“Wij zijn op ons gemak in de strafste ammoniak”

(uit Studentenlied Chemika Leuven, Hugo Van Looy, 1948)

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Source-oriented techniques for improving indoor air quality and assessment of air emissions from pig husbandry

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Dutch translation of the title:

Brongerichte technieken voor de verbetering van de binnenluchtkwaliteit en de bepaling van luchtemissies uit de varkenshouderij

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VOORWOORD – PREFACE

Vaak wordt er gevraagd “hoe gaat het met jouw doctoraat?” en wordt er dan als antwoord gegeven “er is nog wel wat werk aan mijn doctoraat”. In mijn ogen is een doctoraat echter niet het werk van één persoon, maar eerder het werk dat ontstaan is dankzij de (directe of indirecte) hulp en steun van heel wat mensen. Ik zou deze mensen hier dan ook graag voor willen bedanken.

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Tim

SAMENVATTING

De afgelopen decennia vond er een intensivering van de varkenshouderij plaats. Dit heeft geleid tot lokaal verhoogde polluentemissies en daarbij horende nadelige gevolgen voor het milieu en de menselijke gezondheid. Reductietechnieken zijn nodig om deze nadelige gevolgen te verminderen. Idealiter zouden deze reductietechnieken zoveel mogelijk polluenten tezelfdertijd moeten reduceren. Daarnaast zouden betaalbare (brongerichte) reductietechnieken in de stal moeten verkozen worden boven end-of-pipe technieken omdat deze laatste de binnenluchtkwaliteit niet verbeteren. Momenteel worden meestal langdurige en dure metingen ingezet om het potentieel van reductietechnieken in te schatten en om emissiefactoren (EF) te bepalen, die nodig zijn om een wettelijke erkenning te verkrijgen als emissiearme techniek. Om innovatie te stimuleren, zijn goedkopere evaluatiemethodes aangewezen.

In dit proefschrift werden de volgende hoofdaspecten onderzocht:

(1) de invloed van brongerichte technieken op meerdere polluenten in varkensstallen, gebruik makende van een multi-polluent onderzoeksaanpak.

(2) de correlaties tussen de verschillende polluenten in varkensstallen.

(3) de beoordeling van verkorte procedures voor het meten van ammoniak (NH_3) emissies uit varkensstallen om de innovatie van reductietechnieken te stimuleren.

De belangrijkste polluenten in de varkenshouderij zijn NH_3, de broeikasgassen methaan (CH_4) en distikstofoxide (N_2O) en fijn stof (PM: PM_{10}, $PM_{2.5}$ en PM_1). Het belang van deze polluenten in de Vlaamse emissieproblematiek, hun invloed op de omgeving en de menselijke gezondheid, evenals hun bronnen binnenin veestallen wordt besproken in **hoofdstuk 1**. Dit hoofdstuk geeft ook een overzicht van de belangrijkste wetgeving aangaande polluentemissies en reductiedoelen. Zowel reductietechnieken in de stal als end-of-pipe technieken worden kort besproken, evenals technieken en strategieën om polluentemissies te meten.

Hoofdstuk 2 richt zich op brongerichte technieken en de correlaties tussen de binnenluchtconcentraties van de verschillende polluenten in varkensstallen.

In **hoofdstuk 2.1** werd het effect nagegaan van twee reinigingsprotocols en twee stalsystemen op de binnenluchtconcentraties van de polluenten. In deze studie werden er geen significante verschillen in binnenluchtconcentraties van NH_3, CO_2, CH_4, N_2O of PM gevonden over een volledige mestronde wanneer de hokken enkel gekuist werden met bezems en stofzuigers of met bezems en stofzuigers, gevolgd door een inweekstap met water en het grondig kuisen met een hogedrukreiniger. Dit was een teken dat het toepassen van een droge reiniging mogelijk al voldoende was om de polluentconcentraties te verminderen. De binnenluchtconcentraties aan N_2O en PM_1 waren significant lager wanneer de droge reiniging werd toegepast en enkel de eerste maand na reiniging in beschouwing werd genomen. Het was niet geheel duidelijk of het verschil in N_2O binnenluchtconcentraties echt het gevolg was van het gebruik van verschillende reinigingsprotocols of eerder het gevolg was van het ontbreken van data tijdens bepaalde periodes in de eerste maand. Er werd verondersteld dat meer dan de helft van de PM_1 binnenluchtconcentraties afkomstig was van buitenaf. Dit kan, in combinatie met het vier weken verschil in startdatum tussen de rondes met droge en natte reiniging, deels de onverwacht lagere PM_1 binnenluchtconcentraties verklaren bij toepassing van het droge reinigingsprotocol. In vergelijking met een conventioneel stalsysteem resulteerde het gebruik van een wettelijk erkend ammoniakemissiearm (LAE) stalsysteem, gebaseerd op partiële roostervloeren en schuine putwanden, enkel in verlaagde CH_4 binnenluchtconcentraties wanneer de volledige mestronde in beschouwing werd genomen. Er werd geen effect op de andere binnenluchtconcentraties waargenomen. De lagere CH_4 binnenluchtconcentraties in het LAE stalsysteem kunnen het resultaat zijn van een versnelde afvoer van mest via een overloop. Het was evenwel opmerkelijk dat geen significante verschillen in NH_3 concentratie werd waargenomen tussen het LAE stalsysteem en het conventionele stalsysteem. De hogere vervuilingsgraad van de volle vloer in het LAE stalsysteem speelde hierin mogelijk een rol.

De uitgebreide dataset, verkregen in de bovenstaande studie, werd vervolgens gebruikt om correlaties tussen de verschillende gassen en PM fracties te bepalen en om de deeltjesgrootteverdeling (PSD) van het fijn stof te beschrijven (**hoofdstuk 2.2**). Er werden hoge correlaties gevonden tussen de binnenluchtconcentraties van NH_3, CO_2 en CH_4. De

correlatie van deze gassen met de N_2O binnenluchtconcentratie was lager. De hoge correlaties tussen NH_3, CO_2 en CH_4 kunnen mogelijk het gevolg zijn van de gelijkaardige manier waarop deze gasconcentraties toenemen wanneer de dieren groeien, al kan een invloed van het ventilatiepatroon op de gasconcentraties niet uitgesloten worden. Het ongeveer constant blijven van de N_2O binnenluchtconcentraties gedurende de mestrondes is mogelijk de uitleg voor de lagere correlaties tussen NH_3, CO_2 en CH_4 aan de ene kant en N_2O aan de andere kant. Zeer hoge correlaties werden gevonden tussen de PM_{10} en $PM_{2.5}$ binnenluchtconcentraties. Dit kan deels verklaard worden door het feit dat $PM_{2.5}$ een substantieel deel is van PM_{10}. Afhankelijk van het meetinstrument werden er hoge (Grimm spectrometers) of lage (GrayWolf Particle Counters) correlaties gevonden tussen de PM_{10} en $PM_{2.5}$ binnenluchtconcentraties aan de ene kant en de PM_1 binnenluchtconcentraties aan de andere kant. In het algemeen werden er lage correlaties gevonden tussen de verschillende gassen en verschillende stoffracties. Dit duidt op een verschillend gedrag van gassen en fijn stof in de lucht.

Het was opvallend dat er geen grote verschillen in PSD van fijn stof gevonden werden tussen de verschillende stalsystemen of wanneer een verschillend reinigingsprotocol werd toegepast. Hoewel de totale massa aan deeltjes (PM concentraties) significant veranderde (gedurende een dag en gedurende een mestperiode), bleef de PSD ongeveer dezelfde.

Het effect van maalgrootte en pelletering van het varkensvoer was een derde brongerichte reductietechniek die werd bestudeerd in dit proefschrift (**hoofdstuk 2.3**). In compartimenten met gepelleteerd voer waren er hogere PM_{10}, $PM_{2.5}$ en PM_1 binnenluchtconcentraties in vergelijking met compartimenten met meelvoer. Gezien pelletering als een tweede maalstap wordt gezien, kan het pelleteerproces tot verdere verfijning van het voer geleid hebben. Dit werd bevestigd door middel van een natte zeefanalyse. Verder kan het afbrokkelen van de pellets in de voederbakken er toe geleid hebben dat een fijn poeder achterbleef in deze voederbakken. Extra testen in het labo werden uitgevoerd om aanvullende verklaringen te vinden voor deze onverwachte bevindingen. De resultaten van de droptest gaven aan dat meelvoer hogere PM_{10} concentraties veroorzaakte in vergelijking met gepelleteerd voer. Kleine of geen verschillen in $PM_{2.5}$ en PM_1 concentraties werden gevonden. De droptest moet echter eerder gezien worden als een goede maatstaf voor het oppervlakkige (“los”) fijn stof, aanwezig op de pellets. Daarom werd er aanvullend een schudtest uitgevoerd, in een

poging om de wrijvingen tussen de verschillende pellets in het voeder te simuleren. De resultaten van deze schudtest toonden aan dat hogere PM_{10} concentraties bekomen werden met het gepelleteerd voeder in vergelijking met het meelvoeder. Opnieuw werden er kleine of geen verschillen in $PM_{2.5}$ en PM_1 concentraties gevonden. Het fijn malen van de voeders leidde tot hogere PM_{10} concentraties in vergelijking met het grof malen van de voeders. Dit effect was evenwel kleiner dan het effect van het pelleteren van de voeders. Dit kan mogelijk te wijten zijn aan het feit dat niet alle ingrediënten gemalen werden in de voeders omdat sommige ingrediënten reeds vooraf zeer fijn waren of in een vloeibare vorm toegevoegd werden. Het verschil in PM binnenluchtconcentraties tussen beide gepelleteerde voeders kan ook veroorzaakt zijn door een verschil in hardheid van de pellets: pellets van het grof gemalen voer waren harder dan pellets van het fijn gemalen voer.

Bij biggen die meelvoeders kregen, werd een hogere gemiddelde dagelijkse voederopname en een slechtere voederconversie waargenomen ten opzichte van biggen die gepelleteerde voeders kregen. Er werd geen effect van de maalgrootte op de voederconversie gevonden. Biggen die het fijn gemalen meelvoer kregen, hadden een lagere gemiddelde dagelijkse groei, en bijgevolg een lager lichaamsgewicht op 9 weken, dan de biggen die één van drie anders voeders kregen.

In **hoofdstuk 3** werden er een aantal verkorte meetstrategieën onderzocht om een NH_3 EF te schatten voor varkensstallen. Gebaseerd op de uitgevoerde simulaties kon er besloten worden dat het mogelijk was om een EF te schatten met een relatieve fout van maximum 15 % door gebruik te maken van verkorte meetstrategieën. Afhankelijk van de gebruikte dataset waren er hiervoor 21 tot 27 24-uursperioden, 20 tot 29 48-uursperioden, 13 tot 15 7-dagen perioden of 27 tot 84 grab samples nodig. Een geschatte EF met een relatieve fout van maximum 15 % kon ook bekomen worden door wekelijks een grab sample te nemen op een willekeurige werkdag gedurende 28 tot 32 weken.

Een overzicht van de technische problemen die men kan ondervinden tijdens gas- en fijn stofmetingen in het algemeen en in dit proefschrift in het bijzonder, is gegeven in **hoofdstuk 4**.

In de algemene discussie (**hoofdstuk 5**) wordt de doeltreffendheid van brongerichte technieken en hun toekomst als reductietechnieken geëvalueerd. Verder wordt ook het

economisch voordeel van het gebruik van gepelleteerde voeders becijferd. De winst, zowel in tijd als in geld, voor de meest belovende verkorte meetstrategieën werd ook berekend. Aan het einde van dit hoofdstuk worden er een aantal suggesties voor verder onderzoek voorgesteld.

SUMMARY

In the last decades, livestock farming activities, including pig rearing activities, were intensified. This has led to locally increased pollutant emissions and associated adverse effects on the environment and the human health. Mitigation techniques are needed to decrease these adverse effects. Ideally, these mitigation techniques should reduce as many pollutants as possible at the same time. Additionally, affordable (source-oriented) mitigation techniques inside the barn should be chosen over end-of-pipe techniques, since the latter do not improve the indoor air quality. At the moment, long and expensive measurements are needed to assess the potential of mitigation techniques and to determine an emission factor (EF), needed for legal recognition. To stimulate innovation, cheaper evaluation techniques are needed.

In this dissertation the following main aspects were investigated:

(1) the influence of source-oriented techniques on several pollutants in piggeries, using a multi-pollutant research approach.

(2) the correlations between the different pollutants inside piggeries.

(3) the assessment of reduced sampling strategies for ammonia (NH_3) emissions from piggeries in order to stimulate innovation in mitigation techniques.

The main pollutants in pig husbandry are NH_3, the greenhouse gases methane (CH_4) and nitrous oxide (N_2O) and particulate matter (PM: PM_{10}, $PM_{2.5}$ and PM_1). The importance of these pollutants in the Flemish emissions, their influence on the environment and the human health and their sources inside livestock buildings are discussed in **chapter 1**. This chapter also gives an overview of the current legislations regarding pollutant emissions and reduction goals. Both mitigation techniques inside the barn and end-of-pipe techniques are briefly discussed as well as the measuring techniques and strategies to measure pollutant emissions.

Chapter 2 focussed on source-oriented techniques and correlations between the indoor concentrations of the different pollutants inside piggeries.

In **chapter 2.1**, the effect of two cleaning protocols and two housing systems on indoor pollutant concentrations was evaluated. In the present study, no significant differences in indoor concentrations of NH_3, CO_2, CH_4, N_2O or PM over the whole fattening period were found by cleaning the pens between fattening periods with either only brooms and vacuum cleaners or with brooms and vacuum cleaners, followed by a soaking step with water and a thoroughly cleaning with a pressure washer. This was an indication that the application of dry cleaning might already be sufficient to reduce the pollutant concentrations. When only the first month after cleaning was taken into account, N_2O and PM_1 indoor concentrations were significantly reduced when performing the dry protocol. It was not clear if the difference in N_2O indoor concentrations was really a consequence of the cleaning protocols or partially due to the lack of data at some points during the first month. It was hypothesised that the PM_1 indoor concentrations originated for more than 50 % from the outside. This could partially explain, in combination with the four week differences in start-up time between fattening periods with dry or wet cleaning, the unexpected lower PM_1 concentrations for the dry protocol. The use of an officially approved low-ammonia-emission (LAE) housing system, based on partly slatted floors and sloped pit walls, only resulted in reduced indoor CH_4 concentrations as compared to a conventional housing system with fully slatted floors and taking into account the entire fattening period. No effect on the other pollutants could be observed. The lower indoor CH_4 concentrations in the LAE housing system may have been the result of a faster removal of manure via an overflow. It was surprising that no significant differences in indoor NH_3 concentrations were found between the LAE housing system and the conventional housing system. The higher degree of fouling of the solid floor in the LAE housing system might have interfered.

The comprehensive dataset, obtained in the study described above, was subsequently used to determine correlations between the gases and different PM fraction and to describe the particle size distribution (PSD) of PM, as described in **chapter 2.2**. High correlations were found between the indoor concentrations of NH_3, CO_2 and CH_4. The correlations of these gases with indoor concentrations of N_2O were lower. The high correlations between NH_3, CO_2 and CH_4 might reflect the similar effect that animal growth has on these respective gas

concentrations, although an effect of the ventilation rate on the gas concentrations cannot be excluded. Since N_2O indoor concentrations remained more constant during the fattening periods, this could have been the explanation for the low correlations of NH_3, CO_2 and CH_4 with N_2O. Very high correlations were found between the indoor concentrations of PM_{10} and $PM_{2.5}$. The fact that $PM_{2.5}$ is a substantial part of PM_{10} can partially explain this finding. Depending on the measuring instrument, high (Grimm spectrometers) or low (GrayWolf Particle Counters) correlations between the indoor concentrations of PM_{10} and $PM_{2.5}$ on the one hand and the indoor concentrations of PM_1 on the other hand, were found. Generally, low correlations were found between the gases and different PM fractions, suggesting a different behaviour of gases and particles in the air.

It was striking to see that no great differences in PSD were found when comparing PM from the different housing systems and when different cleaning protocols were applied, nor for the count median diameters (CMD), nor for the mass median diameters (MMD). Furthermore, although the total mass of particles (PM concentrations) changed significantly (during a day and during a fattening period), the CMD and MMD values remained about the same.

The effect of grinding intensity and pelleting of pig diets was a third source-oriented mitigation technique that was studied (**chapter 2.3**). Compartments with pelleted diets had higher PM_{10}, $PM_{2.5}$ and PM_1 indoor concentrations as compared to compartments with meal diets. Since pelleting is considered to be a secondary grinding step, the pelleting process may have led to smaller particles. This was confirmed by wet sieve analysis. The degradation of the pellets in the self-feeders, resulting in a fine powder at the bottom of those self-feeders, might also be part of the explanation. Additional laboratory tests were performed in order to find supplementary explanations for these, rather unexpected, findings. Results from the drop test indicated that meal diets gave rise to higher PM_{10} concentrations compared to the pelleted diets and small or no effects on $PM_{2.5}$ or PM_1 concentrations were found. The drop tests were rather a good measure of the superficial (“loose”) PM, present on the pellets. Therefore, in an attempt to simulate the frictions between the different particles in the feed, a shake test was performed. The results indicated that shaking the feeds led to higher PM_{10} concentrations for the pelleted diets as compared to meal diets. Again, small or no effects on $PM_{2.5}$ or PM_1 concentrations were found. Finely grinding of the diets led to higher PM_{10}

indoor concentrations as compared to coarsely grinding of the diets, although this effect seemed smaller than for pelleting the feed. This smaller effect may be due to the fact that not all ingredients were ground before mixing since they were already very fine or in a liquid form. The difference between the indoor PM concentrations for both pelleted diets may also have been caused by the hardness of the pelleted diets: coarsely ground pelleted diets had a higher hardness than finely ground pelleted diets.

Pigs fed meal diets had a higher average daily feed intake (ADFI) and a lower feed efficiency than pigs fed pelleted diets. No effect of grinding intensity on feed efficiency was found. Pigs receiving the finely ground meal diet had a lower average daily gain (ADG) and, as a consequence, a lower bodyweight at 9 weeks of age than pigs receiving coarsely ground meal and finely or coarsely ground pelleted diets.

In **chapter 3**, a number of reduced sampling strategies were assessed to estimate NH_3 EFs of pig fattening facilities. Based on the performed simulations, it was concluded that an estimated EF with a relative error below 15 % could be obtained via reduced sampling strategies, instead of measuring continuously for two consecutive fattening periods. Depending on the tested dataset, 21 to 27 24 hour periods, 20 to 29 48 hour periods, 12 to 16 7 day periods or 27 to 84 single grab samples were needed. An EF with a relative error below 15 % was also obtained by taking weekly grab samples on a random working day for 28 to 32 consecutive weeks.

An overview of the technical problems that can be encountered during gas and PM measurements in general and in this dissertation especially is given in **chapter 4**.

In the general discussion (**chapter 5**), the effectiveness of source-oriented techniques and their future as mitigation techniques are evaluated. The economic advantage of feeding pellets was also evaluated. For the most promising reduced sampling strategies, the gain in measurement costs and measurement time are calculated. At the end of the chapter, directions for future research are suggested.

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LIST OF ABBREVIATIONS

Abbreviation	Description
a	number of animals
ADFI	average daily feed intake
ADG	average daily gain
AED	aerodynamic diameter
BAT	best available techniques
BREF	best available techniques reference document
BW	body weight
CER	CO_2 exhalation rate
CGSD	Geometric standard deviation of the count median diameter
CH_4	methane
C_i	concentration at the air inlet
CMD	count median diameter
C_o	concentration at the air outlet
CO_2	carbon dioxide
CP	crude protein
dEB	dietary electrolyte balance
DOAS	differential optical absorption spectroscopy
EF	emission factor
ER	emission rate
EtOH	ethanol
FTIR	Fourier transform-infrared spectroscopy
G:F	gain over feed
GC	gas chromatography
GHGs	greenhouse gases
GSD	geometric standard deviation
GWP	global warming potential

Abbreviation	Description
H_2S	hydrogen sulphide
ILVO	institute for agricultural and fisheries research
IPPC	integrated pollution prevention and control
IPS	impaction plate pre-separator
IR	infrared
IU	International unit
LAE	low-ammonia-emission
LAS	laser absorption spectroscopy
LSM	least square mean
MeOH	methanol
MGSD	geometric standard deviation of the mass median diameter
MMD	mass median diameter
N_2	molecular nitrogen
N_2O	nitrous oxide
NECs	national emission ceilings
NH_3	ammonia
NH_4^+	ammonium
NMVOCs	non-methane volatile organic compounds
NO	nitric oxide
NO_2	nitrogen dioxide
NO_2^-	nitrite
NO_3^-	nitrate
P	air pressure
PAS	photoacoustic system
PM	particulate matter
P_{ref}	reference air pressure
PSD	particle size distribution
PTFE	polytetrafluoroethylene
Q	ventilation rate

Abbreviation	Description
SACs	special areas of conservation
SID	standardised ileal digestible
SPAs	special protection areas
STEL	short term exposure limit
T	indoor temperature
TCER	tranquil CO_2 exhalation rate
TiO_2	titanium dioxide
T_{ref}	reference temperature
TWA	time-weighted average
UNECE	United Nations economic commission for Europe
VFA	volatile fatty acids
V_m	molar volume
VOCs	volatile organic compounds
VP	vacancy correction factor
w_m	molar mass

1 GENERAL INTRODUCTION

1.1 PIG HUSBANDRY IN FLANDERS

Pig husbandry is an important economic sector in Flanders and represented in 2011 43 % of the total production value from livestock industry and 27 % of the total production value from agriculture and horticulture in Flanders. In Flanders, of all agricultural exploitations in 2011, about 11 % (2959 pig farms) were specialised in pig production. However, the total number of farms which had pigs was even larger (4928 farms in 2011) and comprised 19 % of the total agricultural exploitations. Since 2008, after a period of serious decline, pig production increased steadily in Flanders. In 2011, there were 3.3 % more pigs in Flanders compared to 2005. However, the number of pig farms decreased with 26 % from 2005 to 2011. As a consequence, the average number of pigs per pig farm increased significantly (from 773 in 2001 to 1248 in 2011) (Platteau *et al.*, 2012). This intensification of livestock farming has an impact on the environment due to the emissions of different aerial pollutants (Aneja *et al.*, 2009; Steinfeld *et al.*, 2006). In an attempt to reduce the emissions and adverse effects, policies were developed (e.g. implementation of low-ammonia-emission (LAE) housing systems). However, it has become difficult, especially in a densely populated area like Flanders with locally concentrated livestock farms, for livestock farmers to comply with all regulations (e.g. odour complaints due to proximity of farms and residents). The new regulations regarding conservation objectives for Natura 2000 sites, which will soon come into force in Flanders, will complicate livestock farming in some critical areas. It can be expected that the conflicts between agriculture, nature and nearby inhabitants will only become more pressing in the future. Therefore the challenge to envision pathways towards a sustainable agriculture in Flanders must be taken up urgently. The challenge is to balance environmental and economic requirements and needs.

1.2 MAIN AERIAL POLLUTANTS IN PIG HUSBANDRY

A wide variety of aerial compounds and pollutants exists inside livestock barns. Most of the pollutants are gaseous and include ammonia (NH_3) and the greenhouse gases (GHGs): carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O). Other pollutants consist of a mixture of many types of particle-like pollutants and are called particulate matter (PM). A number of gaseous pollutants (e.g. NH_3, hydrogen sulphide (H_2S) and volatile organic compounds (VOCs)) together with other aerial compounds can cause odour nuisance (NRC, 2003).

In this section, the importance of these different pollutants in pig production will be discussed, together with their formation processes and their possible effects on the environment or human health.

1.2.1 AMMONIA

Agricultural activities are the main source of NH_3 emissions. In 2011, 92 % of the total NH_3 emissions in Flanders originated from agriculture. The largest contribution (83 %) to the NH_3 emissions comes from manure, either directly from the barn or indirectly after land application of manure (VMM, 2012).

Deposition of NH_3 into the environment can lead to eutrophication of waterways and acidification of soils which, in turn, lead to loss of biodiversity and damage to ecosystems (Fangmeier *et al.*, 1994; Krupa, 2003). Emission of NH_3 also has an effect on the formation of secondary PM formation through reactions with sulphuric and nitric acids. This leads to the formation of the ammonium salts ammonium sulphate and nitrate (Erisman & Schaap, 2004). The detrimental effects of PM will be discussed further.

NH_3 is an irritating gas which can have an adverse effect on the human health. The effects on the respiratory system include irritation of the skin, mucous membranes, eyes and the upper respiratory tract (Cole *et al.*, 2000). Clinical signs include coughing, sneezing and salivation (Donham, 2000). Commission Directive 2000/39/EC stated a first list of indicative occupational exposure limits to protect the health and safety of workers from the risks related to chemical agents at work. In this directive, the European Commission recommended a time-weighted average (TWA) limit over an eight hour period of 20 ppmv NH_3 and a short term exposure limit (STEL) over a period of 15 minutes of 50 ppmv NH_3. This

directive was also converted into the Belgian Royal Decree of October 11th, 2002. Up till present it must be noted that no systematic surveys are carried out to check the real conditions inside livestock buildings with regard to these safety levels. Furthermore, research on exposure limits for workers in pig fattening facilities estimated TWA limits of 7 ppmv on the basis of dose-response correlations for human health problems (Donham, 1991).

In pig husbandry, nitrogen is excreted by the pigs either in faeces in the form of proteins or in urine, mainly as urea (Canh *et al.*, 1997). However, a smaller amount of nitrogen, less than 15 % of the total nitrogen excretion, is excreted in urine as creatinine (Figueroa *et al.*, 2002). Hydrolysis of urea is catalysed by the enzyme urease leading to the production of carbonic acid and ammonium (NH_4^+), which in liquid phase is in a pH- and temperature dependent equilibrium with NH_3 (Cortus *et al.*, 2008; Mobley & Hausinger, 1989). The enzyme urease is only present in faeces and is produced by a variety of microbial organisms. Volatilisation can only occur as NH_3 and depends on the concentrations of NH_3 in the manure, the air velocity at the manure surface, the manure pH and temperature and the surface area (Ni, 1999). This enzymatic hydrolysis of urea is the main source of NH_3 from manure. A slower, and less important form of NH_3 formation, is the breakdown of undigested proteins in faeces (Zeeman, 1991).

1.2.2 GREENHOUSE GASES

An important group of aerial pollutants are the GHGs which can contribute to global warming. The most important GHGs in the atmosphere are CO_2, CH_4 and N_2O. However, the net CO_2 emission from livestock, with the exception of CO_2 emission from the use of fossil fuels, is assumed to be zero since the CO_2 originating from the exhalation of the animals is used by plants for their photosynthesis (IPCC, 2006). Therefore, GHGs from livestock buildings mainly consist of CH_4 and N_2O. However for the sake of completeness, CO_2 will also be shortly discussed here.

In 2011, agriculture was responsible for 6 % of the total greenhouse gas emissions in Flanders. The contribution of agriculture to the total CH_4 emission was 79.3 %, with 77 % originating from livestock farming, 2 % from farmland and 0.3 % from fuel consumption. Furthermore, agriculture contributed for 56 % to the total N_2O emission in 2011 in Flanders.

Since only the CO_2 emission from the use of fossil fuels is taken into account, the contribution of agriculture to the total CO_2 emissions is almost negligible (3 %) (VMM, 2012).

CH_4 and N_2O (and CO_2) belong to the GHGs which contribute to the global warming through the greenhouse effect. The greenhouse effect may be simplistically represented as follows: the Earth-atmosphere system absorbs (short-wave) radiation from the sun and re-radiates this energy by means of (long-wave) infrared radiation, maintaining a global energy balance. However, certain gases (called GHGs) and particles in the atmosphere allow (short-wave) sunlight to filter through to the surface, but adsorb the outgoing (long-wave) infrared radiation and re-radiate it, both into space and into the lower atmosphere. Therefore, part of the infrared radiation that would be emitted into space is "trapped" near the surface. The result is a net absorption of energy from short-radiation and heating of the Earth-atmosphere system. When the concentrations of GHGs in the atmosphere increase, the amount of infrared radiation (and thus, heat) that is trapped in the lower atmosphere will increase, thereby increasing the planet's surface temperature (Bowman, 1990; Raval & Ramanathan, 1989; Schneider, 1989). The contribution of a GHG to the global warming is generally expressed as the global warming potential (GWP) and is compared to the GWP of CO_2, which is 1 by definition. On a time horizon of 100 years, CH_4 and N_2O have a GWP of 34 and 298, respectively (IPCC, 2013). Besides its role in the greenhouse effect, N_2O also contributes to the depletion of stratospheric ozone layer (Portmann *et al.*, 2012).

CH_4 is a non-toxic gas, but can become explosive at higher concentrations, with a lower explosion limit of 50 000 ppmv, which is much higher than concentrations normally found within livestock buildings. CH_4 can be classified as an asphyxiant gas, displacing oxygen in the air, which could lead to suffocation. N_2O has a long history as anaesthetic and is only considered toxic when exposed to high concentrations for a long time (Weimann, 2003). However, N_2O concentrations inside livestock buildings rarely exceed a few ppmv. Although CO_2 can be considered a non-toxic gas, it can also be classified as an asphyxiant gas. Donham *et al.* (1991) proposed a TWA limit of 1540 ppmv CO_2 inside livestock barns.

There are two sources of CH_4 production in pig husbandry: (1) enteric fermentation in the animal and (2) release from manure, both driven by anaerobic degradation of organic matter. The CH_4 emission from livestock farming in Flanders can accordingly be split up

(Figure 1.1) into enteric fermentation (60 %) and release from manure (40 %) (VMM, 2012). Emission of CH_4 by pigs through enteric fermentation is estimated to be 4.1 g CH_4 per day (IPCC, 2006), although it is influenced by the diet: enteric fermentation in pigs, and thus CH_4 production, increases with increased dietary fibre content (Jensen & Jørgensen, 1994). Pig husbandry in Flanders is associated with 11 % of the total CH_4 emission from enteric fermentation. Although the CH_4 emission for enteric fermentation from one pig is low, the total CH_4 emission cannot be neglected due to the high number of pigs in Flanders (VMM, 2012). CH_4 is mainly produced by anaerobic digestion of manure via hydrolysis of (hemi-) cellulose, acidogenesis, acetogenesis and methanogenesis. Temperature, pH, (un)availability of oxygen and the presence of inhibiting compounds have an important effect on the CH_4 production from manure (Monteny *et al.*, 2001; Zeeman, 1991). The emission of CH_4 from pig manure is estimated at 32.9 g per day (IPCC, 2006) and pig husbandry is responsible for 84 % of the total CH_4 emission from manure in Flanders (VMM, 2012).

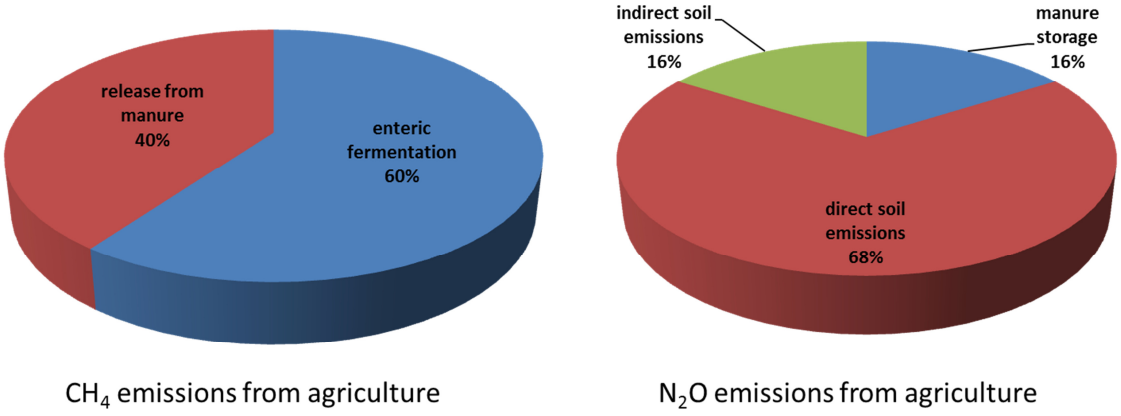

Figure 1.1. Overview of the different sources of CH_4 and N_2O emission from livestock farming in Flanders (VMM, 2012).

N_2O production in manure is a complex process of which the details and conditions are poorly understood. First, urea is converted into ammonium through an ammonification process. Then, under aerobic conditions, ammonium is converted into nitrites (NO_2^-, nitritation) and further into nitrates (NO_3^-, nitratation) by nitrifying bacteria (nitrification process). Finally, nitrate is converted into molecular nitrogen (N_2) through a series of intermediate nitrogen oxide compounds at low oxygen concentrations (denitrification process), with N_2O as intermediate compound (Kebreab *et al.*, 2006; Monteny *et al.*, 2001). In these series of processes, nitrification is the rate-limiting step (Ndegwa *et al.*, 2008). Within the agricultural N_2O emission in Flanders in 2011 (Figure 1.1), only 16 % originated

from manure storage. Other sources of N_2O emission were direct (68 %) and indirect emissions (16 %) from agricultural soils. Direct emissions are the consequence of agricultural activities (e.g. use of fertilizer and manure) which add nitrogen to the soil, increasing the amount of nitrogen which becomes available for nitrification and denitrification processes. Indirect emissions involve nitrogen that is removed from agricultural soils via volatilisation, leaching or runoff into streams and rivers (VMM, 2012). While direct emissions originate from agricultural fields, indirect emissions occur in other locations (e.g. ground and surface waters) than the original nitrogen application site (Mosier *et al.*, 1998).

Carbon dioxide in pig husbandry originates mainly from exhalation by the animals. The CO_2 exhalation rate is influenced by the temperature and animal activity (Ni *et al.*, 1999a). Pigs from 32 to 105 kg produce on average between 41.5 and 73.9 g CO_2 per hour by respiration (Ni *et al.*, 1999a). A second source of CO_2 inside pig buildings is the manure.

Although one research group has reported that the mean release of CO_2 from manure reached an average of 37.5% of the tranquil CO_2 exhalation rate (TCER), which was close to the daily mean CO_2 exhalation rate (CER) (Ni *et al.*, 1999a; Ni *et al.*, 1999b), other researchers have shown that the quantity of CO_2 that is released from manure is less than 5 % of the CO_2 formed by the exhalation of the animals (Anderson *et al.*, 1987; van Ouwerkerk & Aarnink, 1992). More recent research suggested that the release of CO_2 from manure is around 10 % of the CO_2 formed by the exhalation of the animals, provided that manure pits are emptied regularly in a four weeks interval (Pedersen *et al.*, 2008). There are three possible ways of CO_2 formation in manure. The first one is the hydrolysis of urea, already mentioned above, which leads to carbonic acid that decomposes easily into water and CO_2. Anaerobic digestion of organic components in manure is considered as a second CO_2 source (Ni *et al.*, 1999b). Third, CO_2 can be formed from the aerobic degradation of organic matter (Wolter *et al.*, 2004). Manure temperature and the total pig weight have an important influence on the CO_2 release from the manure pit (Ni *et al.*, 1999b).

1.2.3 PARTICULATE MATTER

In 2011, 21 % of the primary PM_{10} emission in Flanders originated from agriculture. Livestock farming attributed 57 % of these emissions. For primary $PM_{2.5}$, agriculture was responsible for 18 % of the total emission in 2011 in Flanders. 26 % was related to livestock farming, but

the bulk of the emission (62 %) from agriculture was due to transport (mainly from the exhaust of agricultural vehicles) (VMM, 2012). Up till now, the agricultural impact on the PM_1 emissions is unknown.

PM emitted to the environment can have multiple negative effects. PM deposition can have an effect on the competitive viability and reproductive fitness of individual plants (Grantz *et al.*, 2003). It is also connected to acidification and nitrogen saturation, which can have an impact on terrestrial ecosystems (Grantz *et al.*, 2003). Particles can also have a direct effect on global warming through scattering and absorption of solar and infrared radiation. An indirect effect of particles as cloud condensation nuclei, affecting the Earth's climate through cloud formation, changes in the cloud lifetime and precipitation, is also observed (IPCC, 2001).

PM can affect human health in three ways: through inhalation of pathogenic and non-pathogenic micro-organisms, through irritation of the respiratory tract and through reduction of the immune resistance to respiratory diseases (Harry, 1978). In contrast to the other pollutants mentioned above, there is no safe level of exposure to PM and also PM concentrations outside livestock houses can give rise to health issues (Pope *et al.*, 2002). For example, studies have shown a positive correlation between increased $PM_{2.5}$ concentrations and increased illness of people (Pope, 2000; Thurston *et al.*, 1994). These illnesses include mainly respiratory problems, but also heart malfunctions (Davidson *et al.*, 2005). Radon *et al.* (2007) questioned about 7000 German rural residents living in a town with a high density of livestock barns and they medically examined about 800 of these residents. This study revealed a higher rate of wheezing without a cold and a lower lung function for residents living close to a large number of animal houses (more than 12 animal houses within 500m of their home). These asthma-like syndromes are also observed in farmers and farm workers (Radon *et al.*, 2007). The role of endotoxins (on the PM particles) towards asthma is less clear: both increased risks due to endotoxins as well as a protection thanks to endotoxins are reported (Radon, 2006; Schulze *et al.*, 2006; von Mutius & Radon, 2008). In recent years, other epidemiological studies and reviews were published on the relation between air pollution and the human health (near livestock houses)(Davidson *et al.*, 2005; Schinasi *et al.*, 2011; Smit *et al.*, 2013; Valavanidis *et al.*, 2008). However, a more detailed description of the influence of PM on the human health is beyond the scope of this dissertation.

In contrast to the pollutants mentioned above, which are chemically well defined, PM is rather a mixture of particles with different properties, sizes and compositions which originates from different sources (U.S.EPA, 2004). The expression "PM" is mainly used in the context of air quality and covers fine solid or liquid particles suspended in a gaseous medium. The same definition holds for "aerosol", but this expression is more commonly used in atmospheric science (Cambra-López *et al.*, 2010). This heterogeneity results in different particles with differences in shape, size, density and chemical composition. The first three properties are combined into one parameter: the aerodynamic diameter (AED). It describes the behaviour of particles in the atmosphere and can be defined as the diameter of a perfect spherical particle, with a density of 1 g cm^{-3} that has the same terminal velocity when settling under gravity as the particle in question (Kulkarni *et al.*, 2011). The different fractions of PM can be defined in multiple ways (modal classification, occupational health classification, size-selective classification), depending on the research area and topic of interest. In this dissertation, only the size-selective classification will be used. PM_{10}, $PM_{2.5}$ and PM_1 are defined as particles that pass through a size-selective inlet with a 50 % efficiency cut-off at 10 µm AED, 2.5 µm AED or 1 µm AED, respectively (U.S.EPA, 2004).

PM can also be grouped on the basis of their origin or source. Primary PM is emitted directly into the atmosphere and originates mainly from mechanical processes, but can also contain micro-organisms, pollen, spores and toxins. Primary PM is mainly contained into the fraction $PM_{10} - PM_{2.5}$. Secondary PM on the contrary, originates mainly from chemical reactions between gases (ammonia, sulphur dioxide, nitrogen oxide and VOCs) and particles in the atmosphere. Secondary PM is mainly contained in the fraction $PM_{2.5}$. Exceptions to this are particles from combustion processes, which are primary in origin, but fall into the fraction $PM_{2.5}$ (Cambra-López *et al.*, 2010). In the context of livestock farming, the main sources of PM are feed, manure and skin parts, but can also contain hair and bedding material (Aarnink *et al.*, 1999; Curtis *et al.*, 1975; Donham *et al.*, 1986; Heber *et al.*, 1988a). Recently, chemical and morphological characterisation, including electron microscopy, of the fractions $PM_{10} - PM_{2.5}$ and $PM_{2.5}$ has shown that, based on particle number, manure was the most abundant source for pigs (41 to 94 % in the fraction $PM_{10} - PM_{2.5}$, 70 to 98 % in the fraction $PM_{2.5}$). When expressed in particle mass, the most important sources were skin parts (0 to 71 % in the fraction $PM_{10} - PM_{2.5}$ and 0 to 79 % in the fraction $PM_{2.5}$) and manure (23 to 92 %

in the fraction PM_{10} – $PM_{2.5}$ and 14 to 95% in the fraction $PM_{2.5}$)(Cambra-López *et al.*, 2011b).

1.2.4 ODOUR

Odour can be defined as a sensation that occurs when the sensory receptors of the human nasal cavity are stimulated by an odorant, i.e. a chemical element (Schiffman *et al.*, 2001; Ubeda *et al.*, 2013). A study by Schiffman *et al.* (2001) revealed 331 different odour-causing VOCs and gases from swine facilities. These compounds consisted of acids, alcohols, aldehydes, amides, amines, aromatics, esters, ethers, fixed gases, halogenated hydrocarbons, hydrocarbons, ketones, nitriles, other nitrogen-containing compounds, phenols, sulphur-containing compounds, steroids and other compounds (Schiffman *et al.*, 2001). Odours from livestock are generally generated under anaerobic conditions by microbial decomposition of organic matter, mostly in the manure (Ubeda *et al.*, 2013). Odour from livestock buildings and agricultural activities (e.g. spreading of manure) is mostly a concern at a very local level. Although some of these compounds can have health effects (mostly irritation of eyes, nose, throat or skin) or environmental effects (e.g. precursors to the formation of tropospheric ozone), odours associated with agricultural emissions are not regulated in response to health or environmental effects but rather in response to nuisance complaints (NRC, 2003).

Investigating odour (nuisance) from livestock is a discipline on its own, which requires other measuring techniques (e.g. olfactometry) as compared to NH_3, GHGs and PM measurements. In this dissertation, odorous emissions from livestock buildings were not measured and, hence, odour will not be discussed in detail.

1.2.5 VOLATILE ORGANIC COMPOUNDS

Volatile organic compounds (VOCs) are compounds which easily vaporise at room temperature (vapour pressure larger than 10 Pa at ambient conditions). Besides CH_4 (already discussed in section 1.2.2), VOCs also consist of fatty acids, nitrogen heterocyclic compounds, sulphides, amines, alcohols, ethers, mercaptans, hydrocarbons and halocarbons (NRC, 2003). These VOCs are also often called non-methane volatile organic compounds (NMVOCs). NMVOCs originate mainly from undigested proteins that decompose in manure (Mackie *et al.*, 1998). The volatile fatty acids, which represent a large portion of the NMVOCs

are responsible for a significant proportion of odour emissions from pig production facilities (Zahn *et al.*, 2001). NMVOCs can also be classified as GHGs, although their direct effect on the greenhouse effect is negligible. However, they have an indirect effect as precursor to tropospheric ozone (IPCC, 2001).

As for odour, the measuring techniques (gas chromatography and mass spectrometry) for the characterisation and quantification of NMVOCs are different as compared to NH_3, GHGs and PM measurements.

1.3 POLICY AND LEGISLATION

As mentioned in section 1.1, the intensification of livestock farming has negative impacts on air, water and soil and gives rise to concerns regarding emissions of the pollutants, described in section 1.2. Policy makers and governmental agencies became aware of this negative impact (Aneja *et al.*, 2008). This awareness, together with growing public concerns, has led to regulations and mitigation strategies for the different pollutants. Already in the 1970s, protocols were adopted in Europe by the UN Economic Commission for Europe (UNECE) under the Convention on Long Range Transboundary Air Pollution to protect the environment and human health (Rosencranz, 1981). In 1999, the Gothenburg Protocol was adopted by the UNECE to abate acidification, eutrophication and ground-level ozone, setting emission ceilings to be reached by 2010 for four pollutants: sulphur oxide, nitrogen oxide, VOCs and NH_3. This protocol was amended in 2012 to include national emission reduction commitments to be achieved in 2020 and beyond. Upper limits for the same four initial pollutants to be met in 2010 were also set for the Member States of the European Union under Directive 2001/81/EC. The National Emission Ceilings (NECs) in this Directive were slightly stricter than those in the Gothenburg Protocol. Currently, the amendment of this Directive is still under preparation, but should set NECS to be respected by 2020 and by 2030 for the four already regulated pollutants.

In Flanders, in order to reduce NH_3 emissions from livestock buildings, a Ministerial Decree was adopted in 2004. This decree required pig and poultry producers in Flanders to use officially approved LAE housing systems when renovating, expanding or building new animal housing. The last decade, this had led to the development and approval of new LAE housing

systems which were added to this decree. The list of all officially approved LAE housing systems for fattening pigs in Flanders can be found in Appendix A. Furthermore, all European (and hence also all Flemish) pig fattening facilities with more than 2000 fatteners are subjected to the European Integrated Pollution Prevention and Control (IPPC) convention. The Intensive Rearing of Poultry and Pigs BREF (Best Available Techniques (BAT) reference document) gives an overview of the BAT, with good agricultural practice as an essential part of it, to reduce NH_3 emissions (European Commission, 2003). Currently, this document is under revision. A draft version is already available (European Commission, 2013).

Low emission spreading of manure to the fields in a way to reduce emissions is also regulated by decrees in Flanders. Furthermore, in order to lower the amount of nitrate in the soil, additional decrees for manure application techniques and fertilisation standards are in force.

With regard to nature conservation, the Flemish government recently approved 36 specific conservation objectives decrees. These objectives serve to protect European habitats and species in Special Areas of Conservation (SACs), determined in the European Habitats Directive (Council Directive 92/43/EEC of 21 May 1992) and Special Protection Areas (SPAs), determined in the Birds Directive (Council Directive 79/409/EEC of 2 April 1979). The measures, taken by the Flemish government, also imply the establishment of a Programmatic Approach to Nitrogen (PAN/*Programmatorische Aanpak Stikstof* in Dutch). This PAN will have substantial consequences for livestock farms, located in or near SACs and can also result in extra generic measures to reduce NH_3 emissions of livestock farms. Livestock farms that significantly contribute to the exceedance of the critical nitrogen deposition value of a SAC will have to take measures to reduce their NH_3 emission levels. In case a livestock farm contributes more than 50 % to the critical deposition value, it may even be forced to stop its activities (E. Brusselman, personal communication).

A national emission reduction commitment for $PM_{2.5}$ for 2020 was added in the amended Gothenburg Protocol and will also be added to the amended Directive 2001/81/EC. PM concentrations in ambient air are regulated by the 2008/50/EC Directive on ambient air quality and cleaner air for Europe. This directive states that the limit value for the daily PM_{10} average concentration is 50 $\mu g\ m^{-3}$ and should not be exceeded more than 35 times in a

calendar year. A limit value of 40 µg m^{-3} was also set for the yearly PM_{10} average concentration. For $PM_{2.5}$ a yearly limit average value of 25 µg m^{-3} was set.

The emissions of GHGs are regulated by the Kyoto Protocol to the United Nations Framework Convention on Climate Change, adopted in 1997 and came in force in 2005. In this protocol, Belgium committed itself to reduce GHG emissions by about 7.5 % in the first commitment period (2008 – 2012) as compared to 1990. The first commitment period ended in 2012 and the second commitment period has not entered into legal force yet. In this second commitment period (2013 – 2020), Belgium should reduce its GHG emissions by 20 % as compared to 1990. Furthermore, it is also possible that a NEC for CH_4 will be added to the amended Directive 2001/81/EC.

Despite the limit values for PM and the Kyoto Protocol, up till now no regulations about PM or GHG emissions from livestock farming are in force in Belgium or Flanders. However, in agreement with the Kyoto Protocol, agriculture in Belgium (and Flanders) will have to reduce their GHG emission with 15 % by 2020 compared to 2005 (Campens *et al.*, 2010).

1.4 EMISSION MITIGATION TECHNIQUES

Overall, emission mitigation techniques can be grouped into mitigation techniques inside the barn and end-of-pipe techniques. Mitigation techniques inside the barn try tackling the emissions at the source (source-oriented techniques), mainly by preventing the formation or the release of the pollutants, or try lowering the pollutant concentrations inside the barn. This has the considerable advantage that not only the emissions but also the indoor concentrations are positively affected. This is in contrast to end-of-pipe techniques which treat the outgoing air and only reduce the emission of pollutants, but do not prevent their formation or release inside the barn environment. Both groups of techniques will be briefly discussed for NH_3, GHGs and PM in the next section. This overview will be limited to reduction techniques at the level of the barn. The processing of manure outside the barn, air guidance techniques outside the barn or reduction techniques for land application of manure are outside the focus of this thesis. Reported reduction percentages can differ considerably between studies as a consequence of differences in housing system, management, climate or measurement technique.

1.4.1 MITIGATION TECHNIQUES INSIDE THE BARN

Different sources of NH_3, GHGs and PM exist inside a livestock building. These sources include the animals, the diet of the animals, the manure and manure pit and the floor of the pens. Different techniques focus on these sources with the goal of reducing the pollutant emissions. All discussed techniques are summarised at the end of this section (Table 1.1). The reported reduction percentages and costs for the different mitigation techniques were calculated with the same denominator and are summarised in Table 1.2 and Table 1.3

1.4.1.1 DIET

Since the amount of protein in the diet is a key factor determining the amount of excreted nitrogen and the subsequent emission of NH_3, it is very important to adjust the amount of protein in the diets to the needs of the pigs over time. This can be achieved by formulating towards an optimal amino acid profile and by a "phase feeding" strategy where the composition of the diet is matched to the requirements of the animals (Aarnink & Verstegen, 2007; Dourmad *et al.*, 1999; Lenis, 1989). The best results in the reduction of nitrogen excretion, up to 50 %, are reached when phase feeding is combined with a perfect balance of essential amino acids and an optimisation of the supply of non-essential amino acids (Bourdon *et al.*, 1997; Dourmad *et al.*, 1999). Modifying the number of diets to decrease the nitrogen excretion can reduce the cost of the feed. However, the supplementation of amino acids to the feed will increase the cost of the feed. According to Dourmad *et al.* (1995) it is possible to reduce the nitrogen excretion by about 20 % with phase feeding, without significantly increasing the cost of main feed ingredients. Compared with the one phase feeding strategy, other researchers report estimated savings of 1.3 € (two phases) and 2.3 € (three phases) per pig (Edwards *et al.*, 2002). However, the cost effectiveness of dietary mitigation techniques is mainly determined by the price of the main feed ingredients, which can fluctuate over time.

Reducing the nitrogen content in the diet is a possible way of reducing nitrogen excretion and consequently NH_3 emissions (Canh *et al.*, 1998a; Philippe *et al.*, 2006). The reduction of the feed nitrogen content can be accomplished through the reduction of crude protein and addition of supplementary synthetic amino acids. This results in a reduction of NH_3 emissions of about 6 to 13 % for every 10 g kg^{-1} reduction in dietary crude protein (Canh *et al.*, 1998b; Hayes *et al.*, 2004; Hobbs *et al.*, 1996; Velthof *et al.*, 2005). Various reduction percentages

are obtained as a result of the various differences in crude protein level between the control and experimental diet. In some studies, these differences were quite large. For instance, Hayes *et al.* (2004) compared diets with a difference in crude protein level of 90 g kg^{-1} (220 g kg^{-1} for the control diet and 130 g kg^{-1} for the experimental diet) while Canh *et al.* (1998b) used a smaller difference in crude protein level of 40 g kg^{-1} (165 g kg^{-1} for the control diet and 125 g kg^{-1} for the experimental diet). However, the reduction of the crude protein level and the supplementation of synthetic amino acids has its limits because other amino acids, which cannot be supplemented, can become limiting. Therefore, it is suggested that the lysine:crude protein ratio should not exceed 6.5 to 6.8 % (Dourmad *et al.*, 1999). Aarnink *et al.* (2010) estimated that reducing the crude protein level (from 165 g kg^{-1} to 135 g kg^{-1}) would cost approximately 6 € per year and per animal place (Aarnink *et al.*, 2010). Although reducing the crude protein content in a diet reduces nitrogen excretion, at present, no effect on N_2O emissions as a result of decreased dietary protein level was found (Clark *et al.*, 2005; Osada *et al.*, 2011). For CH_4 emissions, both reduced and enhanced emissions or no changes in emissions were found when lowering the crude protein content (Clark *et al.*, 2005; Osada *et al.*, 2011; Philippe *et al.*, 2006; Velthof *et al.*, 2005). Enhanced (+10 %) CO_2 emissions from the manure are reported after reducing dietary crude protein content. This unexpected finding was explained by the authors by the differences in microbial activity in the manure from the control and experimental compartment (Clark *et al.*, 2005).

Reducing the urinary pH and thereby reducing the pH of the manure can consequently reduce NH_3 emissions from pig manure. This reduction in urine pH can be accomplished by the addition of acids or acidifier sources to the diet, resulting in NH_3 emission reduction percentages between 15 and 30 % (Aarnink *et al.*, 2008; Kim *et al.*, 2004). The cost of adding 1 % of benzoic acid to the feed is estimated to be approximately 10 € per year and per animal place (Aarnink *et al.*, 2010). Another way of lowering the urine pH may be obtained by altering the acid-base status in pigs, through the dietary electrolyte balance (dEB), which can be calculated as $Na^+ + K^+ - Cl^-$ and is expressed in mEq (Canh *et al.*, 1997; Canh *et al.*, 1998b). Changing 6 g kg^{-1} $CaCO_3$ to $CaCl_2$ in the diet in order to alter the dEB is estimated to cost approximately 9 € per year and per animal place (Aarnink *et al.*, 2010). Additionally, this reduction in urine pH has also the potential to lower (-14 %) the CH_4 emissions in buildings with weekly pit recharge by reducing the pH in the large intestines of the pigs (Kim *et al.*,

2004). Without this weekly pit recharge, the reduction in CH_4 emissions is not observed (Aarnink *et al.*, 2008). Adding acidifier sources to the feed is not believed to lower N_2O emissions (Aarnink *et al.*, 2008).

The addition of dietary fibres (non-digestible carbohydrates) to the diet can shift the nitrogen excretion from urine (nitrogen mainly present in urea) to faeces by incorporating intestinal nitrogen and urea excreted from the blood into bacterial protein (Bakker *et al.*, 1996; Canh *et al.*, 1997; Low, 1985). As already mentioned before, the breakdown of these proteins, and the production of NH_3 is much slower than the production from urea (Zeeman, 1991). Furthermore, fermentation of the dietary fibres in the hind gut of pigs to volatile fatty acids (VFA) can reduce manure pH (Aarnink & Verstegen, 2007). Increasing the dietary fibre content enhances VFA production (Bakker *et al.*, 1996; Canh *et al.*, 1997). However, increasing the fibre level will increase CH_4 production, both from the animal itself as from the manure because of the higher concentrations of bacterial fermentable substrates (Aarnink & Verstegen, 2007; Velthof *et al.*, 2005). CO_2 emissions from manure were reported to decrease (Clark *et al.*, 2005) or increase (Philippe *et al.*, 2009) with increased fibre levels. No effect on N_2O emissions is reported (Clark *et al.*, 2005).

The addition of antibiotics, probiotics, exogenous enzymes, plant extracts or zeolites to the feed have been suggested as measures against NH_3 emission. However, their effectiveness is not always clear (Ndegwa *et al.*, 2008). Furthermore, little is known about the influence of feed additives on GHG emissions. The extra cost of adding probiotics to the diet can be compensated by the increased feed conversion efficiency (European Commission, 2013). Adding animal fat (2.5 to 5 %) or oil (0.5 to 1 %) to the feed has shown to lower total and respirable indoor dust concentrations (Chiba *et al.*, 1985; Chiba *et al.*, 1987; Clark & McQuitty, 1988; Heber & Martin, 1988; Takai *et al.*, 1996). However, other authors report no effects or even increased respirable PM concentrations after adding oil (2 %) to the feed (Welford *et al.*, 1992).

The form in which the feed is delivered to the animals can also influence NH_3 emissions. Reducing the particle size of the diets or pelleting the diets can potentially reduce nitrogen excretion by increasing the surface area of the ingredient particles. This allows for a better interaction with digestive enzymes, higher nitrogen digestibility, a better feed conversion

ratio and consequently less nitrogen excretion (Ferket *et al.*, 2002; Lahaye *et al.*, 2004; Skoch *et al.*, 1983). Furthermore, feeding pelleted diets instead of meal diets has shown to lower indoor PM concentrations (Bundy & Hazen, 1975; Li *et al.*, 1992; Li *et al.*, 1993; Robertson, 1992; Zeitler *et al.*, 1987). However, varying results were obtained depending on the feed delivery system and the season (Bundy & Hazen, 1975; Zeitler *et al.*, 1987). The extra cost to pellet diets is approximately 7 €/tonne (I. Peeters, Aveve, personal communication). Coating of the pellets with fat or lignin can further decrease PM concentrations (Li *et al.*, 1992). Liquid feeding has also been proposed as an option to reduce PM concentrations. However, in practice no unambiguous results were obtained (Dawson, 1990; Robertson, 1992; Takai & Pedersen, 2000; Zeitler *et al.*, 1987).

1.4.1.2 HOUSING SYSTEMS, TECHNIQUES AND MANAGEMENT PRACTICES

Segregating the urine from the faeces immediately upon excretion is an efficient way of reducing the NH_3 emissions. This segregation prevents contact between the urea present in the urine and the enzyme urease. As a consequence no NH_4^+ is formed (Mobley & Hausinger, 1989). Although this technique gives rise to very high reduction percentages on a laboratory scale, application in livestock buildings has been proven to be less efficient. Still a significant reduction (≈ -50 %) in NH_3 emission can be achieved (Ndegwa *et al.*, 2008). Multiple solutions exist to segregate the urine and faeces fraction (Lachance *et al.*, 2005; Von Bemuth *et al.*, 2005). Segregation of urine and faeces is commonly accomplished by using various floor and pit designs, for example V-shaped pit floors with a scraper (Von Bemuth *et al.*, 2005) or conveyor belt systems (Lachance *et al.*, 2005). Depending on the type of scraper system used, various reductions in NH_3 emissions can be achieved. Flat scraper systems do not seem to lower NH_3 emissions (Kim *et al.*, 2008), possibly because the floor is still soiled with a thin film of faeces and manure after scraping. V-shaped scrapers (Von Bemuth *et al.*, 2005) or belt systems (Lachance *et al.*, 2005; Van Kempen *et al.*, 2003) on the other hand are effective (≈ -50 %) in reducing NH_3 emissions. V-shaped conveyor belt systems can reduce CH_4 emissions, probably because almost all CH_4 emission from manure is eliminated through the separate disposal of faeces and urine (Aarnink *et al.*, 2007a). PM_{10} emissions are not affected by this system while N_2O emissions from this system are very low (Aarnink *et al.*, 2007a), although N_2O emissions from outside storage as solid faeces can be drastically enhanced due to higher (de)nitrification (De Vries *et al.*, 2013). The investment cost for a

housing system with a V-shaped manure belt is believed to be lower than for a fully slatted floor system (calculations with the steel price in 2007). This is mainly caused by the higher share of steel among the materials for construction and is therefore dependent of the steel price (Aarnink *et al.*, 2007a; European Commission, 2013).

Reducing the emitting surface of the manure pit can also reduce NH_3 emissions. A housing system with a manure and water channel, both with sloped pit walls to reduce the emitting surface (Figure 1.2), is on the list of officially approved LAE housing systems in Flanders (Appendix A) and should have an emission factor of maximum 1.2 kg NH_3 $year^{-1}$ (animal place)$^{-1}$. However, Van Ransbeeck *et al.* (2013a) found in practice an emission factor of 1.6 kg NH_3 $year^{-1}$ (animal place)$^{-1}$, while van Zeeland (1997) found an emission factor of 1.0 kg NH_3 $year^{-1}$ (animal place)$^{-1}$ for the same system. In contrast to van Zeeland (1997), who performed almost continuous measurements during two consecutive fattening periods, Van Ransbeeck *et al.* (2013a) measured on average minimum 48 consecutive hours per month during two consecutive fattening periods. The length of the fattening periods were also longer for the measurement by Van Ransbeeck *et al.* (2013a)(round 1/2: 144/129 days) as compared to the fattening periods by Zeeland (1997)(111/109 days). The system with sloped pit walls was also successfully tested in housing systems for weaning piglets (van Zeeland & den Brok, 1998). In Belgium, implementing this technique to an existing barn with fully slatted floors is estimated to cost 344 € per animal place in case the originally fully slatted floor is changed into a partially slatted floor or 168 € per animal place in case the floor remains fully slatted (European Commission, 2013). The extra costs to build a new barn of this type as compared to the costs to build a conventional barn with fully slatted floor and manure pit, vary between 86 and 109 € per animal place (European Commission, 2013).

The emitting surface can also be reduced by making use of partially slatted floors instead of fully slatted floors (Oldenburg, 1989; Sun *et al.*, 2008). However, the influence of partly slatted floors on CH_4 and N_2O emissions is not clear: positive, neutral and negative effects have been reported (Guingand *et al.*, 2010; Laguë *et al.*, 2004). No significant differences between CO_2 emissions from partially or fully slatted floors are found (Guingand *et al.*, 2010; Sun *et al.*, 2008). The costs for building a barn with fully-slatted floors or partially slatted floors are deemed similar (European Commission, 2013). The use of partly slatted floors

instead of solid floors can also reduce PM concentrations, because there is less potential for settled dust to resuspend (Dawson, 1990).

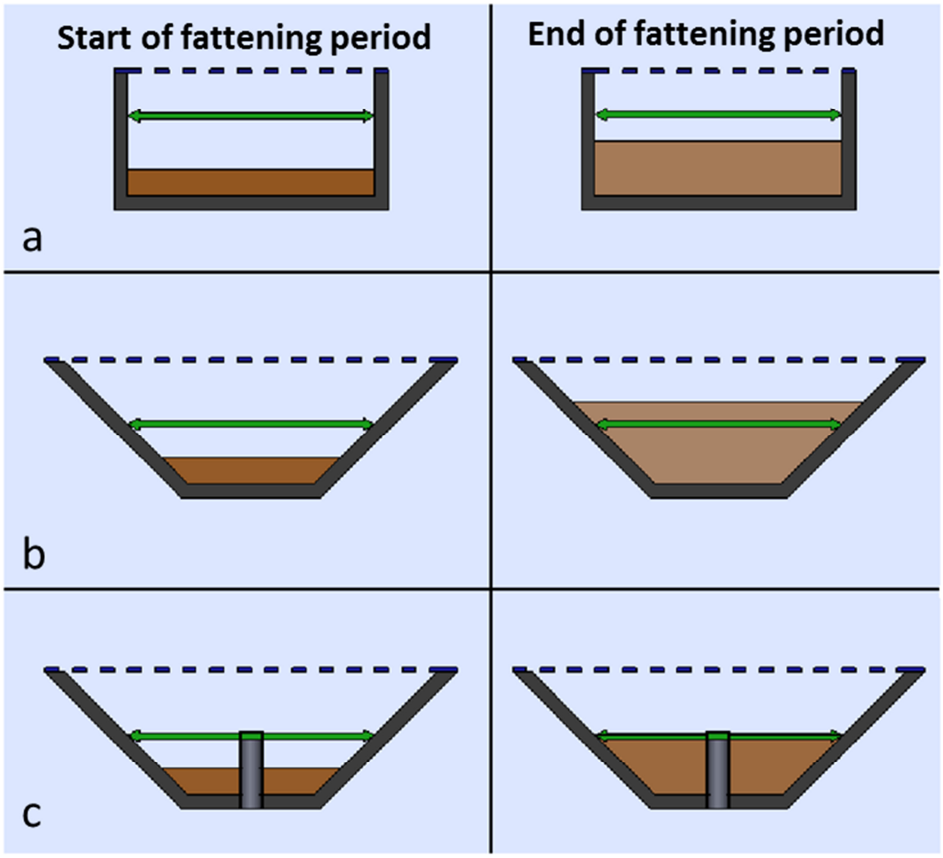

Figure 1.2. Working principle of sloped pit walls. In a standard manure pit (a), the emitting surface area stays maximal throughout the fattening period. By introducing sloped pit walls (b), the emitting surface area is reduced at the start of the fattening period. However, at the end of the fattening period, the emitting surface area could become larger than for a conventional manure pit. This is solved by adding an overflow (c). This figure was adapted from Van Overbeke *et al.* (2010).

Instead of using partially or fully slatted floors with a manure pit, deep litter bedded floor with various bedding materials can also be used. However, a deep litter system with straw is reported to increase NH_3 (+110 %), CO_2 (+14 %) and N_2O (+106 %) emissions in comparison with a fully slatted floor system (Philippe *et al.*, 2007). Though, the NH_3 emission for the fully slatted floor system was rather low in that study. Emissions of N_2O are believed to be higher in the bedded floor system due to the presence of both aerobic and anaerobic conditions in deep litter but not in manure. Higher CO_2 emissions probably originated from higher emissions from the deep litter (Philippe *et al.*, 2007). Changing from a straw based deep

litter system to a straw flow system[i] can reduce GHG emissions (-55 % N_2O, -47 % CO_2 and -46 % CH_4) but further increase NH_3 (+10 %) emissions, possibly due to the daily scraping of the solid manure in the straw flow system (Philippe *et al.*, 2012). However, contrasting results for deep litter systems are reported in other studies. An overview of the emissions of NH_3 for different deep litter systems can be found in Philippe *et al.* (2011). In the United Kingdom, it was estimated that using a straw-based housing system instead of a fully slatted floor system gives rise to an extra cost of approximately 34 € per tonne of produced pig meat or 1.87 € per pig with a final weight of 115 kg (European Commission, 2013). These additional costs are mainly caused by the price of the straw (or other bedding material), but also the extra labour has to be taken into account.

In a preliminary study by Guarino *et al.* (2008) and a follow-up study by Costa *et al.* (2012), the authors claimed that the combination of applying UV lights and titanium dioxide (TiO_2) paint on the compartment walls inside a pig barn, had an effect on CH_4 emissions (15 to 27 % reduction). A reduction (-30 %) in NH_3 emission was also claimed by Guarino *et al.* (2008), but not by Costa *et al.* (2012). However, in both studies, measurement were performed during only one farrowing cycle (Guarino *et al.*, 2008) or one production cycle (Costa *et al.*, 2012). Therefore, the possible effects of the farrowing room or weaning unit on the emissions cannot be assessed. Furthermore, in the study by Guarino *et al.* (2008) lower CO_2 emissions were reported in the TiO_2 treated compartment. No clear explanation was given for this remarkable finding. This may indicate that problems occurred during the gas measurements. Since so far no one has confirmed these results, caution has to be taken when interpreting these results.

Filtration of the indoor air can lower PM concentrations. This can be achieved by using internal recirculating dry air filters (Carpenter & Fryer, 1990), electrostatic precipitators (Rosentrater, 2003) or ionisation systems (Tanaka & Zhang, 1996). The cost to install an ionisation system for a surface of 450 to 600 m² in a fattening pig housing system is

[i] The straw flow system, which is a kind of bedded system, was developed by Bruce (1990). In this system, the lying area is sloped and straw is supplied at the top of this lying area. Through the motion of the pigs, the straw travels down the slope where it is mixed with manure and leaves the pen through a fence onto a scraped passage outside the pen.

estimated to be around 2000 € with an annual operational cost of 8 € per animal place. The installation cost for a system with dry air filters is estimated to be 1.14 € per 30 m^3 of air exchange (European Commission, 2013). Furthermore, large volume housing systems would also require large sizes of these instruments, making them impracticable in livestock buildings (Dawson, 1990).

Ventilation can play an important role in the NH_3, GHG and PM emissions. Higher ventilation rates can increase NH_3 and CO_2 emissions due to the increased air exchange rate above the manure and the reduced resistance of the transfer of NH_3 and CO_2 to the air above the manure (Ndegwa *et al.*, 2008; Ni *et al.*, 1999b). Short periods of increased ventilation ("purge ventilation") can dilute and thus reduce total dust concentrations, however this effect only lasts for a short time (Robertson, 1989). The placement of the air inlet and outlet of the ventilation system can play an important role in reducing NH_3 and PM concentrations. Lower NH_3 concentrations at animal level and lower (-78 %) PM concentrations in the feeding passage are reported when the air inlet is situated near the breathing zones of the pigs and the outlet is situated near the slatted floor and manure pit. However, this alternative placement of the air in- and outlet does not significantly reduce NH_3 emission levels (Aarnink & Wagemans, 1997). Combining ceiling ventilation with pit ventilation has shown to reduce (-42.6 %) indoor NH_3 concentrations by removing NH_3 from the air above the manure before it is transported above the slatted floor. However, due to enhanced air flow rates over the manure, slightly increased (+5 %) NH_3 emissions are reported when using this partial pit ventilation system (Saha *et al.*, 2010). This increase could be overcome by applying an end-of-pipe technique to the pit exhaust.

Ventilation also largely influences the indoor air temperature and local climate inside a pig building. This local climate can influence the dunging and lying locations of the pigs in the pen. Pigs tend to excrete on cold floors or on places were cold air falls into the pen (Aarnink *et al.*, 1996; Hacker *et al.*, 1994; Randall *et al.*, 1983). When indoor temperatures rise, pigs start to excrete on the solid floors, increasing the emission surface (Aarnink *et al.*, 1996; Aarnink *et al.*, 2006; Huynh *et al.*, 2005). Therefore, it is important to establish a correct dunging and lying pattern through the ventilation and air flow pattern (Aarnink & Wagemans, 1997). Reduced pen fouling, independent of ventilation, can be achieved by the

placement of studs on the slatted floors, preventing the pigs from lying there (Aarnink *et al.*, 1997).

Although natural ventilation is considered a BAT regarding electricity use and indoor noise (European Commission, 2003; European Commission, 2013), the effect on the pollutant emissions is less clear. Some authors report lower NH_3 concentrations (Kim *et al.*, 2008) or lower PM_{10} (-63 %), $PM_{2.5}$ (-53 %) and PM_1 (-50 %) concentrations inside naturally ventilated buildings as compared to mechanically ventilated buildings. Others find lower NH_3 EFs when comparing EFs from mechanically and naturally ventilated buildings, found in other studies (Ni *et al.*, 2000). Anyhow, information on pollutant emissions from naturally ventilated pig facilities is still scarce. This is partially because, certainly in Flanders and the more northern European countries, the use of natural ventilation for pig housing systems is limited (European Commission, 2013). Furthermore, the determination of EFs from naturally ventilated buildings is hampered by the lack of a solid reference to determine ventilation rates from naturally ventilated buildings. The CO_2 balance method, heat balance method or tracer gas method are often used to estimate ventilation rates, but can only give a rough estimate and no field reference method is available against which these methods can be evaluated (Kim *et al.*, 2008; Ogink *et al.*, 2013b). Finally, even if a reference measuring technique would exist, comparative emission measurements between natural and mechanical ventilation would not be straightforward since the respective housing systems will probably not have an identical barn layout and management, making it hard to compare only the effect of the ventilation system.

Spraying oil or water mixtures has shown to be effective in reducing PM concentrations (Takai *et al.*, 1995; Takai & Pedersen, 2000). Takai *et al.* (1995) found reductions up to 52 % in respirable PM inside fattening pig housing systems when spraying one to four times each day a rapeseed oil mixture (5 to 20 % oil in water). However, spraying with water is not believed to strongly reduce PM concentrations (Takai & Pedersen, 2000). The investment costs associated with the use of fogging or misting are believed to be between 3.8 and 6 € per fattening pig (European Commission, 2013).

Cleaning practices may affect the indoor air quality and hence emissions. Poor hygienic conditions inside pig buildings have been reported to increase indoor concentrations of NH_3,

CO_2 and airborne and respirable PM (Banhazi *et al.*, 2008a; Banhazi *et al.*, 2008b; Cargill & Banhazi, 1998; Lee *et al.*, 2005). However, attempts to reduce PM concentrations through vacuum cleaning were not successful (van't Klooster *et al.*, 1993).

1.4.1.3 MANURE MANAGEMENT

A possible way to reduce NH_3 emissions from manure is to inhibit the urease enzyme activity with the aid of urease inhibitors (Varel, 1997). However, due to the need for a frequent administration of these inhibitors to the manure and the unknown effects of the inhibitors on the crops after application of the manure to the fields, the additives are rarely used in practice (Ndegwa *et al.*, 2008).

Because the equilibrium in liquid manure between NH_3 and NH_4^+ is determined by pH and temperature, lowering the pH of the manure at constant temperature lowers the amount of NH_3 present in the manure. Between pH 7 and pH 10, the greatest increase in NH_3 release from the manure takes place, while at pH levels below 7, only small amounts are released. Below pH 4.5, almost no volatilisation occurs (Hartung & Phillips, 1994). Therefore, acidification of manure can be used as a NH_3 emission mitigation technique. High reduction rates are accomplished by adding acids [e.g. sulphuric acid (70 - 85 % reduction in NH_3 emission)(Jensen, 2002; Kai *et al.*, 2008; Sørensen & Eriksen, 2009; Stevens *et al.*, 1989) or phosphoric acid (90 % reduction in NH_3 emission)(Al-Kanani *et al.*, 1992)] to the manure. Recently, the acidification of animal slurry was reviewed by Fangueiro *et al.* (2015). In this review, not only the impact of acidification of animal slurry on the NH_3 emissions from the barn, but also its impact on land application and the consequences for combining this technique with other techniques, are discussed (Fangueiro *et al.*, 2015). Manure acidification can be accomplished by pumping the manure to a process tank where the acid is added. Usually, the manure is also aerated to prevent the formation of H_2S. Afterwards, part of the manure is transported back to the barn to ensure that the pH of manure in the barn is low enough. The investment cost for an acidification unit, capable of handling 2400 m³ of manure) is around 200 000 € with an extra cost of 1.4 – 7 € per animal place per year for maintenance, electricity and the cost of the acid (European Commission, 2013). Acidifying additives (e.g. alum or calcium and magnesium salts) can also lower the pH, but cannot maintain a stable pH as opposed to the acids, which makes these less cost-effective. On the other hand, the use of strong acids on a farm can be hazardous (Ndegwa *et al.*, 2008). The

addition of quebracho tannins to manure has shown *in vitro* to strongly reduce (> +85 %) CH_4 emissions, probably by reducing the population of methanogenic microorganisms (Whitehead *et al.*, 2013). However, the effectiveness of most of these substances is discussed only in a limited number of articles. Their effectiveness and applicability on a large scale are not yet clear. Feeding measures to reduce urinary and manure pH were already discussed in section 1.4.1.1 Diet.

Temperature also plays an important role in the NH_4^+– NH_3 equilibrium in manure and the volatilisation of NH_3 from manure. Higher temperatures favour NH_3 concentrations, because of the positive influence of temperature on the dissociation constant between NH_3 and NH_4^+ (Groot Koerkamp *et al.*, 1998). Therefore, reducing the temperature of the manure could possibly reduce NH_3 emissions. Cooling the manure with cooling coils in the floor of the manure pit has shown to reduce NH_3 emissions (-7 – -47 %), however the authors could not exclude the possibility that this reduction was also partially caused by low ambient room temperatures (Andersson, 1998). Cooling of manure to a temperature of 10 °C is predicted to reduce (-74 %) CH_4 emission from pig buildings, but can increase emissions from outside storage afterwards (Sommer *et al.*, 2004). However, cooling of manure can be expensive and is not widely applied.

Binding free NH_4^+ ions in the manure can reduce NH_3 emissions. Popular binding agents for NH_4^+ are zeolite (65-71 % reduction in NH_3 emissions), sphagnum moss (75 % reduction of NH_3 losses) and saponins, although saponins are considered to be less effective (Al-Kanani *et al.*, 1992; Panetta *et al.*, 2005; Portejoie *et al.*, 2003). Even when using the effective binding agents zeolite and sphagnum moss, still large amounts have to be added, making them a financially unattractive option (Ndegwa *et al.*, 2008).

Instead of adding substances to the manure, NH_3 emissions can also be lowered by frequent removal of manure from the manure pit or flushing of the manure pit. This can be on a daily basis (Lim *et al.*, 2004), every two or three days (Lachance *et al.*, 2005) or on a (two) weekly basis (Hartung & Phillips, 1994; Osada *et al.*, 1998). Changing the manure removal or pit flushing rate can also influence the GHG emissions. Weekly removal of manure is reported to reduce (-10 – -19 %) CH_4 and (to a lesser extent) CO_2 emissions (Guarino *et al.*, 2003; Osada

et al., 1998). The effect on N_2O emission is less clear (Guarino *et al.*, 2003; Osada *et al.*, 1998).

Table 1.1. Overview of the different mitigation techniques and their effect on pollutant concentrations or emissions.

Mitigation principle	Mitigation technique	Reduces emissions or concentrations of …[1]	Enhances emissions or concentrations of …[1]	Source-oriented technique ?
Diet	Phase feeding	NH_3		Yes
	Reducing nitrogen content in the feed	NH_3, $CH4_?$	CH_4?	Yes
	Reducing urinary pH (acids or dEB)	NH_3, CH_4?		Yes
	Adding dietary fibres	NH_3, CO_2?	CH_4, CO_2?	Yes
	Adding additives[2]	NH_3?		Yes
	Adding animal fat/oil to the feed	PM?	PM?	Yes
	Pelleting the feed	NH_3? PM?		Yes
	Coating of the pellets with fat	PM?		Yes
Housing systems, techniques & management practices	Segregating urine and manure			
	flat scraper	NH_3?		Yes
	V-shaped scraper	NH_3		Yes
	Belt systems	NH_3, CH_4	N_2O?	Yes
	Reducing emitting surface			
	Sloped pit walls	NH_3		Yes
	Partially slatted floors	NH_3, PM, N_2O?, CH_4?	N_2O?, CH_4?	Yes

[1] If the effect of a mitigation technique is not clear from the literature research, a question mark is put behind the relevant pollutant.

[2] antibiotics, probiotics, exogenous enzymes, plant extracts or zeolites.

Table 1.1. Continued.

Mitigation principle	Mitigation technique	Reduces emissions or concentrations of …[1]	Enhances emissions or concentrations of …[1]	Source-oriented technique?
Housing systems, techniques & management practices	Filtration of the indoor air			
	Dry air filters	PM		No
	Electrostatic precipitators	PM		No
	Ionisation systems	PM		No
	Ventilation management			
	Ventilation management	PM?	NH_3?, CO_2?	No
	Position of ventilators	NH_3, PM		No
	Pit ventilation	NH_3?	NH_3?	No
	Spraying oil and water mixtures	PM		No
	Cleaning practices	NH_3, CO_2		Yes
Manure management	Urease inhibitors	NH_3		Yes
	Acidifying manure			
	by acids	NH_3		Yes
	by acidifying additives	NH_3?		Yes
	Cooling of the manure	NH_3		Yes
	Binding free NH_4^+	NH_3		Yes
	Frequent manure removal	NH_3 , CO_2, CH_4		Yes

[1] If the effect of a mitigation technique is not clear from the literature research, a question mark is put behind the relevant pollutant.

Table 1.2. Overview of the reduction percentages[1] for different mitigation techniques.

Mitigation principle	Mitigation technique	Reduction percentage	Target	Pollutant
Diet	Reducing nitrogen content in the feed (-10 g kg^{-1})	6 to 13 %	Emission	NH_3
	Reducing urinary pH (acids or dEB)			
	addition of 1 % of benzoic acid	15 %	Emission	NH_3
	addition of Ca and P salt	30 %	Emission	NH_3
Housing systems, techniques & management practices	Segregating urine and manure			
	flat scraper	50 %	Emission	NH_3
	V-shaped scraper	50 %	Emission	NH_3
	Belt systems	50 %	Emission	NH_3
	Reducing emitting surface			
	Sloped pit walls	30 to 40 %	Emission	NH_3
	Partially slatted floors	35 to 45 %	Emission	NH_3
	Ventilation			
	Position of ventilators	30 to 70 %[2]	Concentration	NH_3
		78 %	Concentration	PM
	Pit ventilation	43 %	Concentration	NH_3
	Spraying oil and water mixtures	52 %	Concentration	PM
Manure management	Acidifying manure by acids	90 %	Concentration	NH_3
		70 %	Emission	NH_3
	Cooling of the manure	7 to 47 %	Emission	NH_3
	Binding free NH_4^+	65 to 75 %	Emission	NH_3

[1] The reduction percentages are reported “per animal place”, but could also be interpreted as “per animal”. As already mentioned, reported reduction percentages can differ considerably between studies as a consequence of differences in housing system, management, climate or measurement technique.

[2] At animal height.

Table 1.3. Overview of the costs for different mitigation techniques.

Mitigation principle	Mitigation technique	Cost (in €) of the technique per animal place[1]
Diet	Phase feeding	
	two phase	3.5
	three phases	6.2
	Reducing nitrogen content in the feed (-10 g kg^{-1})	2
	Reducing urinary pH (acids or dEB)	
	addition of 1 % of benzoic acid	10
	changing from $CaCO_3$ to $CaCl_2$ (6 g kg^{-1})	9
Housing systems, techniques & management practices	Segregating urine and manure	
	V-shaped belt systems	-22
	Reducing emitting surface	
	sloped pit walls	86 to 109[2]
	Spraying oil and water mixtures	10.3 to 16.2
Manure management	Acidifying manure	
	by acids[3]	1.4 to 1.7

[1] For calculating the costs per animal place, it was assumed that on 1 animal place 2.7 pigs are kept each year (Vrints & Deunick, 2014).

[2] Extra cost compared to building a conventional barn with fully slatted floor and manure pit.

[3] Plus an initial investment cost of 200 000 € to handle 2400 m^3 manure.

1.4.2 END-OF-PIPE TECHNIQUES

Air leaving the barn can be treated to capture or trap different pollutants by means of (bio)filtration or air scrubbing. The ventilation system extracts air from the barn compartments into a central duct and forces it through the (bio)filter or air scrubber. Therefore, these techniques can only be applied in buildings with point extraction (e.g. mechanically ventilated buildings).

Biological and chemical air scrubbers are primarily used to lower NH_3 emissions. In short, an air scrubber consists of a reactor, filled with an inert or inorganic packing material (Figure 1.3). This packing material is sprayed with a washing liquid (e.g. water or an acid solution) and thus wetted. Air is extracted from the barn and introduced into the scrubber. This results in an intensive contact between air and water, resulting in a mass transfer of water soluble compounds (e.g. NH_3) from gas to liquid phase. A fraction of the washing liquid is recirculated, while the other fraction is discharged and replaced by fresh water or acid solution (Melse *et al.*, 2009).

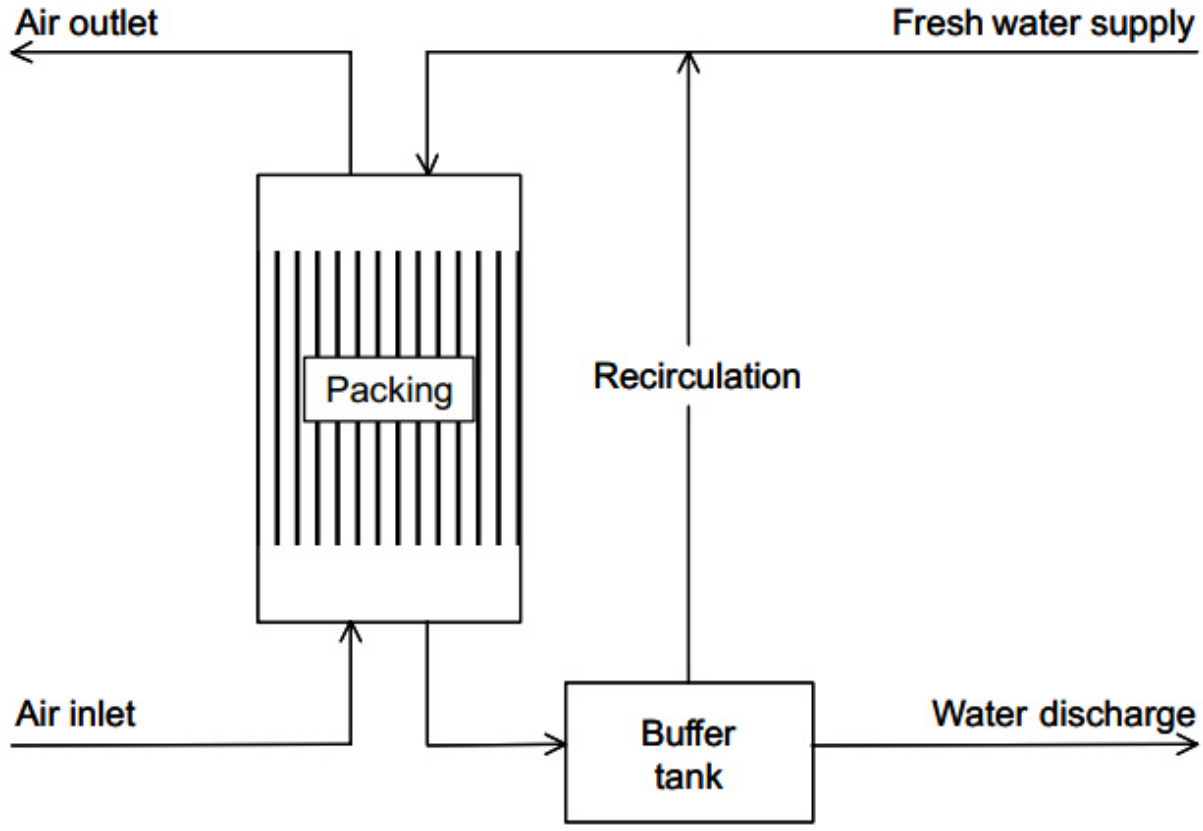

Figure 1.3. Schematic representation of a counter-current air scrubber. This picture was adapted from Melse & Ogink (2005).

Acid scrubbers remove NH_3 by using a sulphuric-acid solution as washing liquid, leading to a pH-driven dissociation of NH_3 into NH_4^+ and hydroxide ions and the subsequent formation of ammonium salts. In order to ensure that the pH of the liquid phase remains low, acid is added to the liquid and ammonium salts are removed with the discharge water which is replaced by fresh water. NH_3 removal efficiencies of 90% to 99% can be achieved (Melse & Ogink, 2005; Melse *et al.*, 2009). Biological scrubbers (also called bioscrubber or biotrickling filter) transform NH_3 into nitrite and nitrate by means of bacterial conversion in a nitrification process. The nitrifying bacteria are present in a biofilm and/or suspended in the washing liquid. The resulting nitrite and nitrate are removed with the discharge water. Average NH_3 removal efficiencies ranging from 35 % to 90 % are reported (Girard *et al.*, 2012; Melse & Mol, 2004; Melse & Ogink, 2005; Scholtens *et al.*, 1988). These lower efficiencies can be caused by inhibition of nitrifying bacteria due to high NH_3 and nitrite concentrations (Melse & Ogink, 2005; Melse *et al.*, 2009). Finally, multi-stage scrubbers are designed to not only remove NH_3, but also to remove other pollutants or compounds from the exhaust air. A multi-stage scrubber combines different types of techniques (e.g. an acid and biological scrubber), each designed to remove a specific pollutant or type of compounds. Therefore it is also called multi-pollutant scrubber (Melse *et al.*, 2009). These multi-stage scrubbers also have the potential to remove PM_{10} and $PM_{2.5}$ from the exhaust air, e.g. by making use of water-curtains (Aarnink *et al.*, 2007b; Ogink & Hahne, 2007). The total costs (investment cost & operational cost, both per year) per year and per animal place for a fattening pig facility with 460 to 700 animals were estimated by Arends *et al.* (2008) to be 17.1 to 19.2 €, 19.0 to 20.3 €, 17.9 to 24.0 € and 18.3 to 20.2 € to use a biological, a 2-stage system (chemical and water stage), a 3-stage system with a water stage, a chemical stage and a biological stage and a 3-stage system with two water stages and a biological stage, respectively. Melse and Ogink (2005) reported investment costs of 33 and 36 € per animal place for a chemical and biological scrubber respectively and yearly operational costs of 11.7 and 11.3 € per animal place, respectively.

Biofilters are usually made up of a filter bed, a mixture of for example wood chips, peat and compost. NH_3 is removed by nitrification and conversion into nitrous and nitric acid (Melse & van der Werf, 2005). However, the applicability of biofilters on a long-term is under question

because of problems that can arise when using biofilters: clogging by accumulation of dust, acidification and inhomogeneous humidification (Melse & Ogink, 2005; Melse *et al.*, 2009).

Literature on the reduction of CH_4 by using a scrubber of biofilter is contradictory. Some authors do not expect that these filters reduce CH_4 emissions, partly due to the low solubility of CH_4 in water. However, methanotrophic bacteria, present in the filter bed can convert CH_4 into CO_2 and H_2O. Due to the relatively low CH_4 concentrations in exhaust air from a barn, very large biofilter volumes would be necessary to reduce CH_4 concentrations significantly (Girard *et al.*, 2011; Melse *et al.*, 2009; Melse & van der Werf, 2005). Formation of N_2O can occur in a biofilter or bioscrubber as a by-product of the nitrification and denitrification processes (Maia *et al.*, 2012; Melse *et al.*, 2009; Melse & van der Werf, 2005).

1.4.3 INTERACTION BETWEEN POLLUTANTS

As mentioned above, reducing the emission of one pollutant can increase the emission of another pollutant. For example, the formation of CH_4 in manure becomes inhibited by the presence of NH_3. Inhibition starts when the free ammonia concentration in the manure reaches 1.1 g-N litre^{-1} (Hansen *et al.*, 1998). Therefore, reducing the concentration of NH_3 in manure may increase the CH_4 production rate. On the other hand, reducing the concentration or emission of one pollutant can simultaneously reduce the concentration or emission of another pollutant. For example, NH_3 (or its ionised form NH_4^+) formation is the first step in the production of N_2O from manure (see section 1.2.2). Reducing the concentration of NH_3 (and NH_4^+) in manure can therefore reduce the N_2O production in the manure. It is also known that NH_3 in the atmosphere plays an important role in the formation of secondary PM (in the form of ammonium salts). Reducing the concentration of NH_3 in the atmosphere can therefore also reduce the formation of ammonium salts and thus secondary PM formation (Erisman & Schaap, 2004). The reduction of NH_3 concentrations inside livestock buildings could perhaps also lead to a reduction of indoor PM concentrations. This would be the case if the formation of ammonium salts is not limited to take place in the atmosphere, but also occurs inside livestock buildings. However, knowledge about the occurrence of ammonium salts inside animal houses is still very scarce. The formation of secondary PM in the atmosphere may take several days. The specific circumstances inside an animal house (high NH_3 concentrations, high relative humidity and enhanced bacterial activity) could speed up this process. Hence, it is not imaginary that

ammonium salts are formed inside livestock buildings. Different authors have characterised aerosols in the vicinity of livestock buildings. For example, Lammel *et al.* (2004) compared background aerosol concentrations with outdoor aerosol concentrations at the farm and concluded that the farm was a source of primary as well as secondary particles, mainly in the form of ammonium and nitrate salts, organic matter and calcium. At the farm, the number of particles in the 1-4 µm fraction increased with a factor 1.7 – 2.1 and with a factor 2.0 to 6.4 in the fraction 4-20 µm as compared to the background levels (Lammel *et al.*, 2004). Since the measurements were not performed inside the barns, it is impossible to determine if the secondary particulate matter was already formed inside the barn or outside the barn. Therefore, Roumeliotis and Van Heyst (2008) chemically characterised $PM_{2.5}$ within an experimental layer house. The results of three different measuring days (and background measurement at the ventilation inlet) indicated that about 50 to almost 100 % of the total $PM_{2.5}$ concentration existed of secondary particulate matter. The differences between the levels of secondary particulate matter from the inside measurements and the measurements at the air inlet could indicate that secondary particulate matter was formed inside the layer house (Roumeliotis & Van Heyst, 2008). The results of a subsequent study by the same research group in a commercial broiler barn indicated that secondary particulate matter formation was enhanced during warmer periods. The mean ratio of the inorganic aerosol concentration and the $PM_{2.5}$ emission was 26 % for the cold period compared to 42 % for the warmer period (Roumeliotis *et al.*, 2010). A recent preliminary study inside a commercial fattening pig facility showed 3 to 4 times higher NH_4^+ concentrations in indoor PM_{10} samples as compared to the NH_4^+ concentrations at the air inlet (Ulens *et al.*, 2014). This is again an indication that secondary PM formation might occur inside livestock buildings.

When reviewing mitigation techniques, not only the need for simultaneous desirable effects on several pollutants is important. Parameters like installation and operational costs, effect on animal health and productivity and effect on human health are also important to keep in mind. These parameters play an important role in determining the successful implementation of these mitigation techniques in practice. In this overview of mitigation techniques, influences on soil emissions (after land application) were not taken into account. It can be concluded that mitigation techniques should always be evaluated taking into

account the most important pollutants and other important parameters simultaneously, instead of performing an individual evaluation.

1.5 ASSESSING EMISSION LEVELS AND REDUCTION EFFICIENCIES

Different techniques exist to measure the different pollutant concentrations. Some pollutants (e.g. NH_3 and GHGs) can be measured simultaneously with one technique, while other pollutants (e.g. PM) rely on a totally different measuring technique. Strategies to measure these pollutants can also vary depending on the application. Distinction can be made between measurements to compare treatments and measurements to define an emission factor.

1.5.1 MEASURING TECHNIQUES

1.5.1.1 GAS MEASURING TECHNIQUES

One of the simplest and most reliable techniques to measure NH_3 is an aqueous acid trap, based on wet chemistry (gas concentration determination in aqueous medium techniques). A known volume of air is passing through an acid solution, trapping the NH_3 in the solution. Afterwards the NH_4^+ concentration in the solution can be determined using colorimetric techniques or via other established analytical methods (Ni & Heber, 2001). This technique is considered to be the standard technique for NH_3 determination (Harper, 2005). Other, relative simple instruments to measure NH_3 and CO_2 concentrations are gas detection tubes. These disposable tubes have a solid surface that undergoes a colour change when a specific gas adsorbs onto the surface. Both active (with pump) and passive (without pump) gas detection tubes exist (Ni & Heber, 2001).

Fourier transform-infrared spectroscopy (FTIR) can be used for both NH_3 and GHGs. In FTIR, an interferometer produces an interference wave which interacts with the air sample, resulting in an interferogram. The Fourier transform of this interferogram yields a spectrum that can be compared with spectra of known samples (Ball, 2001). Different types (e.g. dual-path or open-path) of FTIR spectrometers exist (Hu *et al.*, 2014). Another spectroscopic instrument than can be used to measure NH_3 and GHGs, is an infrared (IR) photoacoustic system (PAS). Unlike the FTIR spectrometer, which analyses the whole IR spectrum, the IR

PAS measures at one spectral band of the IR spectrum (Phillips *et al.*, 2001). The Innova photoacoustic gas monitor 1314 (Innova AirTech Instruments, Santa Clara, CA, USA), which was used in this thesis, works on this principle. An air sample is drawn into the measuring cell. The measuring cell is then closed and the sample is irradiated with an IR beam, chopped at a constant frequency. The wavelength of the incoming beam can be modulated by means of optical filters. At a certain wavelength, specific for each gas, the IR light will be absorbed by the gas molecules. The excitation of the molecules results in heat generation, increasing the temperature in the cell and, since this is a closed cell, the pressure increases. Due to the chopped IR beam, a series of pressure pulses will emerge. These pressure pulses (≈ sound waves) are detected by microphones, situated in the wall of the measuring cell which convert the signal into a voltage differential and ultimately into a concentration (Yamulki & Jarvis, 1999). Laser absorption spectroscopy (LAS) can also be used to measure NH_3, CH_4 and N_2O concentrations. This technique operates on the principle that every molecule absorbs light at a specific wavelength. The emitting laser beam is tuned to a wavelength specific for the molecule of interest while minimising the possibility of absorption by interfering gases. The emitted laser beam is reflected back by a reflector to a transceiver unit and the intensity of the reflected laser beam is measured. Therefore, this technique provides line average concentrations between two measuring points. The path between the emitter and reflector can be either open or closed (Hu *et al.*, 2014; Mosquera *et al.*, 2014). Another option to (indirectly) measure NH_3 concentrations is with chemiluminescence analysers. First, NH_3 has to be catalytically converted (with stainless-steel as converter) in nitric oxide (NO) at very high temperature (795 °C) (Aneja *et al.*, 1978). Afterwards, the generated NO is further oxidized within the chemiluminescence analyser using ozone and nitrogen dioxide (NO_2), in an excited state, is produced. When the NO_2 molecules return to a lower energy state, they release photons, which are detected by the instrument (Phillips *et al.*, 2001).

Due to the high purchase prices of the aforementioned systems for continuous monitoring (FTIR, PAS and chemiluminescence analyser), research is being performed on simple, low price measuring systems which can still monitor continuously as opposed to the wet chemistry method or gas detection tubes. One of these measuring systems is the solid state sensor, which can be used to measure NH_3 or GHG concentrations. This sensor works by measuring the change in a physical property due to adsorption/desorption processes and

chemical reactions on the surface of a sensing element, a solid-state film of a gas-sensitive material (Capone *et al.*, 2003). A review on solid-state sensors was written by Capone *et al.* (2003).

For further reading on this matter, several recent reviews are available. An overview of other techniques to measure NH_3 emissions can be found in a review by Ni and Heber (2001). A review on techniques for NH_3 or, more general, gas emissions, with a focus on agriculture and livestock, can be found in reviews by Philipps *et al.* (2001), Hu *et al.* (2014) and Mosquera *et al.* (2014).

1.5.1.2 PARTICULATE MATTER MEASURING TECHNIQUES

Particulate matter concentrations in animal houses can be measured by different techniques. The most commonly used devices are: a gravimetric sampler, an optical particle counter, a beta attenuation monitor or a tapered element oscillating microbalance.

A gravimetric sampler draws air through a size selective inlet or cyclone. Particles with a size larger than the desired size collapse onto an impaction plate or hit the wall of the cyclone due to centrifugal forces. Particles, within the desired size range, are deposited onto a filter. By pre- and post-weighing the filter, the PM concentration can be calculated and be expressed in mass as mg of PM per m^3 of air.

A beta attenuation monitor also consists of a size selective inlet and a filter. However, instead of a single filter, a filter tape is used which allows for continuous monitoring. The filter tape is radiated with a beta radiation source and the transmission of this beta radiation over the tape is measured by a detector. Transmission before and after sampling is a measure for the mass of collected PM (Jaklevic *et al.*, 1981).

A tapered element oscillating microbalance exists of a hollowed tapered tube with a filter cartridge at the end of the tube. Air passes over the filter into the tube, while particles are collected onto the filter. The frequency of oscillation of the tapered tube (and filter) is dependent upon the physical properties of the tapered tube and the mass of the filter. This frequency changes when particles are deposited onto the filter. This change in frequency is directly related to the mass of the particles on the filter (Patashnick & Rupprecht, 1991).

An optical particle counter detects particles by light scattering inside a measuring cell. The intensity of a scattering light signal is classified to a certain particle size and the scattering light pulse of every particle is being counted. The Grimm 1.109 spectrometer (Grimm Aerosol Technik GmbH & Co. KG, Ainring, Germany), which was mostly used in this thesis, works on this principle. The wavelength of the laser diode, used in this spectrometer, is at 655 nm. By modulating the intensity of the laser beam, particles can be detected in a range of 0.25 µm to 32 µm. Incoming air is focussed and guided through the inner area of a measuring volume, created by focussing the laser beam. Particles in the incoming air emit scattering light when hit by the focused laser beam. This scattering light is detected under a scattering angle of 90° and collected by a receiver diode. The signal, collected at the detector, is finally classified into size channels based on the intensity of the signal. This device counts the number of particles in an air stream with a sampling volume of 1.2 l/min. Conversion to mass concentrations is based on certain algorithms and a calibration procedure (maximum 5 % deviation compared to a reference unit)(manual Grimm 1.109 spectrometer).

1.5.2 MEASURING STRATEGIES

As already mentioned in the introduction of this paragraph, measuring strategies can be split in strategies to measure differences in pollutant concentrations between treatments and strategies to define an emission factor. Strategies to compare between treatments will not be discussed because the number of strategies that are used in literature are almost uncountable. While in some studies whole fattening periods are monitored, other studies report data from measurements spread over the fattening period with variations in frequency and duration. On the other hand, an emission factor should reflect the mass of a certain pollutant that is emitted over a year and in an ideal case, measurements should be performed for one whole year. An emission factor (EF, kg year^{-1} (animal place)$^{-1}$) can be calculated as the cumulative emission of a certain pollutant over one year, divided by the number of animals and corrected for vacancy in the barn (Eq. 1.3). Different emission rate (ER, g h^{-1}) equations for PM (Eq. 1.1) and gas measurements (Eq. 1.2) are used.

$$ER = Q * C_o * 10^{-6} \quad \text{Eq. 1.1}$$

$$ER = Q * [C_o - C_i] * 10^{-6} * \frac{w_m}{V_m} * \frac{P}{P_{ref}} * \frac{T_{ref}}{T} \quad \text{Eq. 1.2}$$

$$EF = \frac{\sum ER}{N}\left(\frac{10^{-3} * 24 * 365}{a}\right) * VP \qquad \text{Eq. 1.3}$$

Where Q is ventilation rate ($m^3\ h^{-1}$), C_o is concentration at the air outlet (ppm for the gasses, $\mu g\ m^{-3}$ for the PM fractions), C_i is concentration at the air inlet (ppm for the gasses, $\mu g\ m^{-3}$ for the PM fractions), w_m is molar mass ($g\ mol^{-1}$), V_m is molar volume at reference temperature and pressure ($m^3\ mol^{-1}$), P is air pressure (hPa), P_{ref} is reference air pressure (1013.25 hPa), T is indoor temperature (K), T_{ref} is reference temperature (273.15 K), N is the number of measuring hours, a is the number of animals and VP is the correction factor for the vacancy (0.9 for fattening pigs).

However, several reduced measuring strategies exist, both for PM as for NH_3. These measuring strategies should take into account the between-farm variance, the within-farm variance, and the instrument measurement variance in order to get a good estimate of the true emission factor. Each of these variances attribute to the overall measurement variance of the mean emission of a housing system (Hofschreuder *et al.*, 2008).

1.5.2.1 GAS MEASURING STRATEGIES

Since NH_3 was the first gas in agriculture that was subjected to mandatory emission reduction, several researchers have tried to find reduced measuring strategies for the determination of NH_3 emission factors. In the Netherlands, a first (slightly) reduced measuring protocol was developed under the Green Label framework (Groen Label, 1996; Mosquera & Ogink, 2011). The goal of this measuring protocol was to accurately estimate the mean annual NH_3 emission of a housing system. The Green Label protocol was still a very elaborate sampling protocol with measurements in two growth cycles (e.g. fattening periods), one in summer and one in winter, both on the same farm. Ammonia concentrations had to be measured on a continuous basis, i.e. every 5 to 10 minutes. Afterwards, hourly means were used in further calculations (Groen Label, 1996). This protocol is also currently used in Flanders to determine NH_3 emission factors. This extensive, expensive and time consuming protocol was followed-up in The Netherlands by an alternative sampling protocol (multiple-location approach), based on measurements at several (four) farms with the same housing system. In this new protocol, six 24-hour sampling periods per farm location, distributed over one year and randomly taken in

subsequent two month periods, are prescribed. For animal categories with growth cycles (e.g. fattening pigs) measurements had to be equally divided over the production cycle (e.g. fattening period) (Ogink *et al.*, 2011). Recently, an alternative sampling protocol (case-control approach) has been suggested. This approach is based on performing simultaneous measurements in both a newly proposed housing system (or in an existing housing system, but with application of a new management strategy; referred as "case" in this protocol) and a reference system (with known emission factor), both located at the same farm. The number of measurements per farm (6) and the conditions concerning spreading of the measurements over time remained the same, but the number of farms decreased from four to two. On the basis of the difference between the emissions in the reference system and the new housing system, the emission factor of the new housing system is estimated (Ogink *et al.*, 2013a). These two alternative protocols (multiple-location approach and case-control approach) are incorporated in the international VERA protocols (VERA, 2011). Mosquera *et al.* (2011) investigated two alternative approaches to shorten the measuring protocol with four farms. The sampling period was shortened from one year to six months, resulting in only three 24-hour measuring periods per farm. Using the results from those six months, either solely or in combination with a mathematical model, led to a small increase in overall random measurement error of the mean emission (Mosquera & Ogink, 2011). Although not specified, the aforementioned protocols could also be used to determine GHG emission factors.

Besides these protocols to determine housing system-specific emission factors, reduced building-specific NH_3 measuring strategies for fattening pigs were proposed by Vranken *et al.* (2004) and later refined by Dekock *et al.* (2009). In the final protocol, a linear model containing ventilation rate, mean weight of the animals and inside and outside temperature, measured at specific times, was used to model the NH_3 emission from a building. In total, four measurement periods (2 before day 70 and 2 after day 70 in the fattening period) per fattening period were needed to get a good estimate of the NH_3 emissions. To get an EF for the building, three fattening periods had to be monitored (Dekock *et al.*, 2009; Vranken *et al.*, 2004).

1.5.2.2 PARTICULATE MATTER MEASURING STRATEGIES

In comparison with NH_3, less reduced strategies exist for PM. Hofschreuder *et al.* (2008) proposed a measuring strategy, similar to the Dutch measuring strategy for NH_3, with measurement at four farms. These authors proposed to perform six 24-hour measurements on each farm, spread over one year, randomly taken in subsequent two month periods. For fattening pigs (or other animal categories with production cycles), measurements also had to be equally divided over the growing period (Hofschreuder *et al.*, 2008). Recently, Van Ransbeeck *et al.* (2012) developed a reduced PM measuring strategy for fattening pigs. This strategy could be used for both indoor PM concentrations and PM emissions, as well as for three PM fractions (PM_{10}, $PM_{2.5}$ and PM_1). Four measuring periods in one fattening period, with two consecutive measuring days per measuring period, were proposed. Sampling should be done during at least two consecutive fattening periods to determine an emission factor. The measuring periods should be between day 1 - day 9, day 29 - day 41, day 57 - day 66 and day 100 – day 120 in the fattening period. If the impact of PM on human or animal health also has to be evaluated, the authors suggest an extra measuring period between day 93 – day 103 to include the expected overall maximum PM concentrations, observed during the period day 93 – day 103 (Van Ransbeeck *et al.*, 2012).

1.6 PROBLEM STATEMENT

The intensification of animal husbandry in Flanders, Europe and the United States has come with great challenges regarding the mitigation of pollutant emissions (NH_3, CO_2, CH_4, N_2O and PM) and the associated detrimental effects on the environment and the human health. These challenges are worldwide recognized by the governments, politicians and concerned citizens. Mitigation strategies are applied and legislation is passed in an attempt to constrain the emissions and to lower their influence on the environment. However, most strategies or legislation only take into account one pollutant or one group of pollutants (e.g. NH_3 or greenhouse gases). For example, in Flanders the decree on emission-low housing systems only addresses NH_3 emissions. Consequently, the approved techniques are not designed for the reduction of, for example, greenhouse gases. Moreover, emissions of other pollutants may even be enhanced by the implementation of a "single pollutant" mitigation technique. Indeed, the currently going pollutant-by-pollutant approach does not take into account the **possible relations among different pollutants** or the extent to which different pollutants can affect each other. Although there is an emerging trend to investigate several pollutants at the same time in a so-called **multi-pollutant research approach**, up to now most studies have been limited to a small group of pollutants.

While **end-of-pipe techniques** have shown great potential in reducing NH_3 and PM emissions, they suffer from some drawbacks. Since pollutants are only reduced at the ventilation exhaust, no improvement of the indoor climate is achieved. Furthermore, these techniques are costly, both in installation and in operation. Besides, due to the need to cluster the exhaust air from different compartments and direct it to one single end-of-pipe technique, this technique can only be used in buildings with central extraction. **Mitigation techniques inside the barn** are usually cheaper, can be mostly applied to a wider range of housing systems and animal categories and, most importantly, can effectively improve the indoor climate. However, the reduction percentages obtained with these techniques are generally lower than those obtained with end-of-pipe techniques. Although numerous studies have been performed on mitigation techniques inside the barn (such as influence of feed and manure and farm management) there still remain ample questions. In order to assess the reduction percentages of mitigation techniques inside the barn or to determine emission factors for new housing systems, **emission measurements** are necessary. These

emission measurements can be both **time consuming and expensive**. Therefore, the accuracy of reduced sampling strategies should be tested, as well as their applicability.

Although already a lot of research has been performed on reducing the emissions of different pollutants from pig husbandry, it is clear that there still remains a lot of work to be done. Optimisations in the field of measuring strategies are also desirable. To summarise, the following knowledge gaps can be identified for future research:

1. Are mitigation techniques inside the barn and, more specifically source-oriented techniques, applicable to multiple pollutants and effective in reducing multiple pollutants at the same time?
2. What are the relations among the different pollutants and how do pollutants affect each other?
3. Can reduced sampling strategies be used to determine emission factors?

1.7 RESEARCH OBJECTIVES & THESIS OUTLINE

In this dissertation, the indoor air quality of and emissions from **pig facilities** were studied. Data on multiple pollutants (**multi-pollutant approach**) were collected through performing different measurement campaigns at commercial and experimental farms where a limited number of new or not yet extensively studied **source-oriented mitigation techniques** were applied. This data could then be used to assess the **effectiveness** of these source-oriented techniques (knowledge gap 1), but also to generate hypotheses concerning the observed **correlations between the various pollutants** (knowledge gap 2). Already existing datasets were used to assess the **applicability of reduced sampling strategies** for the determination of NH_3 emissions (knowledge gap 3).

With the identified knowledge gaps in mind (see 1.6 Problem statement), different research objectives can be formulated. This dissertation aimed to answer the following research questions:

- What is the influence of a dry or wet cleaning protocol and of a conventional or low-ammonia-emission housing system on indoor pollutant concentrations of a commercial pig fattening facility? (knowledge gap 1; chapter 2.1)
- Based on data from the experiment above: What are the correlations between the gases (NH_3, CO_2, CH_4 and N_2O) and the PM fractions? (knowledge gap 2) Do different cleaning protocols and housing systems lead to differences in the distribution of particle sizes? (chapter 2.2)
- What is the effect of grinding intensity and feed form on the indoor PM concentrations and growth performances of weanling pigs inside a pig nursery? (knowledge gap 1; chapter 2.3)
- Is it possible to obtain a good estimate of an NH_3 emission factor via reduced measurement strategies which do not take into account possible influencing parameters? (knowledge gap 3; chapter 3)
- Based on literature and on the experiences gained in this dissertation: What are the main problems associated with performing gas and PM measurements inside livestock buildings? (chapter 4)

2 SOURCE-ORIENTED TECHNIQUES

This chapter is divided into three parts. The first part covers the influence of pen cleaning techniques and housing systems on the indoor concentrations of PM, NH_3, CO_2, CH_4 and N_2O in a fattening pig facility. In the second part and based on the data obtained in the first trial, the correlations between the indoor concentrations of the different pollutants were calculated and a particle size distribution (PSD) analysis was performed to get an overview of the dominant size ranges in fattening pig facilities. Finally, the influence of grinding intensity and pelleting of the diet on the indoor PM concentrations and the growth performance of weanling pigs was studied.

2.1 THE EFFECT OF DIFFERENT PEN CLEANING TECHNIQUES AND HOUSING SYSTEMS ON INDOOR CONCENTRATIONS OF PARTICULATE MATTER, AMMONIA AND GREENHOUSE GASES (CO_2, CH_4, N_2O)[ii]

2.1.1 INTRODUCTION

Emissions of ammonia (NH_3), greenhouse gases (methane (CH_4) and nitrous oxide (N_2O)) and particulate matter (PM) from pig housing systems have an impact on the environment. Ammonia emissions can lead to eutrophication and acidification of waterways and soils (Krupa, 2003). The emissions of greenhouse gases contribute to global warming, which is considered to be a major threat to the global environment (Flessa *et al.*, 2002). PM is strongly associated with human health problems (Bates, 2000). Furthermore, PM can be a carrier of endotoxins and microorganisms, facilitating the transmission of pathogenic microorganisms and the transportation of odorous compounds which can cause a nuisance for nearby inhabitants (Hooda *et al.*, 2000; Oehrl *et al.*, 2001; Seedorf *et al.*, 1998; Yuan *et al.*, 2010; Zhao, 2011).

Over the past few decades pig production in Flanders has intensified (European Commission, 2003; Van Gijseghem *et al.*, 2002). To minimise the environmental impact of this production intensification, new legislation has been implemented, especially with regard to NH_3 emissions. All European pig fattening facilities with more than 2000 fatteners, are subjected to the European Integrated Pollution Prevention and Control (IPPC) convention. The Intensive Rearing of Poultry and Pigs BREF (Best Available Techniques (BAT) reference document) gives an overview of the BAT, with good agricultural practice as an essential part of it, to reduce NH_3 emissions. Regarding housing systems, the main principles to reduce ammonia emissions are: reduction or cooling of the emitting manure surfaces, quick removal of manure out of the barn or the use of surfaces (e.g. slats and manure channels) which are

[ii] **Adapted from: Ulens, T., Millet, S., Van Ransbeeck, N., Van Weyenberg, S., Van Langenhove, H., & Demeyer, P.** (2014). The effect of different pen cleaning techniques and housing systems on indoor concentrations of particulate matter, ammonia and greenhouse gases (CO_2, CH_4, N_2O). *Livestock Science, **159***:123-132.

smooth and easy to clean. Other possibilities are end-of-pipe techniques (e.g. chemical wet air scrubber or bioscrubber) (European Commission, 2003).

Legislation passed in 2004 requires pig and poultry producers in Flanders to use officially approved LAE housing systems when renovating, expanding or building new animal housing. These housing systems are usually more expensive than conventional housing systems. Furthermore, some of these techniques are pure end-of-pipe techniques. Such end-of-pipe techniques are not expected to reduce indoor concentrations of ammonia.

The indoor air quality of pig housing is gaining increasing attention in relation to human and animal health (Banhazi *et al.*, 2008b; Wathes *et al.*, 1998). Exposure to high indoor concentrations of NH_3, CO_2 or PM can negatively affect the health of workers in pig houses (Asmar *et al.*, 2001; Laitinen *et al.*, 2001; Von Essen & Donham, 1999; von Essen & Banks, 2009) and of the pigs themselves (Busse, 1993; Donham, 1991; Donham, 2000; Lee *et al.*, 2005; Urbain *et al.*, 1999).The suggested maximum allowed CO_2 concentration for workers (5000 ppmv) is rarely exceeded inside pig houses (Choudat *et al.*, 1994; CIGR, 1992).

According to the study of Banhazi *et al.* (2008a; 2008b) a decrease in pen cleanliness results in higher indoor concentrations of ammonia, airborne bacteria and respirable particles. These researchers stated that improved pen cleanliness can be considered the most practical recommendation for decreasing concentrations of ammonia, respirable particles and bacteria (Banhazi *et al.*, 2008a; Banhazi *et al.*, 2008b). Recently Chen *et al.* (2011) developed a empirical emission model for commercial swine finishing barns based upon a two-year emission dataset from a commercial swine finishing farm. The vacancy period of the barn and the emissions after high pressure washing were included. In this dataset, they observed a reduction in the emissions of NH_3 and PM_{10} after wet cleaning of the barns. However, due to the limited amount of data for the empty-barn and power washing conditions, it was difficult to make accurate estimations of the influence of cleaning on the emissions (Chen *et al.*, 2011).

Information on this topic is scarce. Most studies evaluate only one single or a few important pollutants simultaneously. Furthermore, the cleaning techniques used usually differ greatly and are not always applicable in practice. Therefore we used a multi-pollutant approach to explore the effect of two practically applicable pen cleaning techniques on the indoor

concentrations of NH_3, greenhouse gases (CO_2, CH_4 and N_2O) and PM in two types of housing systems for fattening pigs. In this study, the chosen pen cleaning techniques were 1) dry cleaning versus 2) dry and wet cleaning with an additional disinfection step.

2.1.2 MATERIALS AND METHODS

2.1.2.1 LOCATION OF THE MEASUREMENTS

The study was conducted in a commercial fattening pig barn (Diksmuide, Belgium) with all-in/all-out management. Two types of housing systems were studied: (1) four conventional compartments with fully slatted floors (Figure 2.1, compartments A, B, G and H) and (2) four low-ammonia-emission compartments with reduced emission surfaces (i.e. partly-slatted floors with a central convex solid floor, a manure channel with sloped pit walls and a water channel (Figure 2.2))(Figure 2.1, compartments C, D, E and F). All compartments had one exhaust fan and automated temperature-controlled ventilation. Fresh air entered the compartments through an opening in the lower part of the door (door ventilation). The diameter of the exhaust fan was 0.45m in the low-ammonia-emission compartments and 0.5m in the conventional compartments. In both housing systems, the exhaust fan was situated above the corridor. To check the ventilation pattern, a smoke test was performed in each compartment. Phase feeding was applied in all compartments, with pelleted feed and water available ad libitum. The feed was delivered automatically by a feeding chain in the open troughs. An overview of the main characteristics of the different compartments is shown in Table 2.1.

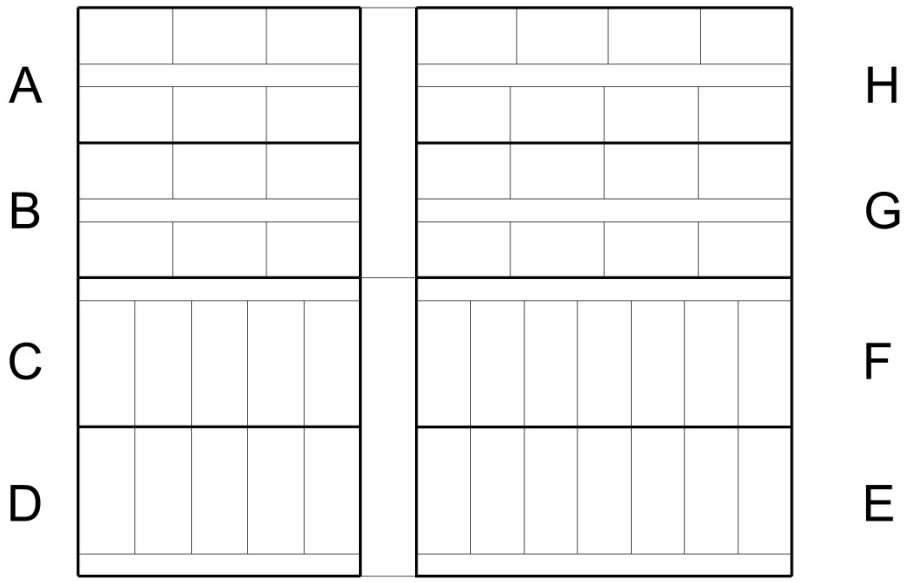

Figure 2.1. Two-dimensional floor plan of the barn with indication of the different compartments.

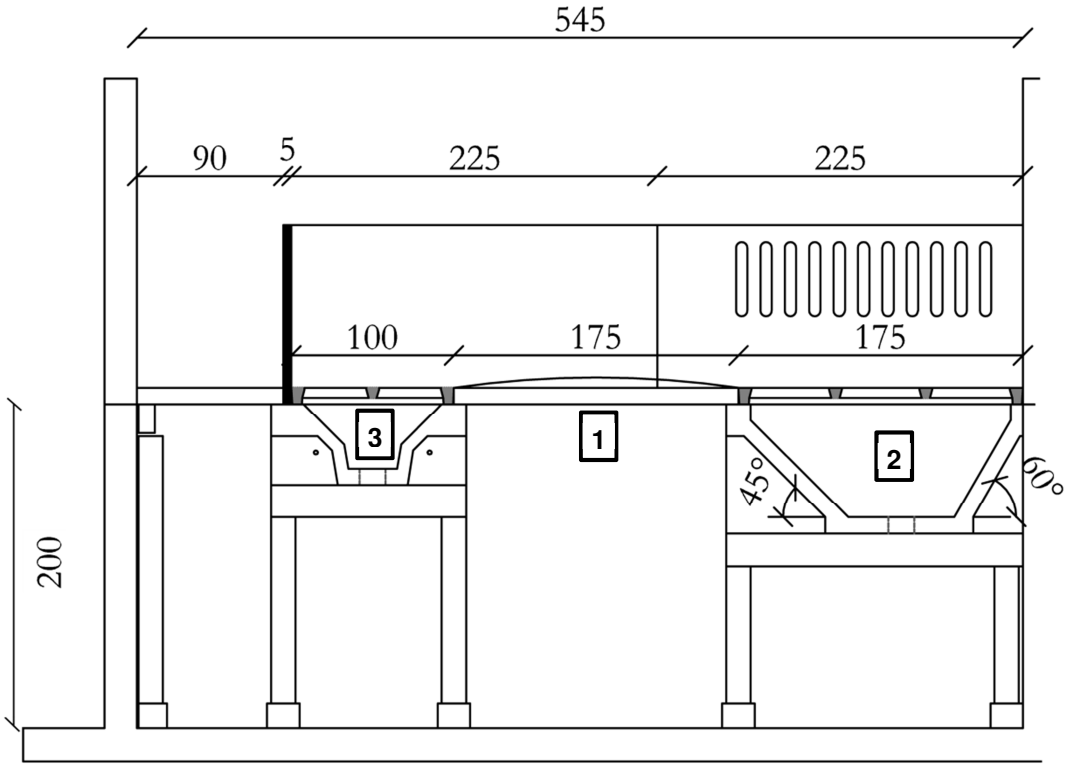

Figure 2.2. Schematic overview of the manure pit in the low-ammonia-emission compartments with partly-slatted floors and a central convex solid floor [1], a manure channel with sloped pit walls [2] and a water channel [3] (adapted from N.V. Betonbedrijf R. Dobbelaere –Bonte).

Table 2.1. Overview of the main characteristics of the different compartments.

Compartment	Type[1]	#pens	#pigs/pen	pen dimension	% open floor area	Area (m²) per pig
A	C	6	10	3.25m x 2m	11.5%	0.65
B	C	6	10	3.25m x 2m	11.5%	0.65
C	LAE	5	14	2m x 4.5m	8.8%	0.64
D	LAE	5	14	2m x 4.5m	8.8%	0.64
E	LAE	7	13	1.9m x 4.5m	8.7%	0.66
F	LAE	7	13	1.9m x 4.5m	8.7%	0.66
G	C	8	10	3.25m x 2m	11.5%	0.65
H	C	8	10	3.25m x 2m	11.5%	0.65

[1] C: conventional compartment; LAE: low-ammonia-emission compartment.

2.1.2.2 EXPERIMENTAL DESIGN

All measurements were performed between August 2011 and June 2012 (Table 2.2). Two fattening periods were monitored per compartment and the eight compartments were divided into two groups with a difference of four weeks between the start of the first group and the start of the second group. Group 1 comprised compartments A, C, F and H. Group 2 comprised compartments B, D, E and G. At the start of each fattening period, ten-week-old piglets with a weight of approximately 23 kg were randomly divided over the compartments.

Table 2.2. Overview of the start and end date of the different fattening periods.

Fattening period	Type[1]	Cleaning protocol	Compartments	Start	End
1	C	Dry	A, H	8/08/2011	19/12/2011
1	LAE	Dry	C, F	8/08/2011	19/12/2011
1	C	Wet	B, G	6/09/2011	17/01/2012
1	LAE	Wet	D, E	6/09/2011	17/01/2012
2	C	Dry	A, H	27/12/2011	8/05/2012
2	LAE	Dry	C, F	27/12/2011	8/05/2012
2	C	Wet	B, G	25/01/2012	5/06/2012
2	LAE	Wet	D, E	25/01/2012	5/06/2012

[1] C: conventional compartment; LAE: low-ammonia-emission compartment.

Two types of cleaning protocols were compared, namely a “dry” (group 1) and a “wet” (group 2) protocol. Cleaning was always performed prior to the start of every fattening period by the farmer according to standard practices. In both protocols, the pens were cleaned with brooms and a vacuum cleaner. The manure pit for the conventional housing system or the manure and water channels for the low-ammonia-emission housing system were emptied before the start of each fattening period. For the “wet” protocol the floor of the pens was subsequently soaked with water and then the floor and, in the case of the low-ammonia-emission housing system, the manure and water channel were cleaned with a pressure washer. In the “dry” protocol the manure (pit) and/or water channels were only emptied, but not cleaned with a pressure washer. Finally, for the “wet protocol”, the pens were disinfected using Virocid® (CID LINES N.V., Ieper, Belgium). After this disinfection step the pens stayed empty for at least two more days, allowing them to dry. No cleaning practices or removal of manure were carried out during the fattening periods. An overflow in the manure channel of the low-ammonia-emission housing system prevented the manure from flooding.

A parallel study (Michiels *et al.*, 2015), conducted at the same commercial fattening pig barn and during the same period, did not allow to change the cleaning protocols between the different compartments between the fattening periods.

2.1.2.3 MEASURING EQUIPMENT

Indoor concentrations of NH_3, CO_2, CH_4 and N_2O were measured using an Innova photoacoustic gas monitor 1314 (Innova AirTech Instruments, Santa Clara, CA, USA) connected to a multipoint sampler (CBISS, A1-Envirosciences ltd., Wirral, Merseyside, UK), which allowed us to sample all eight compartments sequentially. The gas monitor was calibrated by the Dutch Metrology Institute VSL according to ISO/IEC 17025. Sampling was performed in the middle of the central pen at 0.8m above the slatted floor, which is representative of the height of an adult pig. About one hour was needed to sample all eight compartments sequentially.

Particulate matter was sampled using two Grimm 1.109 spectrometers (Grimm Aerosol Technik GmbH & Co. KG, Ainring, Germany) and two GrayWolf Particle Counters - Handheld 3016IAQ (GrayWolf Sensing Solutions, Shelton, CT, USA). All instruments were equipped with

a temperature and relative humidity sensor to monitor indoor temperature and relative humidity. The manufacturer calibrated both types of instruments. At a certain point in the measuring campaign one of the Grimm spectrometers showed unlikely values; therefore an additional calibration was performed on this spectrometer on March 30th 2012.

The instruments were placed in self-constructed iron cages (1.4 x 0.8 x 0.6m) attached to the slatted floor in the middle of the pens. The air inlet of the instruments was placed at 0.8m above the slatted floor. For the Grimm spectrometers, data was collected with a one-minute interval. To minimise the risk of clogging the pump of the GrayWolf Counters, only one measurement was done every 15 minutes. A rotation scheme was setup whereby the four monitors were rotated twice weekly among the different compartments. To ensure the equivalence of the four PM monitors, bi-weekly side-by-side measurements were performed inside a randomly-chosen compartment for 30 minutes. These potential differences between the PM monitors were calculated on the basis of one-minute interval data. When the differences between the mean PM_{10}, $PM_{2.5}$ or PM_1 concentrations exceeded 10 %, the results from an aberrant monitor were corrected on the basis of these measurements (data not shown). Corrections were only made during the first fattening period after a check-up of both Grimm spectrometers by the manufacturer. One of both spectrometers was considered aberrant and gave consistently lower values. These were corrected based on the side-by-side measurements. The Grimm spectrometers were chosen as the reference monitors to compare the data from the GrayWolf Counters with (Van Ransbeeck *et al.*, 2013b).

Ventilation rates were monitored using free running impellers (Fancom, Panningen, the Netherlands) and logged using a DEWE-43 data logger (DEWETRON Ges.m.b.H, Graz-Grambach, Austria). The outdoor temperature, relative humidity and atmospheric pressure were monitored using the Vantage Pro2 weather station (Davis Instruments Corp., Hayward, CA, USA).

2.1.2.4 DATA ANALYSIS

Erratic or outlier data was excluded for further analysis. Aberrant measurements occurred because (1) the compartment was entered for purposes of animal management (only for PM measurements), (2) the measuring instrument was repositioned (only for PM measurements) or (3) instrument failure (for PM and gas measurements). Data processing

revealed that the ventilation data gathered from August 2011 to April 2012 was erroneous. These data were not used. Different attempts were made to estimate the ventilation rates and validate these with the correct ventilation data between May and June 2012. However, both the use of a carbon dioxide balance (CIGR, 2002) and linear regression including outdoor and indoor temperatures did not result in comparable estimates of the ventilation data. In addition, at certain times no continuous measurements were obtained due to (1) the rotation scheme of the PM monitors and (2) maintenance or repair of the measuring device (e.g. gas data during fattening period 1; see Table 2.3). Table 2.3 provides an overview of the data used for statistical analysis of the four possible combinations of housing system and cleaning protocol. For each combination in each fattening period 2 compartments were taken into account.

Table 2.3. Total number of daily mean measurement results (gasses, PM, outdoor temperature) for the different combinations of cleaning protocol and housing system per fattening period. Each of these combinations comprised 2 compartments.

	Fattening Period	Gases	PM	Outdoor temperature
Wet protocol	1	136	127	268
LAE[1]	2	242	109	266
Wet protocol	1	135	134	268
C[1]	2	241	116	266
Dry protocol	1	128	119	268
LAE[1]	2	230	111	268
Dry protocol	1	128	128	268
C[1]	2	231	106	268

[1] C: conventional compartment; LAE: low-ammonia-emission compartment.

Data analysis was performed on daily averages using SAS/STAT software mixed procedure (SAS 9.3, Cary, NC, USA). A linear mixed model was built using the different gas and PM concentrations as dependent variables. Residuals were assumed to be normally distributed, with a null expectation, based on graphical evaluation (QQ-plot of the residuals). Fattening period was considered a random variable to correct for repeated measurements within a fattening period. Compound symmetry was used as autocorrelation structure. Day in

fattening period (linear and quadratic), outside temperature, cleaning method and housing system were investigated as independent variables in a backward stepwise regression process. In addition, interaction between cleaning method and housing system was tested for all dependent variables. However, this interaction was not statistically significant for any model, and hence it was excluded from the model. Statistical significance was considered for $P < 0.05$.

Figures of the indoor concentrations of the gases and PM for each fattening period were obtained by averaging and grouping the data per two weeks in the period for the four combinations of housing system and cleaning protocol, respectively.

2.1.3 RESULTS AND DISCUSSION

2.1.3.1 EVOLUTION OF THE POLLUTANT CONCENTRATIONS ACROSS THE FATTENING PERIODS

Large differences were observed in the evolution of the pollutant concentrations between the two fattening periods. For this reason, these two periods will be discussed separately. These differences are most probably due to seasonal effects. Fattening periods which started during summer months have generally lower indoor concentrations than fattening periods which started during winter months (Duchaine *et al.*, 2000; Takai *et al.*, 1998). During the first fattening period which started during summer, concentrations of NH_3, CO_2, PM_{10} and $PM_{2.5}$ gradually increased over time (Figure 2.3, Figure 2.4 and Figure 2.7). On the other hand, concentrations of CH_4, N_2O and PM_1 varied only slightly during this fattening period (Figure 2.5 and Figure 2.6). The latter is in accordance with the findings of Mosquera *et al.* (2010). The rather high concentrations for all pollutants at the beginning of fattening period 2 (first two weeks for the wet protocol, third and fourth week for the dry protocol) which started during winter are probably due to weather conditions with low outside temperatures (Figure 2.8). As a consequence of these lower temperatures, the ventilation rate decreased and because no heating devices were present in the compartments, the air inlets were partially closed in order to maintain a suitable indoor temperature. Furthermore, the concentrations at the end of the second fattening period were lower than at the end of the first fattening period. Again, ventilation rate as determined by outside temperature probably played a major role.

The encountered NH_3 indoor concentrations in this study were quite high compared to NH_3 indoor concentrations found in literature. For example, Van Ransbeeck *et al.* (2013a) also investigated indoor concentrations in different Flemish commercial fattening pig barns and found mean NH_3 indoor concentrations between 13.7 and 22.1 ppmv. The results from a large-scale sampling campaign across Northern Europe showed mean NH_3 concentrations between 12.1 and 18.2 ppmv for fattening pigs kept on slatted floors. In this sampling campaign, a maximal NH_3 concentration of 58.6 ppmv was measured (Groot Koerkamp *et al.*, 1998). However, in contrast to these studies, which measured NH_3 concentrations at 1.6m above the slatted floor (Van Ransbeeck *et al.*, 2013a) or at 1.5m and 2.5m (Groot Koerkamp *et al.*, 1998), gas concentrations in this study were measured at 0.8m above the slatted floor. This could partly explain the higher concentrations, found in this study. As already stated above, the low outdoor temperatures, during the first two weeks for the wet protocol and the third and fourth week for the dry protocol, will most certainly have led to lower ventilation rates. Due to the inverse relation between ventilation rate and NH_3 indoor concentrations, this reduced ventilation rate gave rise to higher NH_3 indoor concentrations (Kim *et al.*, 2007).

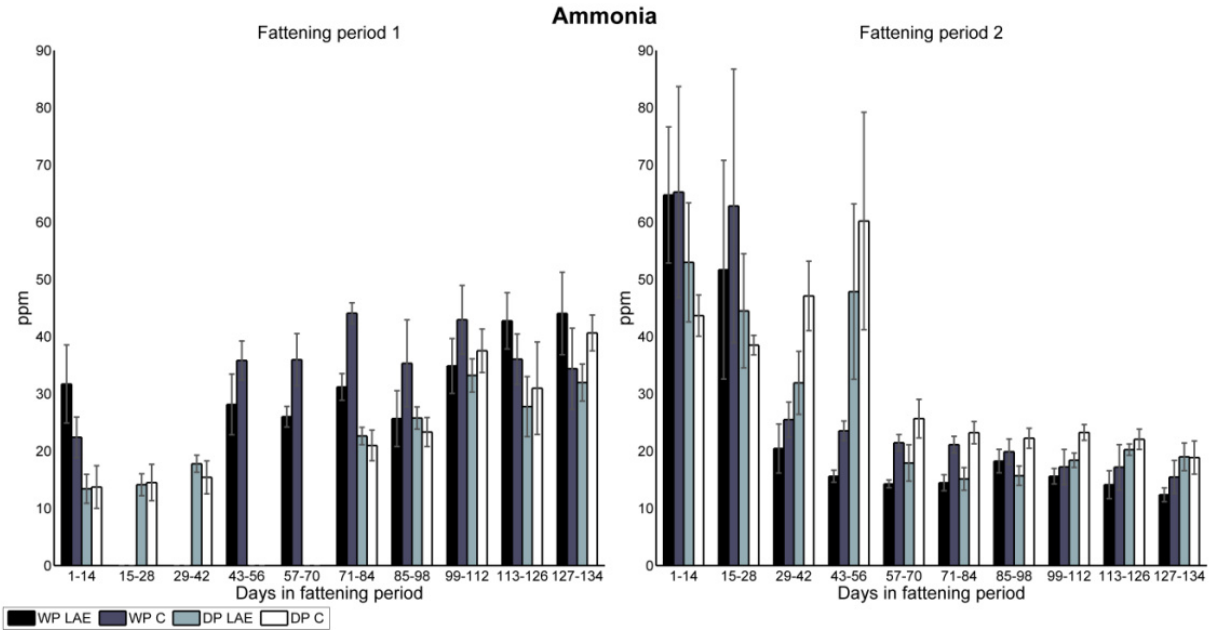

Figure 2.3. Indoor concentrations of NH_3 for each fattening period, averaged and grouped per two weeks for the four combinations of housing system and cleaning protocol (WP: "Wet" Protocol, DP: "Dry" Protocol, LAE: low-ammonia-emission, C: conventional).

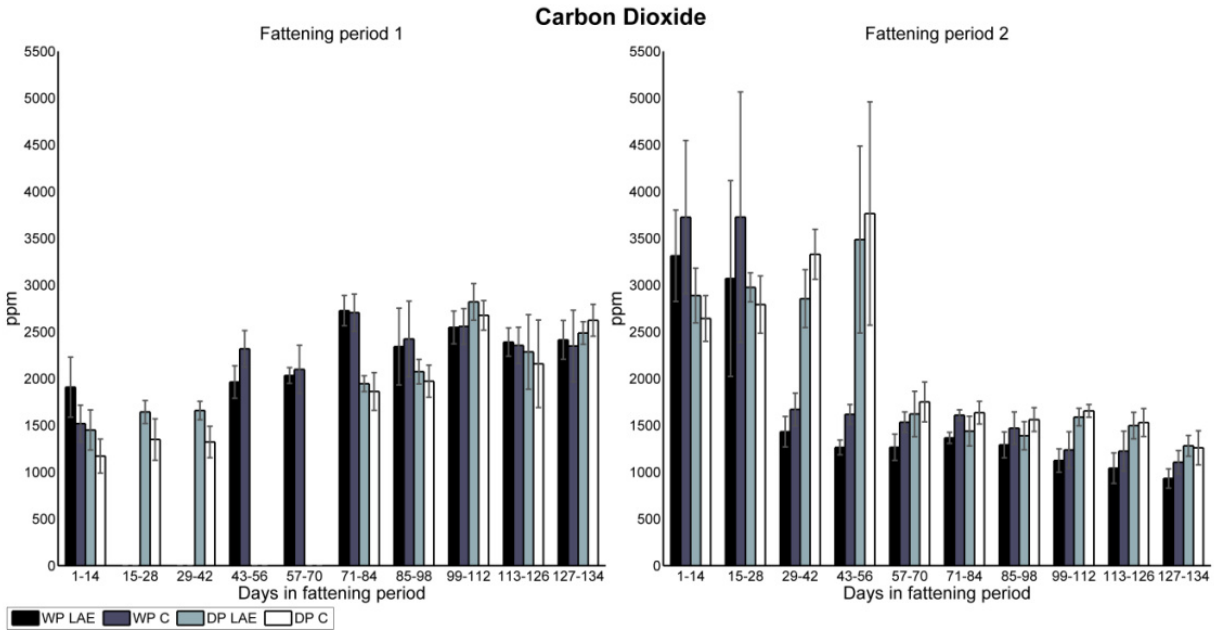

Figure 2.4. Indoor concentrations of CO_2 for each fattening period, averaged and grouped per two weeks for the four combinations of housing system and cleaning protocol (WP: "Wet" Protocol, DP: "Dry" Protocol, LAE: low-ammonia-emission, C: conventional).

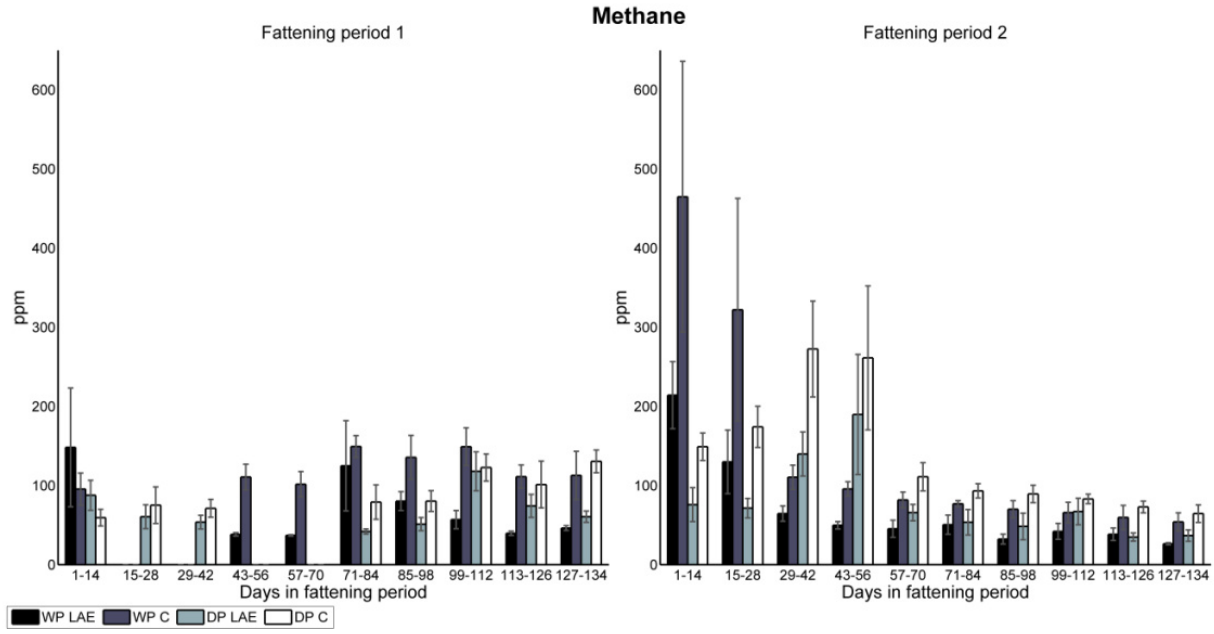

Figure 2.5. Indoor concentrations of CH_4 for each fattening period, averaged and grouped per two weeks for the four combinations of housing system and cleaning protocol (WP: "Wet" Protocol, DP: "Dry" Protocol, LAE: low-ammonia-emission, C: conventional).

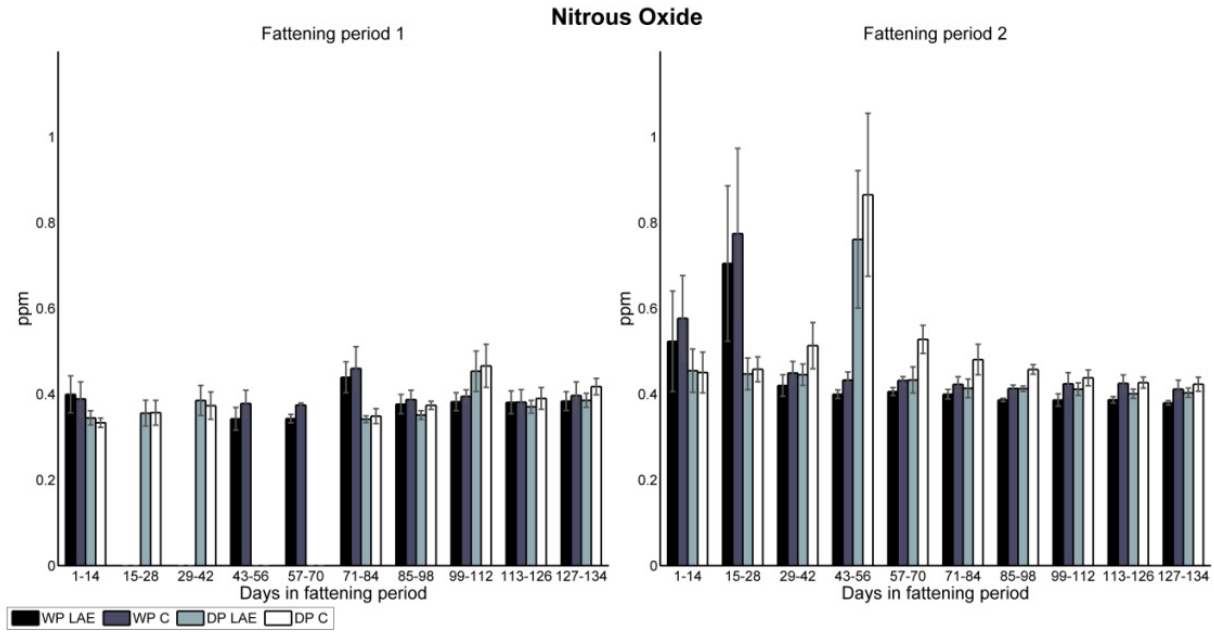

Figure 2.6. Indoor concentrations of N_2O for each fattening period, averaged and grouped per two weeks for the four combinations of housing system and cleaning protocol (WP: "Wet" Protocol, DP: "Dry" Protocol, LAE: low-ammonia-emission, C: conventional).

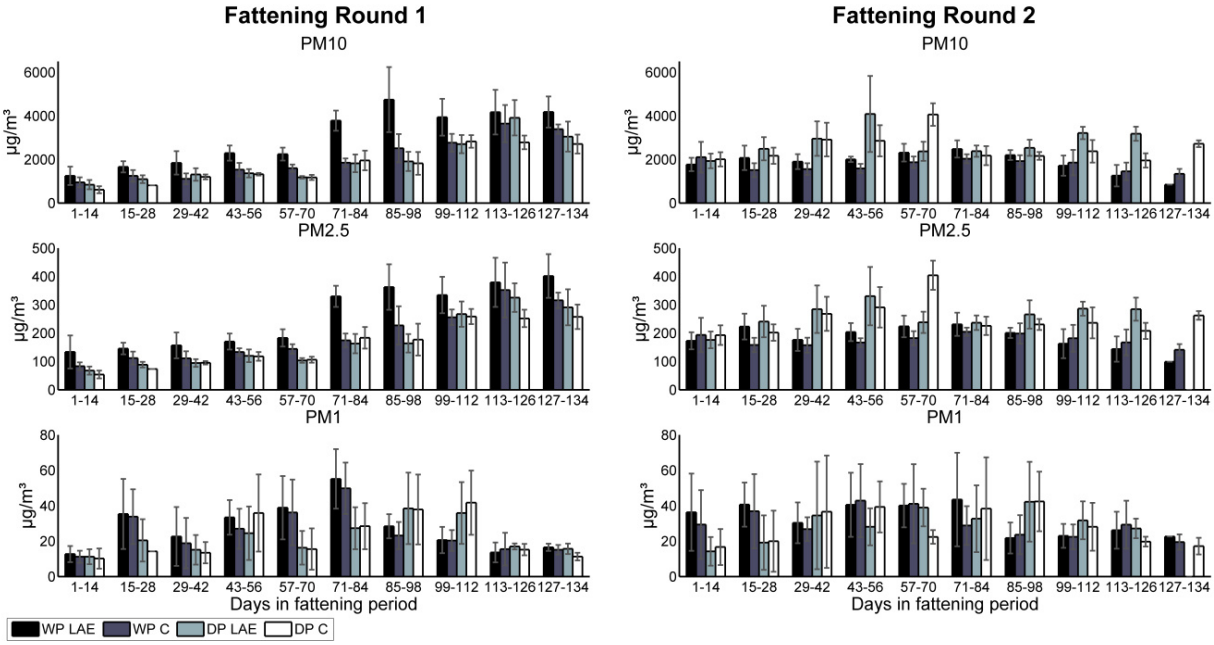

Figure 2.7. Indoor concentrations of PM_{10}, $PM_{2.5}$ and PM_1 for each fattening period, averaged and grouped per two weeks for the four combinations of housing system and cleaning protocol (WP: "Wet" Protocol, DP: "Dry" Protocol, LAE: low-ammonia-emission, C: conventional).

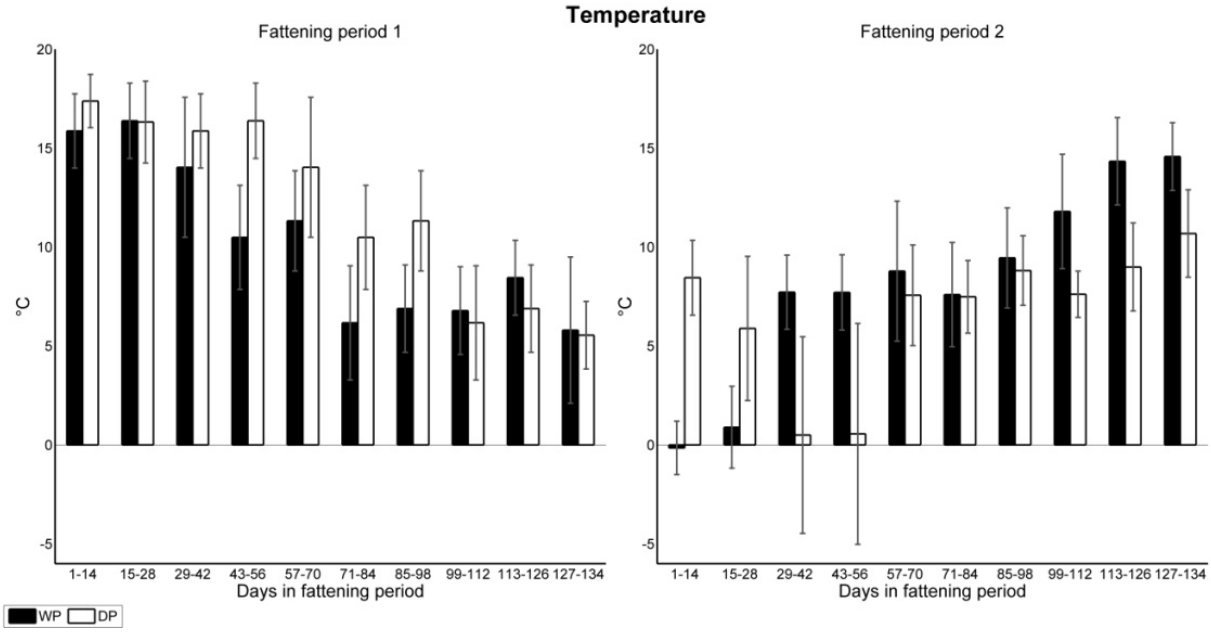

Figure 2.8. Outdoor temperature for each fattening period, averaged and grouped per two weeks for the two cleaning protocols (WP: "Wet" Protocol, DP: "Dry" Protocol).

2.1.3.2 INFLUENCE OF CLEANING PROTOCOL AND HOUSING SYSTEM ON INDOOR POLLUTANT CONCENTRATIONS DURING THE FIRST MONTH AFTER CLEANING

To test for a short-term effect of cleaning protocol or of the housing system, the first 30 days of the fattening periods were analysed separately.

INFLUENCE OF CLEANING PROTOCOL

The use of different cleaning protocols had a significant effect on the N_2O and PM_1 indoor concentrations during the first 30 days. Lower N_2O ($P = 0.01$) and PM1 ($P < 0.0001$) concentrations were observed after performing the "dry" cleaning protocol. A significant effect of the different cleaning protocols on NH_3, CO_2, CH_4, PM_{10} or $PM_{2.5}$ indoor concentrations was not found. The difference in PM_1 concentration (Least Square Mean $(LSM)_{dry}$: 16.4 ± 1.2 µg/m³; LSM_{wet}: 31.2 ± 1.4 µg/m³) between the two cleaning protocols was the opposite of what we expected. There is no clear explanation for this finding. It might be partially caused by the four-week difference in start up date between the compartments where the "dry" protocol was performed and the compartments where the "wet" protocol was performed. Other research also indicated that about 52% of the particles inside animal houses and smaller than 1 µm (= PM_1) is not formed inside the animal house but rather originates from outside (Aarnink *et al.*, 2011). This would mean that the outdoor PM_1 concentration on a given day can have an important effect on the PM_1 indoor concentrations

at that time. Nevertheless, this cannot completely explain the difference in PM_1 concentrations between the two cleaning protocols. The difference in N_2O concentrations (LSM_{dry}: 0.43 ± 0.02 ppmv; LSM_{wet}: 0.51 ± 0.02 ppmv) for the first month should be interpreted with caution. There is a lack of data for the period from day 15 to 28 in fattening period 1 for the wet protocol (Figure 2.6). There was also a low mean outside temperature (Figure 2.8) during these days in fattening period 2, leading to the high concentrations of N_2O (Figure 2.6). However, this lack of data in period 1 and the low outside temperatures for period 2 are also seen for the other gases without a significant effect between the two cleaning protocols.

INFLUENCE OF HOUSING SYSTEM

No effect of the housing system on any of the studied pollutants could be found during the first 30 days (Table 3). However, a trend (P = 0.09) was observed for CH_4 concentrations, with lower concentrations in the low-ammonia-emission housing system (LSM: 105 ± 26 ppmv) as compared to the conventional housing system (LSM: 175 ± 26 ppmv).

Table 2.4. Least square means and standard error with level of statistical significance for cleaning protocol and housing system for the different pollutants and calculated over two fattening periods.

		Cleaning protocol			Housing system		
		Wet protocol	Dry protocol	P-value	LAE[1]	Conventional	P-value
Ammonia (ppmv)	first month	38.9 ± 3.6	33.8 ± 3.5	0.33	36.8 ± 3.6	35.9 ± 3.5	0.85
	entire fattening round	30.6 ± 1.7	26.8 ± 1.7	0.14	27.3 ± 1.7	30.1 ± 1.7	0.26
Carbon dioxide (ppmv)	first month	2389 ± 108	2355 ± 105	0.83	2393 ± 105	2350 ± 105	0.79
	entire fattening round	2016 ± 114	2045 ± 114	0.86	2012 ± 114	2049 ± 114	0.82
Methane (ppmv)	first month	165 ± 27	115 ± 26	0.21	105 ± 26	175 ± 26	0.09
	entire fattening round	101 ± 7	93 ± 7	0.51	74 ± 7	121 ± 7	**0.003**
Nitrous oxide (ppmv)	first month	0.51 ± 0.02	0.43 ± 0.02	**0.01**	0.46 ± 0.02	0.48 ± 0.02	0.41
	entire fattening round	0.43 ± 0.01	0.44 ± 0.01	0.42	0.42 ± 0.01	0.44 ± 0.01	0.12

[1] LAE: low-ammonia-emission compartment.

Table 2.4. Continued.

		Cleaning protocol			Housing system		
		Wet protocol	Dry protocol	P-value	LAE[1]	Conventional	P-value
PM_{10} ($\mu g/m^3$)	first month	1497 ± 164	1448 ± 163	0.83	1536 ± 163	1409 ± 162	0.59
	entire fattening round	2146 ± 159	2215 ± 159	0.76	2393 ± 159	1968 ± 159	0.08
$PM_{2.5}$ ($\mu g/m^3$)	first month	143 ± 15	135 ± 15	0.72	146 ± 15	132 ± 15	0.54
	entire fattening round	201 ± 12	208 ± 12	0.69	219 ± 12	190 ± 12	0.12
PM_1 ($\mu g/m^3$)	first month	31.2 ± 1.4	16.4 ± 1.2	**<0.0001**	24.5 ± 1.3	23.2 ± 1.3	0.5
	entire fattening round	28.7 ± 1.4	26.3 ± 1.4	0.25	27.8 ± 1.4	27.2 ± 1.4	0.75

[1] LAE: low-ammonia-emission compartment.

2.1.3.3 INFLUENCE OF CLEANING PROTOCOL AND HOUSING SYSTEM ON INDOOR POLLUTANT CONCENTRATIONS DURING THE ENTIRE FATTENING PERIOD

In addition to the analysis for the first 30 days, we also analysed the entire fattening period. This allowed us to determine whether the observed differences in PM_1 and N_2O concentrations for the different cleaning protocols were still valid when considering the entire fattening period. It also provided more information about the effect of housing system during an entire fattening period.

INFLUENCE OF CLEANING PROTOCOL

Different cleaning protocols yielded no significant effect on any of the studied pollutants (Table 2.4). A study by Heber *et al.* (1988b) suggested that poor hygiene in piggery buildings, evaluated by the depth of floor litter, is related to increased airborne dust concentrations (Heber *et al.*, 1988b). Other studies compared one form of cleaning with no cleaning at all and found significant differences. For example, Cargill and Banhazi (1998) showed that concentrations of respirable particles and airborne bacteria decreased with 21.1 % and 25.4 %, respectively, after thoroughly cleaning between batches as compared to no cleaning between batches (Cargill & Banhazi, 1998). In a similar study with weaner pigs, a significant elevation of total dust (53.3 %), NH_3 (117 %) and CO_2 (37.9 %) concentrations was found in a 'dirty' indoor environment, with no cleaning and disinfection before the start of the fattening period, versus a clean environment where cleaning and disinfection was performed before the start of the fattening period. However, they detected no differences on the respirable dust, total bacteria and endotoxin concentrations (Lee *et al.*, 2005). In this study, the comparison of two practical protocols did not show any significant difference. This indicates that dry cleaning alone may sufficiently reduce pollutant concentrations and that the addition of extra wet cleaning steps doesn't significantly reduce the concentrations further. Van't Klooster *et al.* (1993) did not find a significant reduction in the indoor PM concentrations when evaluating the effect of weekly vacuum cleaning inside a pig finishing facility. For NH_3, multiple studies have shown that concentrations in piggeries increase with a decrease in pen hygiene (Aarnink *et al.*, 1996; Aarnink *et al.*, 1997; Ni *et al.*, 1999c). Pen hygiene was defined by Aarnink *et al.* (1996) as the surface area of urine-fouled floor. Aarnink *et al.* (1997) however, also included the frequency of urination and defecation on various locations in the pen. Ni *et al.* (1999c) considered the proportion of the solid floor

surface that was covered by a mixture of urine and faeces. The lack of a significant effect on any of the pollutants in the current study may have several causes. The effect of the cleaning protocol might be the strongest during the first four weeks after cleaning, therefore it might be possible that the effect during that period is lost when considering the entire fattening period. This might be the case for the PM_1 and N_2O concentrations, as we observed significant differences when considering the first 30 days, but no significant differences when considering the entire fattening period. Or the effect of the cleaning protocol could be relatively small and might have been overshadowed by differences in ventilation rate for the different compartments. This presumption could not be ascertained because of instrument failure during this period. It is also possible that the tested cleaning protocols did not differ sufficiently from each other. Finally, it is also possible that the four week difference in start up date between compartment with dry and wet cleaning and the decision not to change the treatments between different manure periods, have reduced the power of the experiments to differentiate between the cleaning protocols.

INFLUENCE OF HOUSING SYSTEM

When comparing housing systems, we only found a significant effect (P = 0.003) on the indoor CH_4 concentrations with lower concentrations in the low-ammonia-emission housing system (LSM_{LAE}: 73.60 ± 7.03 ppmv; LSM_C: 120.67 ± 7.03 ppmv). This difference was already seen as a trend when looking at the first 30 days, but was not yet significant. Both the manure pit in the conventional housing system and the water and manure channels in the LAE housing system, were emptied before the start of each fattening period. However, the manure in the conventional housing system was completely collected in the manure pit (if the amount of manure which remained on the slatted floor is not taken into account). In the LAE housing system on the other hand, manure could be removed from the compartment via the overflow present in the manure channel. This could be the reason for the observed lower indoor CH_4 concentrations. One would expect that, as a consequence of the reduced emission surface, the use of a LAE housing system would lead to lower NH_3 indoor concentrations as compared to a conventional housing system. However, this was not observed. One possible explanation is that part of the NH_3 concentration inside the low-ammonia-emission housing system originates from the soiled solid floor and, together with the NH_3 volatilisation from the reduced manure pit, results in the same concentrations as

can be found in the conventional housing system. It has been suggested that an important part of the total NH_3 emission from a pig house originates from the pen floor (Hoeksma *et al.*, 1992). The concentration levels of N_2O are generally very low, allowing small differences to remain unnoticed. Research on the influence of a reduced emission surface on the PM_{10} and $PM_{2.5}$ concentrations is scarce. Mosquera *et al.* (2010) found that animal houses with reduced emission surfaces had 39 % more PM_{10} and 19 % more $PM_{2.5}$ emissions as compared to animal houses with fully slatted floors (Mosquera *et al.*, 2010). In our study, no differences could be detected for PM_{10}, $PM_{2.5}$ or PM_1 indoor concentrations. There is no ready explanation for this finding.

2.1.4 CONCLUSIONS

This study compared the effects of two cleaning protocols and two housing systems on indoor pollutant concentrations at a commercial fattening pig barn. Data analysis showed that the use of a low-ammonia-emission housing system under field conditions in this study does not seem to lower the indoor concentrations of NH_3, CO_2, N_2O or PM. The CH_4 indoor concentrations were reduced, however, in the low-ammonia-emission system. In order to generalise the conclusion about the performance of low-ammonia-emission housing systems under field conditions, measurements on multiple farms are necessary.

In this study and for this farm, with the technical problems of the study in mind, it was not possible to differentiate between the two cleaning protocols as applied between the fattening periods. The use of a more extensive cleaning protocol, with an additional wet cleaning and disinfection step, did not seem to lead to consistently lower indoor concentrations for the studied pollutants. This can also suggest that dry cleaning by itself may already be sufficient to reduce the pollutant concentrations or that the extra steps in the wet cleaning protocol do not result in a better indoor air quality. However, additional experiments on multiple farms will be necessary to validate this hypothesis.

2.2 CORRELATIONS BETWEEN AERIAL POLLUTANTS AND PARTICLE SIZE DISTRIBUTIONS OF PARTICULATE MATTER INSIDE A PIG FATTENING FACILITY[iii]

2.2.1 INTRODUCTION

Inside livestock barns, a wide variety of aerial pollutants can affect indoor air quality (NRC, 2003). The main pollutants are particulate matter (PM), ammonia (NH_3) and the greenhouse gases (GHGs) carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O). High indoor concentrations of one or several of these pollutants can negatively affect human health (Asmar *et al.*, 2001; Von Essen & Donham, 1999; von Essen & Banks, 2009) or the health and productivity of the animals (Busse, 1993; Donham, 1991; Donham, 2000; Urbain *et al.*, 1999). Furthermore, emissions of these pollutants can have adverse effects for the environment and for nearby inhabitants (Flessa *et al.*, 2002; Krupa, 2003; Oehrl *et al.*, 2001; Zhao, 2011).

The gases commonly found in livestock barns are formed through different processes and in various locations throughout the barn. Ammonia and CH_4 are mainly formed from manure and released from either the manure pit (and almost entirely for CH_4) or the floor of the pens (Canh *et al.*, 1997; Zeeman, 1991). Carbon dioxide is mainly released via the pigs' respiration (Ni *et al.*, 1999a), but a significant part also originates from the manure pit (Ni *et al.*, 1999b). Details about the N_2O formation processes inside a livestock barn are still poorly understood (Monteny *et al.*, 2001). Nitrous oxide is assumed to originate completely from the manure present (Philippe, 2013). In contrast to CH_4 production, which requires anaerobic conditions, N_2O production first requires an initial aerobic reaction followed by an anaerobic process (Monteny *et al.*, 2001). Gaseous pollutants are defined by a single molecule, whereas particulate matter is a mixture of many types of particles that differ in size, shape, chemical composition and density (Pedersen *et al.*, 2000). The main sources of PM inside fattening pig facilities are feed, manure and skin. PM can also contain or adsorb micro-organisms, toxins or residues of veterinary products (Cambra-López *et al.*, 2011a; Cambra-López *et al.*, 2011b). Although the formation processes and their locations can differ

[iii] **Adapted from: Ulens, T., Millet, S., Van Weyenberg, S., Van Der Meeren, P., Van Langenhove, H., & Demeyer, P.** (2015). Correlations between aerial pollutants and particle size distributions of particulate matter inside a pig fattening facility. *Submitted.*

widely, it seems plausible that interactions and correlations between the pollutants may exist. However, most of the mitigation strategies developed to minimise the emissions of these pollutants do not take these possible interactions and correlations into account. However, more recent sampling strategies incorporate measurements of several pollutants at the same time in their strategy and evaluate the mitigation strategies for different pollutants (VERA, 2011).

To date, only few studies reported correlations between the emissions of the different pollutants. For example, Philippe *et al.* (2007) found high correlations between NH_3 emissions and CH_4 (R = 0.77) and CO_2 (R = 0.76) emissions for fattening pigs kept on a slatted floor. A high correlation (R = 0.71) between CH_4 emissions and CO_2 emissions was also found. Studies about the correlation of indoor concentrations are even scarcer. To our knowledge, Van Ransbeeck *et al.* (2013a) are the only researchers who reported correlations between the indoor concentrations of all the main aerial pollutants (NH_3, GHGs and PM) inside fattening pig facilities. While it is impossible to distinguish causal relationships from co-evolving patterns, calculating correlations between the different pollutant concentrations can help to generate hypotheses concerning underlying causes for the observed correlations.

To cope with the heterogeneous nature of PM and the associated highly irregular shape and variety in density of the particles, the behaviour of the different particles is commonly described by the aerodynamic diameter (AED). The AED of an irregularly shaped particle is defined as the diameter of a sphere with a standard density (1000 kg/m³) that would have the same settling velocity in air as the particle (Zhang, 2004). A wide range of particles with different AED can be found inside livestock houses (Harry, 1978). Particle size distribution (PSD) analysis may help to describe and understand this heterogeneity in AED. Indeed, the PSD of PM is perhaps the most important physical parameter determining particle behaviour. PSD analysis can also be used to develop mitigation strategies or techniques to identify the particle size ranges that should be removed (Dawson, 1990). The changes associated with growing animals would make it likely that the amount and/or distribution of PM may evolve over time. Similarly, Dawson (1990) suggested that mitigation techniques

may affect PSD. However, to our knowledge, the effect of different mitigation techniques on PSD is not clear at the moment.

Therefore, the aims of the current study were 1) to investigate the correlations between the indoor concentrations of NH_3, GHGs and PM in a commercial fattening pig barn and 2) to perform PSD analysis in order to get an overview of the dominant size ranges in fattening pig facilities. An additional goal was to make a first assessment of the effect of different housing systems and cleaning protocols on PSD.

2.2.2 MATERIALS AND METHODS

2.2.2.1 EXPERIMENTAL DESIGN

The measurements were performed at a commercial fattening pig facility in Diksmuide, Belgium during two fattening periods (August 2011 to June 2012). Two types of housing systems were used: conventional compartments with fully slatted floors (C) and low-ammonia-emission compartments with reduced emission surfaces by means of sloped pit walls and partially slatted floors (LAE). Two cleaning protocols were performed in the different compartments, namely a “dry” (D) and a “wet” (W) protocol. A detailed description of the housing systems and more information about the cleaning protocols can be found in chapter 2.1.

2.2.2.2 MEASURING EQUIPMENT

Indoor gas concentrations (NH_3, CO_2, CH_4 and N_2O) were measured using a photoacoustic gas monitor (Innova 1314, Innova Air Tech Instruments, Santa Clara, CA, United States) connected to a multipoint sampler (CBISS, A1-Envirosciences ltd., Wirral, United Kingdom). Sampling was performed in the middle of the central pen at 0.8m above the slatted floor. This height is representative of the height of an adult pig.

Particulate matter was sampled using two spectrometers (Grimm 1.109, Grimm Aerosol Technik GmbH & Co. KG, Ainring, Germany) and two particle counters (GrayWolf Particle Counters type Handheld 3016IAQ, GrayWolf Sensing Solutions, Shelton, CT, United States). The Grimm spectrometers make use of scattered light photometry inside an optical measuring cell that enables detection of each individual particle. Both the Grimm spectrometers and the GrayWolf Particle Counters use a laser diode light source. Scattered

light is collected and focused onto a photo diode that converts the bursts of light into electrical pulses. The amplitude of the pulses is used as the measure of the particle size.

Van Ransbeeck *et al.* (2013b) revealed that the Grimm spectrometers were equivalent to a reference instrument in accordance with EN 123412 for the measurement of indoor PM_{10} concentrations (Van Ransbeeck *et al.*, 2013b). The spectrometers and particle counters were placed in self-constructed iron cages (1.4 x 0.8 x 0.6m) attached to the slatted floor in the middle of the pens with the air inlet of the instruments at 0.8m above the slatted floor. For the Grimm spectrometers, data were collected with a 1-min interval. To minimise the risk of clogging the pump of the GrayWolf Counters data were collected every 15 minutes during 1 min. The equivalence of the four PM monitors was checked using a bi-weekly 30-min, side-by-side measurement inside a randomly-chosen compartment (chapter 2.1). The Grimm spectrometers were chosen as the reference monitors to correct the data from the GrayWolf Counters (Van Ransbeeck *et al.*, 2013b).

More information about the measuring setup and characteristics of the different instruments can be found in chapter 2.1.

2.2.2.3 DATA ANALYSIS

Raw data from the gas and PM measurements were converted into hourly means. For PM measurements, data from periods with human interference in the compartment (e.g. entering of the compartment for purposes of animal management or repositioning of the measuring instrument) were deleted from the dataset. For both gas and PM measurements, data from periods with instrument failure were excluded from the dataset.

CORRELATIONS

Comparison of the data from the different housing systems and cleaning protocols revealed no significant differences in most gas and PM concentrations (chapter 2.1). Therefore, all correlations were calculated based on the full dataset. As mentioned previously (chapter 2.1), ventilation measurements were performed. However, instrument failures rendered these measurements unreliable and they were not used. Therefore, it was not possible to take ventilation effects into account.

The full dataset contained approximately 30 000 hourly means of gas concentrations and 18 000 hourly means of PM concentrations. Within the hourly means of PM concentrations, both data from the Grimm spectrometers (9 000 hourly means) and from the GrayWolf Particle Counters (9 000 hourly means) were present.

Correlations between the different gas concentrations and PM fractions were calculated using SPSS Statistics 21.0 (SPSS Inc., Chicago, IL, USA). Relations between the different pollutants were checked for non-linearity by graphical presentation. Using the Kolmogorov-Smirnov test and based on visual inspection of QQ-plots, it was shown that the data were not normally distributed ($P < 0.05$). Therefore, Spearman's rank correlation coefficients were calculated. All statistical tests were performed at 0.05 significance level.

PARTICLE SIZE DISTRIBUTION (PSD)

For the PSD analysis, only the data from the Grimm 1.109 spectrometers were used. These spectrometers are capable of counting the number of particles in 31 size ranges with the following lower limits (µm): 0.25, 0.28, 0.30, 0.35, 0.40, 0.45, 0.50, 0.58, 0.65, 0.70, 0.80, 1.0, 1.3, 1.6, 2.0, 2.5, 3.0, 3.5, 4.0, 5.0, 6.5, 7.5, 8.5, 10.0, 12.5, 15.0, 17.5, 20, 25, 30 and 32. The 32 µm size range was not included in the analysis due to uncertainties about its upper limit. The Grimm 1.109 spectrometers were used in two conventional and two low-ammonia-emission compartments. Both cleaning protocols were used in each type of housing system.

To represent the PSD, the count median diameter (CMD) and the mass median diameter (MMD), together with their respective geometric standard deviation (GSD) were calculated, using equations adapted from Zhang (2004). Both diameters were calculated based upon the number of particles for the 30 size ranges of the Grimm spectrometers. For purposes of calculation we assumed that all particles were spherical and had the same density.

The CMD (in µm) is defined as the geometric mean diameter of the number–weighted PSD. For a lognormal distribution, the geometric mean equals the median. The CMD was calculated using Eq. 2.1.

$$\mathrm{CMD} = \exp\left[\frac{\sum F_i \ln d_i}{\sum F_i}\right] \qquad \text{Eq. 2.1}$$

Where:

F_i: number of particles per m³ in size range *i*

$\sum F_i$: total number of particles per m³

d_i: mean diameter of the lower and upper limit of size range *i*, in µm

The geometric standard deviation of the CMD (CGSD) is a dimensionless quantity with a value greater than 1.0 and is a measure for the width of the number-weighted aerodynamic particle size distribution. The CGSD for the number-weighted PSD was calculated using Eq. 2.2.

$$\mathrm{CGSD} = \exp\left[\frac{\sum F_i(\ln d_i - \ln CMD)^2}{\sum F_i}\right]^{1/2} \quad \text{Eq. 2.2}$$

The MMD (in µm) is defined as the diameter for which half the total mass of particles is larger and half is smaller than this size. The MMD was calculated using Eq. 2.3.

$$\mathrm{MMD} = \exp\left[\frac{\sum F_i d_i^3 \ln d_i}{\sum F_i d_i^3}\right] \quad \text{Eq. 2.3}$$

The geometric standard deviation of the MMD (MGSD) is also a dimensionless quantity with a value greater than 1.0 and is a measure for the width of the mass-weighted aerodynamic particle size distribution. The MGSD was calculated using Eq. 2.4.

$$\mathrm{MGSD} = \exp\left[\frac{\sum F_i\, d_{pi}^3(\ln d_i - \ln MMD)^2}{\sum F_i d_i^3}\right]^{1/2} \quad \text{Eq. 2.4}$$

CMD, MMD and their respective GSD were calculated on hourly data from the two consecutive fattening periods in the four compartments. These calculations were automated in R3.0.1 (R Core Team, 2013).

2.2.3 RESULTS

2.2.3.1 CORRELATIONS BETWEEN DIFFERENT POLLUTANTS

High correlations (R > 0.8) between NH_3 and CO_2 were found for the entire dataset (Table 2.5). High correlations (R > 0.6) were also found between NH_3 and CH_4 and between CO_2 and CH_4. Lower correlations (R <= 0.5) were found between NH_3 and N_2O, between N_2O and CO_2 and between N_2O and CH_4.

Very high correlations (R > 0.95) were found between PM_{10} and $PM_{2.5}$ when analysing data from both the Grimm spectrometers and GrayWolf Particle Counters (Table 2.6). When analysing the full dataset, lower correlations (R < 0.3) were found between PM_{10} and PM_1 and between $PM_{2.5}$ and PM_1. However, higher correlations (R > 0.5) between PM_{10} and PM_1 and between $PM_{2.5}$ and PM_1 were found when analysing only the data from the Grimm spectrometers, but not from the GrayWolf Particle Counters (Table 2.6).

Table 2.5. Spearman's rank correlation coefficients between the different gases.

		Spearman's rank correlation coefficient
Ammonia	Carbon Dioxide	0.87[a]
Ammonia	Methane	0.70[a]
Ammonia	Nitrous Oxide	0.40[a]
Carbon Dioxide	Methane	0.63[a]
Carbon Dioxide	Nitrous Oxide	0.31[a]
Methane	Nitrous Oxide	0.48[a]

[a] Significant at P < 0.001.

Table 2.6. Spearman's rank correlation coefficients between the different PM fractions based upon the full PM dataset, the Grimm dataset and the GrayWolf dataset.

		Spearman's rank correlation coefficient		
		Full dataset	Grimm	GrayWolf
PM_{10}	$PM_{2.5}$	0.97[a]	0.95[a]	0.99[a]
PM_{10}	PM_1	0.24[a]	0.59[a]	0.07[a]
$PM_{2.5}$	PM_1	0.27[a]	0.69[a]	0.09[a]

[a] Significant at P < 0.001.

Positive correlations were found for the full dataset between the gases NH_3 and CO_2 and PM fractions PM_{10} and $PM_{2.5}$ (Table 2.7), whereas negative correlations were found between the gases and PM_1. No significant correlations were found between CH_4 and PM_{10}, $PM_{2.5}$ and PM_1. Correlations between gases and PM_1, both calculated from the Grimm dataset or from the GrayWolf dataset, were low (R < 0.4)(Table 2.7). Furthermore, all other measured correlations between the different gases and PM fractions were low or non-significant for the full dataset (Table 2.7).

Table 2.7. Spearman's rank correlation coefficients between the different gases and the PM fractions.

		Spearman's rank correlation coefficient		
		Full dataset	Grimm	GrayWolf
Ammonia	PM_{10}	0.24[a]	0.26[a]	0.18[a]
Ammonia	$PM_{2.5}$	0.23[a]	0.30[a]	0.12[a]
Ammonia	PM_1	-0.04[a]	0.17[a]	-0.17[a]
Carbon Dioxide	PM_{10}	0.33[a]	0.33[a]	0.28[a]
Carbon Dioxide	$PM_{2.5}$	0.30[a]	0.34[a]	0.21[a]
Carbon Dioxide	PM_1	-0.02[b]	0.21[a]	-0.14[a]
Methane	PM_{10}	-0.006	-0.04[b]	0.03[b]
Methane	$PM_{2.5}$	-0.017	0.04[b]	-0.015
Methane	PM_1	0.011	0.02	-0.004
Nitrous oxide	PM_{10}	0.11[a]	0.08[a]	0.12[a]
Nitrous oxide	$PM_{2.5}$	0.17[a]	0.20[a]	0.12[a]
Nitrous oxide	PM_1	0.22[a]	0.20[a]	0.27[a]

[a] Significant at P < 0.001.

[b] Significant at P < 0.05.

2.2.3.2 PARTICLE SIZE DISTRIBUTION

Examples of a differential number-weighted and differential mass-weighted particle size distribution are given in Figure 2.9. Few differences were found between the mean CMD values for the different compartments and fattening periods, with values ranging from 0.43 to 0.49 µm (Figure 2.10). Also the mean CGSD values did not differ largely from one another with values between 2.01 and 2.29 (data not shown). Mean MMD values ranged from 10.73

to 12.18 µm (Figure 2.11) with MGSD values ranging from 1.88 to 1.97 (data not shown). More outliers were found for the MMD values from the low-ammonia-emission housing system. In general, for the four combinations of housing system and cleaning protocol, very similar PSDs were found.

For neither the CMD values nor the MMD values could a daily pattern in the evolution of these values be found (Figure 2.12). No clear evolution of these values through the fattening period could be found either (Figure 2.13).

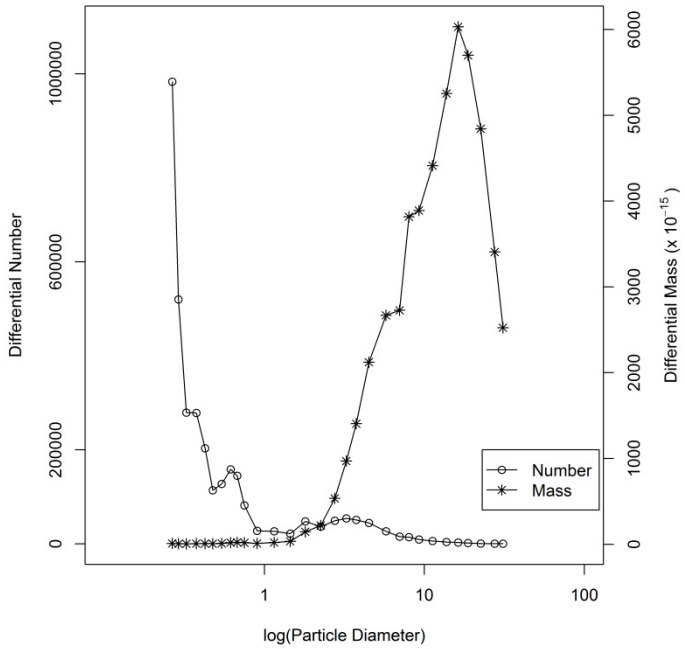

Figure 2.9. Example of a differential number-weighted (circles) and mass-weighted (asterisk) PSD (data from 10 AM till 11 AM for the 118th day in fattening round 1 in the conventional compartment where dry cleaning was performed)

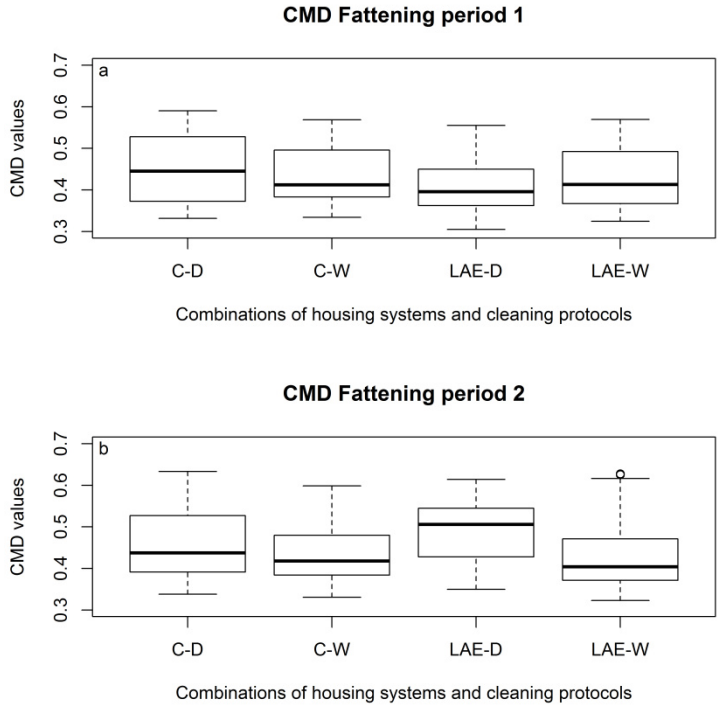

Figure 2.10. Overview of the CMD values for the different combination of housing systems and cleaning protocols in both fattening periods (C: conventional compartment; LAE: low-ammonia-emission compartment; D: "dry" cleaning protocol; W: "wet" cleaning protocol).

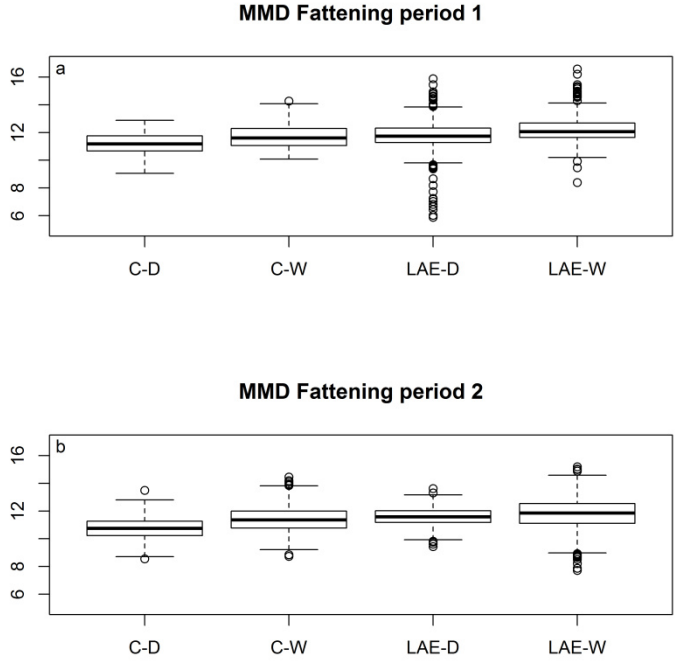

Figure 2.11. Overview of the MMD values for the different combination of housing systems and cleaning protocols in both fattening periods (C: conventional compartment; LAE: low-ammonia-emission compartment; D: "dry" cleaning protocol; W: "wet" cleaning protocol).

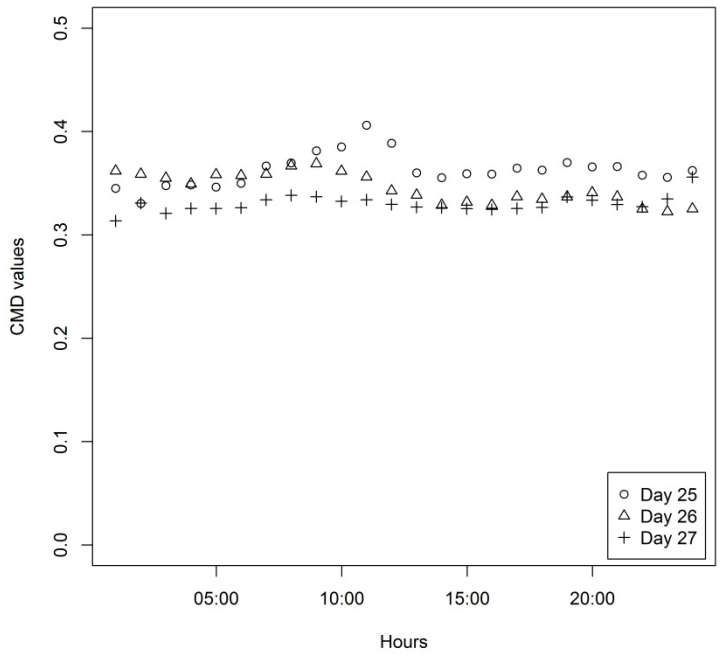

Figure 2.12. Hourly CMD values from 3 consecutive days in the first fattening period for the low-ammonia-emission compartment where the dry cleaning protocol was executed. No clear diurnal pattern could be observed.

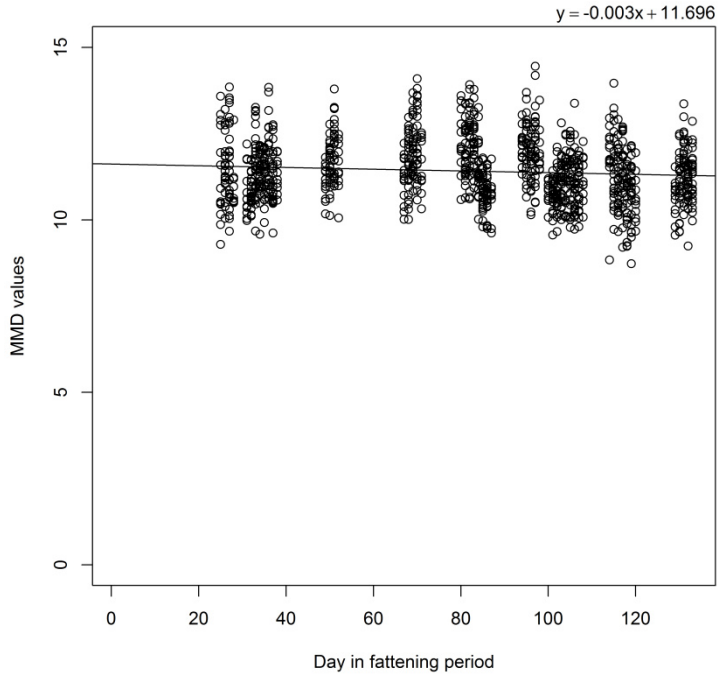

Figure 2.13. Hourly MMD values in the second fattening period for the conventional compartment where the wet cleaning protocol was executed. The trend line and the equation of the trend line, with an almost horizontal slope, reflect the lack of evolution of the MMD values through the fattening period.

2.2.4 DISCUSSION

2.2.4.1 CORRELATIONS

Because the results were obtained from an observational study, it is not possible to establish causal relationships between the different gases and PM fractions. Nevertheless, the calculated correlations presented in this study are based upon a large number of hourly means and can hold valuable information about potential interactions/relations between the different pollutants. However, additional research will be needed to confirm these relationships and to explore the reasons for correlation.

When calculating the correlations, the autocorrelation between the hourly measured gas and PM concentrations was not taken into account. Although this autocorrelation will certainly have an effect on the calculated correlations, this effect will be mainly on the number of degrees of freedom. If corrections would be made to take into account the autocorrelations, this would reduce the number of degrees of freedom. Furthermore, since most of the correlations found in this study, were very significant ($P < 0.001$), it is unlikely that they would not be significant anymore after correction for autocorrelation (Bartlett, 1946; Bayley & Hammersley, 1946).

As mentioned in the introduction, the gases found in livestock buildings have different formation processes and are formed in various locations within the building. Nevertheless, high correlations were found between the indoor concentrations of NH_3, CH_4 and CO_2. These high correlations were also found by Van Ransbeeck *et al.* (2013a). In that study, correlations were calculated based upon two fattening periods in 7 barns on 3 farms. Instead of continual measurements, they performed on average one 48-hour measurement each month. Furthermore, in that study the indoor gas and PM concentrations were measured at 1.6m (human height) (Van Ransbeeck *et al.*, 2013a).

While an influence of the ventilation rate on the indoor gas concentrations (and the obtained correlations) cannot be excluded, a similar increase of the respective gas concentrations throughout a fattening period might be part of the reason for the observed highly correlated indoor gas concentrations. Indeed, the CO_2 exhalation rates increase through a fattening period as a consequence of increasing pig weight (Ni *et al.*, 1999a), but NH_3 concentrations also tend to increase over a fattening period due to the increased feed

consumption and consequently higher manure production as the pigs become older and heavier (Aarnink *et al.*, 1995; Philippe *et al.*, 2007). Volatilisation of CH_4 from manure is affected by the amount of excreted manure (Jarret *et al.*, 2011), which is also likely to increase throughout a fattening round.

High correlations between the emissions of NH_3 and CO_2 and between the emissions of NH_3 and CH_4 are observed in other studies (Philippe *et al.*, 2007). However, one should be careful when comparing correlations based upon indoor concentrations or based upon emission data, especially when the indoor concentrations are not measured at the air outlet.

Correlations between indoor concentrations of N_2O with indoor concentrations of NH_3, CO_2 and CH_4 were low in this study. This is in contrast with Van Ransbeeck *et al.* (2013a), who found higher correlations between N_2O and NH_3 (R = 0.57), CO_2 (R = 0.65) or CH_4 (R = 0.59). Unlike NH_3, CO_2 and CH_4, the indoor concentrations of N_2O remain more constant during a fattening period (Mosquera *et al.*, 2010; chapter 2.1). This could be a possible explanation for the lower correlations found between the indoor concentrations of N_2O and the other gases. Besides, the low correlation between NH_3 and N_2O may directly result from the formation processes of both gases. Nitrous oxide production in manure starts with the oxidation of ammonium into nitrite and is then converted into nitrate. Denitrification converts the nitrate into molecular nitrogen (N_2), with N_2O as an intermediate compound (Kebreab *et al.*, 2006). It has been shown that higher N_2O losses are correlated with lower ammonium concentrations (Brown *et al.*, 2000). If NH_3 volatises from the manure, less ammonium will be available in the manure to be converted to N_2O.

Indoor concentrations of PM_{10} and $PM_{2.5}$ correlated well in the current study. The observed high correlation between PM_{10} and $PM_{2.5}$ indoor concentrations was also found by Van Ransbeeck *et al.* (2013a) inside livestock buildings and by Marcazzan *et al.* (2001) in ambient air (R = 0.97 in winter, R = 0.93 in summer). This can partially be explained by the fact that $PM_{2.5}$ is a substantial part of PM_{10}. Nevertheless, in the current study the mean ratio PM_{10}:$PM_{2.5}$ was about 10:1, while Marcazzan *et al.* (2001) found a ratio of 3:2 in ambient air. Most of the PM inside livestock buildings is primary in origin and can mainly be found in the coarse (PM_{10}-$PM_{2.5}$) fraction. This is especially the case for PM originating from feed, animal hair and skin as well as manure (Cambra-López *et al.*, 2011a). Particles in the fine ($PM_{2.5}$)

fraction are mostly formed through chemical reactions between gases and particles. These secondary processes occur to a lesser extent inside livestock buildings and part of the mechanically generated particles can fall into the $PM_{2.5}$ size range (Cambra-López *et al.*, 2010).

Van Ransbeeck *et al.* (2013a) found a high correlation between $PM_{2.5}$ and PM_1 indoor concentrations (R = 0.77) and a lower correlation between PM_{10} and PM_1 indoor concentrations (R = 0.46). This corresponds quite well with our observed correlations as calculated on the basis of the data from the Grimm spectrometers, which is the same type of instrument as used by Van Ransbeeck *et al.* (2013a). However, when using the GrayWolf Particle Counters, correlations between both fractions were much lower in our study. This indicates that the observed correlations with PM_1 are dependent upon the measuring instrument used. However, both instruments offer a counting efficiency of 50 % at AED of 0.3 µm and of 100 % for all particles larger than 0.45 µm (manufacturer's website; Schmoll *et al.*, 2010). The relative humidity inside the barn can also play an important role. At high relative humidity, water molecules risk of being recognised as particles by the optical instrumentation which can falsify the measurements. To overcome this problem, the Grimm spectrometers are equipped with an air mixing device which can add particle-free dry air to the sample airflow. This system is activated when the relative humidity exceeds 85 % (manual Grimm spectrometer). The GrayWolf Particle Counters however are not equipped with such a device and therefore do not correct for high relative humidity.

While indoor concentrations of PM_{10}, $PM_{2.5}$ and PM_1 changed throughout the fattening periods, the lack of relevant correlations between the gases and the PM fractions suggest a different behaviour of PM or gas particles in the air. This is in agreement with the findings of Van Ransbeeck *et al.* (2013a).

2.2.4.2 Particle Size Distribution

The mean CMD values found in the current study (ranging from 0.43 to 0.49 µm) are similar to the value (0.40 µm) found by Lai *et al.* (2012) who analysed the PSD in different pig buildings using identical Grimm spectrometers. The mean MMD values found in the current study (ranging from 10.73 to 12.18 µm) correspond well with values found by Maghirang *et al.* (1997) in a pig nursery (ranging from 10 to 19 µm) using a cascade impactor. Heber *et al.*

(1988a) found MMD values between 3.51 and 5.14 µm on dust samples from commercial swine finishing buildings. However, they used a resistive-pulse particle analyser with a very narrow dynamic range (1.76 to 7.01 µm) (Heber *et al.*, 1988a). Lee *et al.* (2008) captured dust particles on filters from pig finishing, farrowing and gestation buildings and analysed the PSD with four types of instruments. For pig finishing buildings, depending on the instrument, Lee *et al.* (2008) found MMD values between 10.7 and 20.7 µm. The GSD values found in the current study for the number- and mass-weighted distribution were all larger than 1.22, indicating that the aerosols in all compartments were polydisperse.

Despite the different housing systems and cleaning protocols observed in this study, CMD and MMD values showed no great differences. However, as reported previously (chapter 2.1), indoor mass concentrations of PM_{10}, $PM_{2.5}$ and PM_1 changed throughout the fattening periods. Furthermore, the lack of a clear pattern over a day or over a fattening period is in contrast with the observed diurnal pattern and day to day pattern during a fattening period found for PM_{10}, $PM_{2.5}$ and PM_1 concentrations (Van Ransbeeck *et al.*, 2012). This indicates that, although the total mass of particles (PM concentrations) changed significantly (during a day and during a fattening period) inside the barn, the CMD and MMD values remained about the same.

It is known that besides PM concentrations and density, the size of the particles is an important parameter for the development and operation of reduction strategies or techniques (Zhang, 2004). In the current study, the lack of differences in CMD and MMD values between the different housing systems and cleaning protocols indicates that reduction techniques will probably need to target the same particle size ranges regardless of housing system and cleaning protocol. However, extended research in other livestock buildings will be necessary to verify that CMD and MMD values are in the same range as found here. Furthermore, when applying existing reduction techniques (e.g. electrostatic precipitators, dry filters or wet scrubbers) in the present livestock buildings, the same set-up settings could be used to tackle the most important particle size ranges.

2.2.5 CONCLUSIONS

The results from the present study showed high correlations between the indoor concentrations of NH_3, CO_2 and CH_4. This was possibly related with co-evolving patterns due to ventilation rate or a similar increase for the respective gas concentrations throughout a fattening period. None of the indoor concentrations of these gases correlated well with the indoor concentrations of N_2O. PM_{10} and $PM_{2.5}$ indoor correlations also correlated well, but no high correlations were found between these PM indoor concentrations and the gas concentrations. No differences in PSD could be found between different housing systems or cleaning protocols. Further research will be needed to confirm the findings and possible explanations from this study.

2.3 EFFECT OF GRINDING INTENSITY AND PELLETING OF THE DIET ON INDOOR PARTICULATE MATTER CONCENTRATIONS AND GROWTH PERFORMANCE OF WEANLING PIGS[iv]

2.3.1 INTRODUCTION

The presence of particulate matter in ambient air affects air quality. A significant part of PM originates from agriculture. Inside pig barns, the main sources of PM are manure, skin parts and feed (Aarnink *et al.*, 1999; Cambra-López *et al.*, 2011b; Heber *et al.*, 1988a). Indoor PM concentrations generated from feed depend on the feed composition, the type of feed, the pelleting process and the delivery method (Bundy & Hazen, 1975). Despite the current major concerns about PM, information on feed based mitigation strategies to reduce PM is limited to a few studies, all older than a decade. Feeding pellets instead of meal diets has been shown to lower PM production or indoor PM concentrations (Bundy & Hazen, 1975; Guingand, 1999; Li *et al.*, 1993). However, in the study by Bundy and Hazen (1975) this was only true for floor feeding, but not for feeding with self-feeders. Zeitler *et al.* (1987) only found a significant reduction in PM with a diameter smaller than 5 µm in winter when feeding pellets instead of meal to the pigs. In summer, no differences in indoor PM concentrations could be found when feeding meal or pellets to the pigs (Zeitler *et al.*, 1987). It is unclear to what extent grinding intensity affects PM concentration. Also, whereas both pelleting and grinding intensity have been shown to affect feed efficiency (Healy *et al.*, 1994; Jensen & Becker, 1965), to our knowledge, no simultaneous study has been conducted on the influence of particle size and form of the feed on the growth performance of weaned piglets and the PM concentrations inside pig nurseries. Therefore, in the present experiment, a trial was designed to investigate the interaction between particle size and feed form on indoor PM concentrations and performance of pigs.

2.3.2 MATERIALS AND METHODS

All animal-based procedures followed Belgian and EU legislation (Council Directive 86/609/EEC). No procedures required approval from the local ethics committee.

[iv] **Adapted from: Ulens, T., Demeyer, P., Ampe, B., Van Langenhove, H., & Millet, S.** (2015). Effect of grinding intensity and pelleting of the diet on indoor particulate matter concentrations and growth performance of weanling pigs. *Accepted in Journal of Animal Science.*

2.3.2.1 HOUSING AND ANIMALS

The experiment, conducted at the Institute of Agricultural and Fisheries Research (ILVO) (Melle, Belgium), was divided into 4 weaning rounds, beginning on 18 July 2013 and ending 14 November 2013. A total of 576 weanling pigs (Piétrain boar x RA-SE genetics hybrid sow, Ra-Se genetics holding N.V., Lokeren, Belgium) were used. In each weaning round, 144 pigs were divided over 4 compartments (Figure 2.14, 6 pens of 6 animals per compartment). The pigs were blocked according to sex (2 classes) and body weight (BW) (3 classes), leading to 1 weight class and sex combination per compartment. The compartment was considered the experimental unit. After evenly distributing the piglets over the compartments, each compartment was assigned to 1 of the 4 treatments: finely ground meal, pelleted finely ground feed, coarsely ground meal or pelleted coarsely ground feed. This was repeated 4 times, resulting in 4 replicates per treatment. No treatment was assigned twice to the same compartment in the different weaning rounds. One compartment (6.3 m x 3.6 m) consisted of 8 identical pens (2.16 m x 0.88 m), 4 on each side, with a central 0.35-m exhaust fan in the wall. Per compartment, 2 pens were left empty. The measuring instruments were placed in one of these empty pens. Each pen had a fully slatted plastic floor, 2 nipple drinkers and 1 self-feeder (a four-space feeding trough). Feed was poured manually into the self-feeders. Feed was added (approx. 25 kg) when the feeders were almost empty.

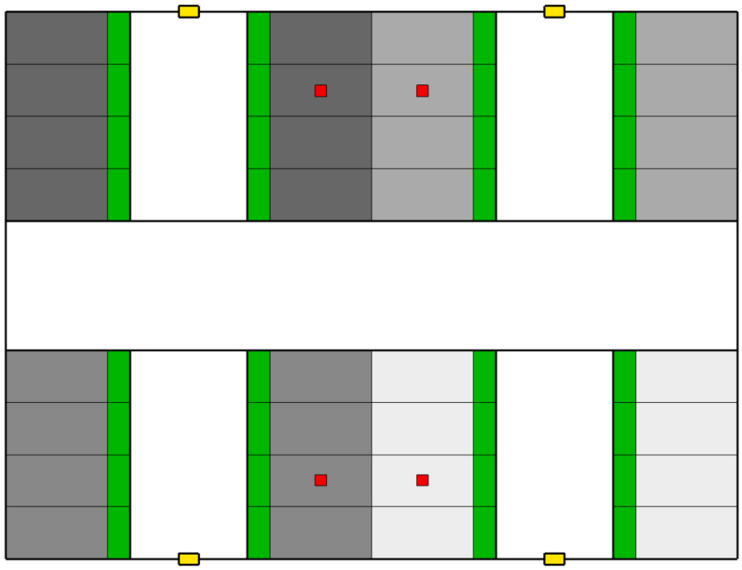

Figure 2.14. Overview of the four compartments: different shades of grey represent different compartments with their pens; the green rectangles represent the self-feeders; the yellow rectangles represent the exhaust fans and the red squares represent the sampling points.

2.3.2.2 DIET COMPOSITION

The ingredient and nutrient content of the feed were exactly the same in all 4 treatments (Table 2.8). The feed was processed using a hammer mill (Promill Type A36, Promill-Stolz S.A., Serville, France) provided with a 1.5 mm (feed A & B) or 6 mm (feed C & D) screen depending on treatment. Corn, wheat, barley, full fat soybeans and soybean meal were ground before mixing. Other ingredients were bought grounded or were liquids (Table 2.8). Per batch, half of the feed was kept as meal (feed B & D) and the other half was pelleted (feed A & C) using cold pelleting equipment (Promill Type A36, Promill-Stolz S.A., Serville, France; die characteristics: 6.0 mm hole size, 60.0 mm wall thickness, 10 rows, 124 holes per row, 490.0 mm flange diameter, 359.0 mm inside diameter, 155.0 mm width, and 78.0 mm track width), leading to pellets with 6 mm diameter. The feeds were provided *ad libitum*.

Table 2.8. Ingredient and nutrient[1] composition of the experimental diet (as-fed).

Item	Content
Ingredient, g/kg	
Maize[2]	254.6
Barley[2]	250.0
Wheat[2]	150.0
Toasted full-fat soybean[2]	99.1
Soybean meal[2] (49% CP)	80.0
Premix[3]	60.0
Beet molasses	30.0
Wheat gluten feed (77% CP)	20.0
Potato protein	20.0
Nutrisure[4]	10.0
L-Lys HCL	6.2
Monocalcium phosphate	5.3
Limestone	3.7
NaCl	2.9
L-Thr, 98%	2.4
DL-Met, 99%	2.4
L-Val , 96.5%	1.2
Soy oil	1.1
L-Trp, 98%	0.9
Phytase (5000 IU/g)	0.1

[1] Values were calculated according to CVB, 2007 (CVB, 2007).

[2] Ingredient was ground before mixing.

[3] The premix contained 80% dairy products and 20% vitamin and mineral premix, providing the following quantities of vitamins and minerals per kilogram of diet: vitamin A, 15,000 IU; vitamin D3, 2,000 IU; vitamin E, 100 mg; vitamin K, 2 mg; vitamin B1, 2.5 mg; vitamin B2, 7.5 mg; vitamin B5, 20 mg; vitamin B6, 5 mg; vitamin B12, 0.04 mg; vitamin C, 100 mg; niacin, 30 mg; choline, 324 mg; folic acid, 3 mg; biotin, 0.15 mg; Ca, 516 mg; P, 419 mg; Mg, 165 mg; Na, 353 mg; Cl, 1,375 mg; K, 1,227 mg; S, 234 mg; Fe, 100 mg; Cu, 160 mg; Mn, 60 mg; Zn, 100 mg; I, 2 mg; Se, 0.4 mg.

[4] A mixture of calcium salts of the following organic acids: lactic acid, formic acid, citric acid monohydrate, orthophosphoric acid and propionic acid (DSM Nutritional Products, Kaiseraugst, Switzerland).

Table 2.8. Continued.

Item	Content
Analysed and calculated composition	
Net energy[1], kcal/kg	2340.7
Dry matter[5], g/kg	888.0
Crude ash[6], g/kg	44.3
Crude fiber[6], g/kg	31.7
Crude protein[6], g/kg	185.0
Ether extract[6], g/kg	47.4
Calcium[1], g/kg	5.6
Phosphorus[1], g/kg	5.2
Digestible phosphorus[1], g/kg	3.5
SID[7] lysine[1], g/kg	11.9
SID methionine+cysteine[1], g/kg	7.5
SID threonine[1], g/kg	7.7
SID tryptophan[1], g/kg	2.6
SID leucine[1], g/kg	12.1
SID valine[1], g/kg	8.3
SID isoleucine[1], g/kg	6.2

[1] Values were calculated according to CVB, 2007 (CVB, 2007).

[5] A mean value for the 4 diets is shown. The individual DM values were 885 g/kg (finely ground meal diet), 881.3 g/kg (coarsely ground meal diet), 892.5 g/kg (finely ground pelleted diet) and 893 g/kg (coarsely ground pelleted diet).

[6] A mean value for the 4 diets is shown.

[7] SID: standardised ileal digestible.

2.3.2.3 FEED CHARACTERISTICS

To determine nutrient composition, separate feed samples from all 4 diets were ground through a 1-mm screen (Brabender Wiley, Rheotec, Maarkedal, Belgium). Moisture was determined by drying at 103 °C (EC, 1971). Crude ash was obtained by incineration at 550 °C (ISO, 2002). Crude protein (Nx6.25) was determined according to Kjeldahl (ISO, 2005). Crude fat was extracted using petroleum ether after hydrolysis with HCl (ISO, 1999). Crude fibre

was obtained using the Ankom Fibre Analyser (Ankom Technology, Macedon, NY, USA) after boiling with sulphuric acid first and then with sodium hydroxide (EC, 1992).

The particle size distribution of the feeds was determined by wet sieve analysis according to Millet *et al.* (2012b). In short, 50 g of feed was placed in a tumbler and 1000 mL of water heated to 30 °C was added to the feed. The mixture was stirred with a spatula after 30 min. After an additional 30 min, the feed/water suspension was deposited onto the top of a sieve tower (sieves: 4.75, 3.35, 2.36, 1.18, 0.60, 0.30 mm) which was placed on a bowl with a downspout, then washed with 10 L cold, distilled water (water pressure = $1x10^5$ Pa). Afterwards, the sieve tower was dried (65 °C) in a ULE 800 ventilated oven (Memmert GmbH & Co KG, Schwabach, Germany) overnight, cooled down in a desiccator and each sieve was weighed again. Each feed was measured in duplicate. The particle size distribution of the meal diets was also determined by dry sieve analysis (ASAE, 1994). For this test, a sample of 300 g was sieved using a set of sieves with apertures of 9.5, 4.75, 3.35, 2.36, 1.18, 0.60, 0.30 mm respectively and a pan placed on a AS200 shaker (Retsch, Haan, Germany) for 5 min with an amplitude of 1.80 mm. The analysis was done in triplicate. The hardness of the pelleted feeds is expressed as the force (N) needed to break a pellet as measured by means of a Kahl Pellet Hardness Tester (Amandus Kahl Nachf., Hamburg, Germany). This was measured on 25 pellets for each pelleted diet and the average of these 25 measurements was considered the hardness value.

2.3.2.4 PERFORMANCE TRAITS

All pigs were weighed individually at the beginning of the trial, after two weeks and at the end of the weaning round. Feed consumption per pen was recorded and average daily feed intake (ADFI) was calculated for each weaning round. Average daily gain (ADG) and gain over feed (G:F) were also calculated over the entire weaning round on a pen basis.

2.3.2.5 INDOOR AIR QUALITY

Two Grimm 1.109 spectrometers (Grimm Aerosol Technik GmbH & Co. KG, Ainring, Germany), mounted in weatherproof housing, were used to monitor the indoor PM concentrations. Three different PM fractions were monitored: PM_{10}, $PM_{2.5}$ and PM_1. The fractions PM_{10}, $PM_{2.5}$ and PM1 are defined as particle matter which passes through a size-selective inlet with a 50 % efficiency cut-off at 10 µm aerodynamic diameter (AED), 2.5 µm

AED or 1 µm AED, respectively (U.S.EPA, 2004). The same instruments were used to log indoor temperature and relative humidity. Sampling was performed in an empty pen, 0.8 m above the slatted floor (Figure 2.14). Data were collected at intervals of 1 min. Because the number of Grimm spectrometers was smaller than the number of compartments to be sampled, a rotation scheme was used. Every day the Grimm spectrometers were moved to a different compartment, resulting in approximately one 24-h measurement per pen every 2 days. Ventilation rates were monitored using free running impellers (Fancom, Panningen, the Netherlands) and automatically logged using F-Central FarmManager software (Fancom, Panningen, the Netherlands) at a 5-min interval.

An Innova photoacoustic gas monitor 1314 (Innova AirTech Instruments, Santa Clara, CA, USA) connected to a multipoint sampler (CBISS, A1-Envirosciences ltd., Wirral, Merseyside, UK) was used to measure the indoor concentrations of NH_3, CO_2, CH_4 and N_2O sequentially in the four compartments. Sampling was performed in the middle of the central pen at 0.8m above the slatted floor (Figure 2.14). However, due to technical problems, it was not possible to measure the gas concentrations correctly and hence, this data is not presented here. More information on the technical problems encountered in this study can be found in section 4.1.

Outdoor conditions (temperature, relative humidity and atmospheric pressure) were monitored using the Vantage Pro2 weather station (Davis Instruments Corp., Davis, CA, U.S.A.).

2.3.2.6 DATA ANALYSIS

Raw data were combined into hourly means. Data from periods where a disturbance of the pigs was observed (e.g. entering of the compartment for purposes of animal management, repositioning of the measuring instrument or instrument failure) were excluded for further analysis. The compartment was considered the experimental unit, with pen as repeated measure within a compartment for performance results.

For determining the effect of grinding intensity and feed form on the growth performance, linear mixed models were built (using the GLIMMIX procedure in SAS9.4 (SAS Inst. Inc., Cary, NC)) using the different performance parameters (BW, ADFI, ADG and G:F from 4 to 6 weeks, from 6 to 9 weeks and from 4 to 9 weeks of age) as dependent variables. Grinding intensity,

feed form, sex and the interaction between particle size and feed form were investigated as independent variables. Compartment and weaning round were included as random effects. Except for BW at 4 weeks, models were corrected for the initial BW at 4 weeks.

For indoor air quality measurements, linear mixed models were built (using the GLIMMIX procedure) using the PM concentrations (PM_{10}, $PM_{2.5}$ and PM_1) as dependent variables. To achieve normally distributed residuals, PM concentrations were log transformed. Grinding intensity, feed form, their interaction, ventilation rate and indoor relative humidity were investigated as independent variables. Non-significant interactions were removed from the final model. Random effects for compartment and weaning round were included in the model to correct for repeated measurements within a compartment and within a weaning round. Corrections were also made for measuring device for PM_{10} and $PM_{2.5}$, but not for PM_1 concentrations.

The data was assumed to be normally distributed, based on graphical evaluation (histogram and QQ-plot of the residuals). Statistical significance was considered for $P < 0.05$ and a trend was considered for $0.05 < P < 0.1$.

2.3.2.7 ADDITIONAL LAB TESTS

To verify and to explain the results obtained inside the nurseries, 2 lab tests were performed: the “drop test” and the “shake test”.

During the drop test, 300 g of the diet was dropped from 1 m above the floor into a bucket using a device of our own design (Figure 2.15). This was repeated 4 times (4 trials) for each diet in a Latin square design. A Grimm 1.109 spectrometer, with a 6-sec time interval, was placed directly beside the bucket to record PM_{10}, $PM_{2.5}$ and PM_1 concentrations during 3 min.

During the shake test, 100 g of each diet was shaken for 20 minutes at 300 motions per minute using a mechanical shaker (Universal Shaker SM-30, Edmund Bühler GmbH, Tübingen, Germany). Particulate matter measurements were performed with a 6-sec time interval at 0.2 m above the mechanical shaker with a Grimm 1.109 spectrometer (Figure 2.16). The shake test was repeated 4 times (4 trials) for each diet in a Latin square design.

For both laboratory tests, data analysis was performed using the GLIMMIX procedure in SAS9.4. The trial was considered the experimental unit, with measurements every 6 seconds as repeated measure within a trial. For determining the effect of grinding intensity and feed form on the PM concentrations from the drop and shake tests, a linear mixed model was built using the PM concentrations (PM_{10}, $PM_{2.5}$ and PM_1) as dependent variables. To achieve normally distributed residuals, PM concentrations were log transformed for the drop test, PM concentrations for the shake test were already normally distributed. Grinding intensity, feed form and the interaction between both were investigated as independent variables. For the drop test, time after the drop was also added to the model. Residuals were assumed to be normally distributed, based on graphical evaluation (histogram and QQ-plot of the residuals). Statistical significance was considered for $P < 0.05$.

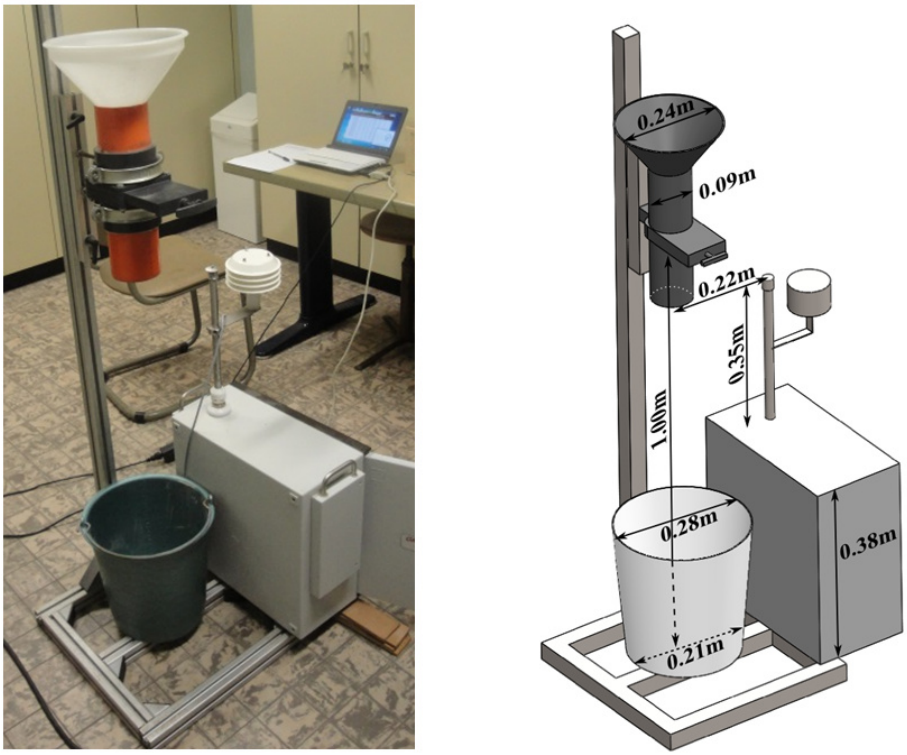

Figure 2.15. Device (our own design) used for the drop test. From each diet, 300 g was dropped from 1 m into a bucket. Particulate matter concentrations were measured with a Grimm 1.109 spectrometer (Grimm Aerosol Technik GmbH & Co. KG, Ainring, Germany).

Figure 2.16. Mechanical shaker and Grimm 1.109 spectrometer (Grimm Aerosol Technik GmbH & Co. KG, Ainring, Germany), without the weatherproof housing, used for the shake test. From each diet, 100 g was shaken for 20 min.

2.3.3 RESULTS

2.3.3.1 FEED CHARACTERISTICS

Within diets with the same grinding intensity, particle size distribution was comparable, despite a larger proportion of particles in the < 0.30 mm fraction in the pelleted diets (Table 2.9). The particle size of finely ground diets was lower than the particle size of the coarsely ground diets (Table 2.9 & Table 2.10). The pelleted diets did not have particles in fractions larger than 3.35 mm (coarsely ground) or 2.36 mm (finely ground). Pelleted diets had a lower mean particle size within each grinding intensity as compared to meal diets. Hardness values of the pellets were lower in the finely ground pelleted diet (77.8 ± 15.0 N) compared to the coarsely ground pelleted diet (93.6 ± 14.5 N).

Table 2.9. Particle size distribution of the diets measured in duplicate using wet sieve analysis.

Feed form	Meal		Pellets	
Particle size	Finely ground	Coarsely ground	Finely ground	Coarsely ground
< 300 µm, %	57.40	46.90	62.24	48.15
301-600 µm, %	19.55	16.11	20.06	12.99
601-1180 µm, %	21.69	15.89	16.92	25.53
1181-2360 µm, %	1.02	17.93	0.78	11.87
2361-3350 µm, %	0.23	2.61	0.00	1.46
3351-4750 µm, %	0.00	0.57	0.00	0.00
> 4750 µm, %	0.11	0.00	0.00	0.00
Mean particle size, µm	400	700	350	610

Table 2.10. Particle size distribution of the meal diets measured in triplicate using dry sieve analysis.

Particle size	Finely ground	Coarsely ground
< 300 µm, %	45.03	30.67
301-600 µm, %	32.78	22.46
601-1180 µm, %	16.26	26.11
1181-2360 µm, %	2.94	17.85
2361-3350 µm, %	0.86	2.42
3351-4750 µm, %	1.11	0.44
4751-9500 µm, %	1.02	0.04
> 9500 µm, %	0.00	0.00
Mean particle size, µm	550	790

Table 2.11. Effect of grinding intensity and feed form on indoor PM fractions[1].

	Meal		Pellet		P-value	
Fraction, µg m^{-3}	Finely ground	Coarsely ground	Finely ground	Coarsely ground	Particle size	Feed form
PM_{10}	171 [74.4;390]	147 [64.2;337]	597 [261;1365]	515 [225;1178]	0.014	<0.001
$PM_{2.5}$	24.6 [11.4;52.8]	22.3 [10.4;47.9]	71.9 [33.4;155]	65.1 [30.3;140]	0.065	<0.001
PM_{1}	9.6 [7.2;12.8]	8.9 [6.7;11.9]	12.9 [9.7;17.1]	12.0 [9.0;15.9]	0.223	<0.001

[1] Back-transformed LSMeans are given with back-transformed 95% CI.

2.3.3.2 INDOOR AIR QUALITY

The indoor PM_{10}, $PM_{2.5}$ and PM_1 concentrations were higher in the compartments where pellets were fed to the pigs ($P < 0.001$) (Table 2.11). Within feed form, feeding the finely ground feed resulted in higher indoor PM_{10} concentrations ($P < 0.05$). A tendency to a similar effect of grinding intensity was observed for $PM_{2.5}$ ($P < 0.1$), but not for PM_1 concentrations. No interaction between feed form and grinding intensity was found on any PM fraction.

Ventilation rate and indoor relative humidity had an effect ($P < 0.001$) on all PM fractions. In this study, an increase in ventilation rate of 1 m^3/h gave rise to increased PM_{10} (+ 0.0047 $\mu g/m^3$), $PM_{2.5}$ (+ 0.0041 $\mu g/m^3$) and PM_1 (+ 0.0019 $\mu g/m^3$) concentrations. An increase in the indoor relative humidity also increased the indoor PM concentrations. An increase of 1 % in indoor relative humidity corresponded with increases of 0.054 $\mu g/m^3$, 0.056 $\mu g/m^3$ and 0.036 $\mu g/m^3$ respectively for indoor PM_{10}, $PM_{2.5}$ and PM_1 concentrations. It should be kept in mind that these effects were established at 0.8m above the slatted floor.

2.3.3.3 PERFORMANCE

In total, 5 pigs (3 on the coarsely ground meal diet, 1 on the finely ground meal diet and 1 on the finely ground pelleted diet) died over the course of the experiment. One pig that received the finely ground pelleted diet was eliminated from the experiment for health reasons.

Between 4 and 6 weeks of age, the pigs consumed more of the coarsely ground feed ($P < 0.001$). In this period, we observed a tendency for higher feed intake on the meal versus pelleted diet ($P < 0.1$) without an interaction between grinding intensity and form (Table 2.12). However, between 6 and 9 weeks of age ($P < 0.05$) and over the entire experiment ($P < 0.05$), grinding intensity and form did interact on ADFI. On the pellets, grinding intensity did not affect ADFI significantly. On the meal diet however, the piglets consumed more ($P < 0.05$) of the coarsely ground meal than the finely ground meal. Both groups consumed more feed than the groups receiving pellets.

An interaction between grinding intensity and feed form on ADG was found for the 2 periods ($P < 0.05$ between 4 and 6 weeks of age and $P < 0.05$ between 6 and 9 weeks of age) and for the whole experiment ($P < 0.001$). For the periods 4 to 6 and 4 to 9 weeks of age, pigs fed on finely ground meal had lower ADG ($P < 0.05$) than pigs of the other 3 groups. However, for

the period 6 to 9 weeks of age, it was not possible to further differentiate between groups using Tukey's *post hoc* test.

An interaction between grinding intensity and form on BW at 6 ($P < 0.05$) and 9 ($P < 0.001$) weeks of age was found. Pigs that received the finely ground meal had lower BW ($P < 0.05$) than pigs of the other 3 groups at both time points.

Between 4 and 6 weeks of age, an interaction ($P < 0.001$) between grinding intensity and feed form on G:F was found. Pigs receiving finely ground meal had a lower G:F ($P < 0.05$) than pigs receiving coarsely ground meal, which was in turn lower ($P < 0.05$) than pigs receiving pellets, with no differences in the 2 particle sizes when fed as pelleted diets. For the periods between 6 to 9 weeks and over the entire experiment, an interaction between grinding intensity and form was not observed. Pigs fed meal had lower ($P < 0.001$) G:F than pigs fed pelleted feeds. Across the entire experiment, grinding intensity did not significantly affect G:F.

Sex did not affect the measured parameters significantly, except for the daily gain between 6 and 9 weeks of age ($P < 0.05$, data not shown). Male pigs had a higher daily gain during that period than female pigs.

Table 2.12. Effect of grinding intensity and feed form on the growth performance of the pigs.

	Meal		Pellets			*P*-value		
	Finely ground	Coarsely ground	Finely ground	Coarsely ground	SEM	Grinding intensity	Form	Grinding intensity x Form
Body weight, kg								
4 weeks	8.01	8.04	8.01	7.98	0.13	0.989	0.924	0.914
6 weeks	10.67[a]	11.33[b]	11.47[b]	11.5[b]	0.15	<0.001	<0.001	0.002
9 weeks	21.44[a]	22.97[b]	23.02[b]	22.7[b]	0.25	0.080	0.008	<0.001
Average daily feed intake, g								
4-6 weeks	294[b]	324[a]	282[b]	292[ab]	4.61	<0.001	0.078	0.126
6-9 weeks	802[b]	869[a]	735[c]	710[c]	11.41	0.188	<0.001	0.004
4-9 weeks	599[b]	651[a]	554[c]	543[c]	7.92	0.059	<0.001	0.004
Average daily gain, g								
4-6 weeks	190[a]	235[b]	246[b]	251[b]	4.73	<0.001	<0.001	0.002
6-9 weeks	513	555	549	534	6.17	0.781	0.514	0.002
4-9 weeks	384[a]	427[b]	428[b]	421[b]	4.64	0.075	0.009	<0.001
Gain over feed, g:g								
4-6 weeks	0.637[a]	0.719[b]	0.871[c]	0.858[c]	0.012	0.015	<0.001	<0.001
6-9 weeks	0.641[a]	0.638[a]	0.743[b]	0.753[b]	0.007	0.892	<0.001	0.431
4-9 weeks	0.642[a]	0.656[a]	0.771[b]	0.776[b]	0.007	0.521	<0.001	0.530

[a,b,c] Within a row, values without a common superscript differ (P < 0.05) according to Tukey-Kramers' *post hoc* test.

2.3.3.4 LABORATORY TESTS

An interaction ($P < 0.05$ for PM_{10}, $P < 0.05$ for $PM_{2.5}$, $P < 0.05$ for PM_1) between grinding intensity and feed form on PM concentrations was found (Table 2.13). PM_{10} concentration differed ($P < 0.05$) between the 2 meal diets, but for the pelleted diets, no significant differences were found. Finely ground meal diet showed the highest PM_{10} concentration in the drop test. According to Tukey's *post hoc* test, no differences were found between the coarsely ground meal diet and the coarsely ground pelleted diet. Finely ground pelleted diet had the lowest PM_{10} concentration in the drop test. A difference in $PM_{2.5}$ concentrations was only found between the finely ground meal and the coarsely ground meal ($P < 0.05$), the pellets gave intermediate values. For PM_1 concentrations, although an interaction ($P < 0.05$) between grinding intensity and feed form was found, it was not possible to further differentiate between diets.

An interaction ($P < 0.05$) between grinding intensity and feed form was observed in the shake test for PM_{10} concentrations (Table 2.13). Shaking the pelleted diets gave rise to higher PM_{10} concentrations than shaking the meal diets, with highest PM_{10} concentrations for the finely ground pelleted diets. No differences in PM_{10} concentrations between the 2 meal diets were found. For $PM_{2.5}$ concentrations, only the grinding intensity had an effect ($P < 0.05$), with coarsely ground diets giving rise to higher $PM_{2.5}$ concentrations as compared to the finely ground diets. Shaking the coarsely ground meal resulted in the highest $PM_{2.5}$ concentrations. For PM_1 concentrations, neither the grinding intensity nor the feed form nor the interaction of both had a significant effect.

Table 2.13. Effect of grinding intensity and feed form on indoor PM fractions obtained from the drop and shake tests[1].

	Meal		Pellet		P-value		
Fraction, μg m^{-3}	Finely ground	Coarsely ground	Finely ground	Coarsely ground	Particle size	Feed form	Particle size x form
Drop test							
PM_{10}	327^{a} [241;445]	172^{b} [127;234]	89^{c} [66;121]	107^{bc} [79;145]	0.126	<0.001	0.013
$PM_{2.5}$	14.8^{a} [12.6;17.4]	10.2^{b} [8.7;12.0]	11.2^{ab} [9.6;13.2]	12.9^{ab} [10.9;15.2]	0.139	0.793	0.005
PM_{1}	7.9 [6.6;9.6]	5.8 [5.7;8.4]	6.9 [4.8;7.0]	8.1 [6.7;9.7]	0.364	0.270	0.020
Shake test							
PM_{10}	18.8^{c} [16.1;21.4]	21.1^{c} [18.4;23.7]	49.5^{a} [46.9;52.2]	42.5^{b} [39.9;45.2]	0.074	<0.001	0.002
$PM_{2.5}$	6.7^{b} [5.8;7.5]	8.8^{a} [8.0;9.7]	8.0^{ab} [7.2;8.9]	8.7^{a} [7.8;9.5]	0.003	0.136	0.072
PM_{1}	4.9 [3.8;6.1]	6.3 [5.1;7.4]	5.7 [4.6;6.9]	6.5 [5.4;7.7]	0.063	0.331	0.635

[a,b,c] Within a column, values without a common superscript differ (P < 0.05) according to Tukey's *post hoc* test.

[1] Back-transformed LSMeans are given with back-transformed 95% CI.

2.3.4 DISCUSSION

The results of the current study indicate that both the physical form and grinding intensity of the feed affect largely the growth characteristics of weaning pigs as well as the indoor PM concentrations.

This direct link between PM and feed was also found by other authors for finishing pigs (Heber *et al.*, 1988a) and weaned piglets (Aarnink *et al.*, 1999). Surprisingly however, pelleted diets gave rise to higher PM concentrations as compared to meal diets (more pronounced for PM_{10} and $PM_{2.5}$ than for PM_1). The effect of the grinding intensity seemed smaller, but nevertheless finely grinding the diet led to larger PM concentrations. It must be noted that ingredients which were already very fine or in a liquid form, were not ground before mixing. This may be one explanation for the smaller effect of grinding intensity on the indoor PM concentrations. Furthermore, the hardness of the coarsely ground pelleted diets was higher than the hardness of the finely ground pelleted diets. To our knowledge, no other studies have reported an increase in PM concentrations when feeding pellets to pigs. A possible explanation is that pelleting, which is considered to be a secondary grinding step (Grosse Liesner *et al.*, 2009), leads to smaller particles. This has been shown by Wolf *et al.* (2010) and is in line with the slightly higher amount of fine particles in the pelleted diets as determined with the wet sieve analysis, which can eventually lead to a smaller mean particle size. Similarly, Bundy and Hazen (1975) attributed the lack of difference between pellets and meal in PM concentrations to the degradation of the pellets, which become a fine powder in the bottom of the self-feeders. Nevertheless, the fractions determined by the sieve analysis (with the smallest sieve at 300 µm), or the "visible fines" are far larger than PM fractions. Therefore, the results of the wet sieve analysis could be a better indicator for the breakdown of the diets in the animal rather than as an indicator for the production of PM. So, there might be no direct link between the results from the sieve analysis and the observed indoor PM concentrations. In an attempt to retrieve a better indicator of the production of PM and to find further possible explanations for the observed differences in PM concentrations, some additional laboratory tests on the experimental feeds were performed. Tests similar to the drop test have been used previously (Gast & Bundy, 1986; Heber & Martin, 1988). Dropping the feed into a bucket gave rise to higher PM_{10} concentrations for the meal diets compared to the pelleted ones. This is the opposite of what was found in our pig barn

experiments. However, during these experiments, data recorded during the dropping of the feeds into the self-feeders were not included in our dataset. For $PM_{2.5}$ and PM_1 concentrations, small or no differences between the different diets were found. In an attempt to simulate the frictions between the different particles in the feed that can occur when the piglets were eating, a shake test was performed. The results of this test indicated that shaking the feeds led to higher PM_{10} concentrations for the pelleted diets, with highest PM_{10} concentrations coming from the finely ground pelleted diet. Again, for $PM_{2.5}$ and PM_1 concentrations, small or no differences were found. These results were in line with the observations in the pig compartments. Whereas the drop test, which involved 1 single action, might be a good measure of the superficial ("loose") PM, the shake test caused frictions in the sample and might indicate PM production as a result of agitation, possibly similar to frictions caused when pigs were eating. The shake test could also be considered as an indicator for the pellet durability. Optimising the laboratory tests (e.g. different drop height, different motions per minute, different placement of the Grimm spectrometer) might clarify the obtained results even further. The effect of both ventilation rate and indoor relative humidity on PM concentrations was very small (but significant) and seemed negligible compared to the observed differences in indoor PM concentrations.

In the current study, pigs fed meal diets had higher ADFI than pigs fed pelleted diets. However, an interaction between the physical form and grinding intensity was found: grinding intensity only had an effect on the meal diets with higher ADFI for the coarsely ground meal. Other researchers also found that reducing the grinding intensity reduced the ADFI in weaning pigs (Healy *et al.*, 1994; Mavromichalis *et al.*, 2000). The same was found for finishing pigs (Wondra *et al.*, 1995b). O'Doherty *et al.* (2001) also found decreased ADFI with pelleted feed. According to the authors, this can be a consequence of the increased energy value of the diets after pelleting, as feed intake is correlated with the energy value of the diets (Henry, 1985). Furthermore, before entering the trial, all piglets received creep feed, a meal diet, in the farrowing unit. The change from one form of diet to another can lead to lower feed intakes. This was already proven for turkeys when changing from crumbs to pellets (Lecuelle *et al.*, 2010). Although care was taken to limit spillage and no significant feed losses were observed during the course of the experiments, it cannot be excluded that part of the higher ADFI for meal diets versus pelleted diets was caused by greater spillage of

the feed due to feed sticking to the mouth of the pig or being thrown out of the trough during rooting (O'Doherty *et al.*, 2001). This would also explain the lower feed efficiency found for pigs receiving meal.

Also for ADG, an interaction was also found between the form and grinding intensity. Finely ground meal had a lower ADG than the other 3 diets. No differences could be detected between pelleted feeds and coarsely ground meal. In other studies, some authors found a positive effect of pelleting on ADG (Wondra *et al.*, 1995a; Wondra *et al.*, 1995b), whereas others did not observe an improvement (Vande Ginste & De Schrijver, 1998). Wondra *et al.* (1995b) found no significant effect of reducing particle size on ADG, whereas Mavromichalis *et al.* (2000) found an increase in ADG when increasing the grinding intensity of the diets from 1.3 mm to 0.6 µm, but no increase was found when particle size was reduced to 0.4 mm. To the contrary, in the current study, finely grinding the diet caused slower gain, probably because of reduced feed intake. However, the coarsely ground diet had a mean particle size that was already comparable to the finely ground diet in the study of Mavromichalis *et al.* (2000).

The physical form of the feed had a clear impact on the feed efficiency. In line with other research (O'Doherty *et al.*, 2001; Vande Ginste & De Schrijver, 1998; Wondra *et al.*, 1995b), pelleting the feed gave rise to higher G:F. The pelleting process makes the nutrients in the feeds more accessible to digestive enzymes and can potentially improve the nutritional quality process (Vande Ginste & De Schrijver, 1998; Wondra *et al.*, 1995b). Also, as mentioned before, reduced feed losses may be an explanation (O'Doherty *et al.*, 2001). One would expect that reducing the particle size gives rise to an increased surface area which makes the nutrients more accessible to digestive enzymes (Mavromichalis *et al.*, 2000; Wondra *et al.*, 1995b). Decreased particle size has been linked to an increased ileal digestibility of proteins and amino acids (Lahaye *et al.*, 2004). This is in line with the finding of Healy *et al.* (1994) and Wondra *et al.* (1995b), who observed better feed efficiency in finely ground diets. Similarly, Mavromichalis *et al.* (2000) found increased G:F when decreasing feed particle size from 1.3 to 0.6 mm, but a decrease in G:F when decreasing the particle size further to 0.4 mm. Chae *et al.* (2000) found no effect of reductions in particle size on digestibility of amino acids in the ileum. In the current study, finely grinding the meal diet led to a lower feed efficiency during the first 2 weeks of the trial. It seems logical to

assume that this is a result of lower feed intake and daily gain, which indicates that a relatively high amount of the feed was used to cover maintenance energy requirements.

The present results show a contradiction between indoor air quality and performance results: whereas performance was best on the pelleted diets, these gave rise to a higher amount of PM in the barn. Besides, intestinal health was not taken into account. It is known that finely grinding the feed increases the risk of developing gastric ulcers (Grosse Liesner *et al.*, 2009; Millet *et al.*, 2012b; Wondra *et al.*, 1995b). Furthermore, because pelleting the diets gives rise to a reduction in particle size (Wolf *et al.*, 2010), pelleting can also increase the development of gastric ulcers and is considered equivalent to a secondary grinding process. Indeed, feed processing affected ulcer scores in slaughtering pigs (Millet *et al.*, 2012a).

As with performance results and the risk on gastric ulceration, it seems plausible that the amount of particulate matter depends on the intensity and the type of feed processing (e.g. using steam pelleting, expanding or extrusion processes) or the ingredient composition. Besides the health of the animals, the health of the farmer is another important parameter not taken into account in the current study. Upscaling in pig production has created full-time occupations in which workers are exposed to PM more frequently and extensively (Pedersen *et al.*, 2000). Several studies have reported a relationship between exposures to PM in livestock buildings and respiratory symptoms in farm workers (Donham *et al.*, 1995; Radon *et al.*, 2001; Reynolds *et al.*, 1996). Furthermore, emissions of high PM concentrations can influence the local air quality and can cause health problems for the nearby inhabitants (Pope *et al.*, 2002). Therefore, further research will be necessary before guidelines can be recommended for optimising feed structure that also account for performance, indoor air quality, health of the animals and the health of the farmers and nearby inhabitants.

2.3.5 CONCLUSION

Whereas pelleting improved performance results, it also gave rise to higher indoor PM concentrations. The effect of grinding intensity on PM concentration was less pronounced, but still visible. Moreover, finely grinding the meal diet negatively affected performance results.

3 REDUCED SAMPLING STRATEGIES

This chapter is dedicated to the comparison of different reduced sampling strategies to sample NH_3 emissions and to determine an NH_3 emission factor. Computerised simulations were run to determine the effect of different reduced sampling strategies.

3.1 EVALUATION OF SAMPLING STRATEGIES FOR THE DETERMINATION OF AMMONIA EMISSIONS FROM FATTENING PIG FACILITIES

3.1.1 INTRODUCTION

Ammonia was the first gas in agriculture subjected to mandatory emission reduction. As a consequence, LAE housing systems were introduced in Flanders since 2004. Pig and poultry farmers are obliged to use officially approved LAE housing systems when renovating, expanding or building new animal housing. Innovative farmers can also ask permission to build new LAE housing systems of which the reduction potential towards NH_3 is presently not yet established through measurements. The NH_3 emission from these housing systems has to be measured by an officially approved (research) institute in order to determine an EF. This is traditionally done by measuring the NH_3 concentrations and ventilation rates frequently (at least every hour) during long periods (> 200 days for fattening pigs), covering warm and cold seasons (Groen Label, 1996). However, the costs associated with this methodology are high (up to € 50 000) (Dekock *et al.*, 2009). Therefore, researchers developed several reduced sampling strategies to obtain an NH_3 EF for a housing system (Mosquera & Ogink, 2011; Ogink *et al.*, 2011; Ogink *et al.*, 2013a) or for a specific barn (Ogink *et al.*, 2000; Vranken *et al.*, 2004).

Up to now, all reduced sampling strategies suggested in literature take into account the parameters that influence NH_3 emissions, such as the increasing live weight of the pigs during a fattening period and the seasonal variations in NH_3 emissions. A different method, that does not N_2O emissions from a wastewater treatment plant (Daelman *et al.*, 2013). In the present study, an introductory analysis on a limited number of datasets was performed to evaluate if a similar methodology could be applicable for the determination of an NH_3 EF for pig houses. Therefore, we evaluated a methodology which investigated the accuracy of the estimated EF solely as a function of the sampling frequency and strategy. When comparing the different sampling strategies, the measuring costs (both labour costs of a technician and the operating costs of the gas monitor) associated with each strategy and the number of barns for which an EF could be estimated each year were also taken into account. It must be noted that the sampling strategies were only compared for the determination of

an EF for a specific barn, and not for a housing system. In the future, analyses on multiple datasets are desirable to confirm the obtained results.

3.1.2 MATERIAL AND METHODS

3.1.2.1 DATASETS

Two datasets were used to evaluate the influence of reduced sampling strategies on the estimated NH_3 emission factor from fattening pig facilities. The datasets originated from measurements performed to assign Green Label certificates of different animal housing systems in the Netherlands, applying the official Dutch measurement protocol for ammonia emission factors.

The first dataset was collected in a conventional fattening pig facility between March 13th, 1996 and November 18th, 1996 during two consecutive fattening periods (Figure 3.1, A & B and Figure 3.2, A & B). During the measurements, 130 fattening pigs were present in the pig unit. The dataset consisted of 5628 hourly outgoing (exhaust) and incoming (inlet) NH_3 concentrations (mg m^{-3}) and ventilation rates (m^3 $hour^{-1}$). The dataset had a completeness of 97.0 % for fattening period 1 and 95.2 % for fattening period 2.

The second dataset was collected in a LAE fattening pig facility between January 28th, 2001 and September 18th, 2001 during two consecutive fattening periods (Figure 3.1, C & D and Figure 3.2, C & D). During the measurements, 144 fattening pigs were present in the pig unit. The dataset consisted of 5338 hourly outgoing NH_3 concentrations (mg m^{-3}) and ventilation rates (m^3 $hour^{-1}$). No information on the incoming NH_3 concentrations (background level) was present. The dataset had a completeness of 92.2 % for fattening period 1 and 98.2 % for fattening period 2.

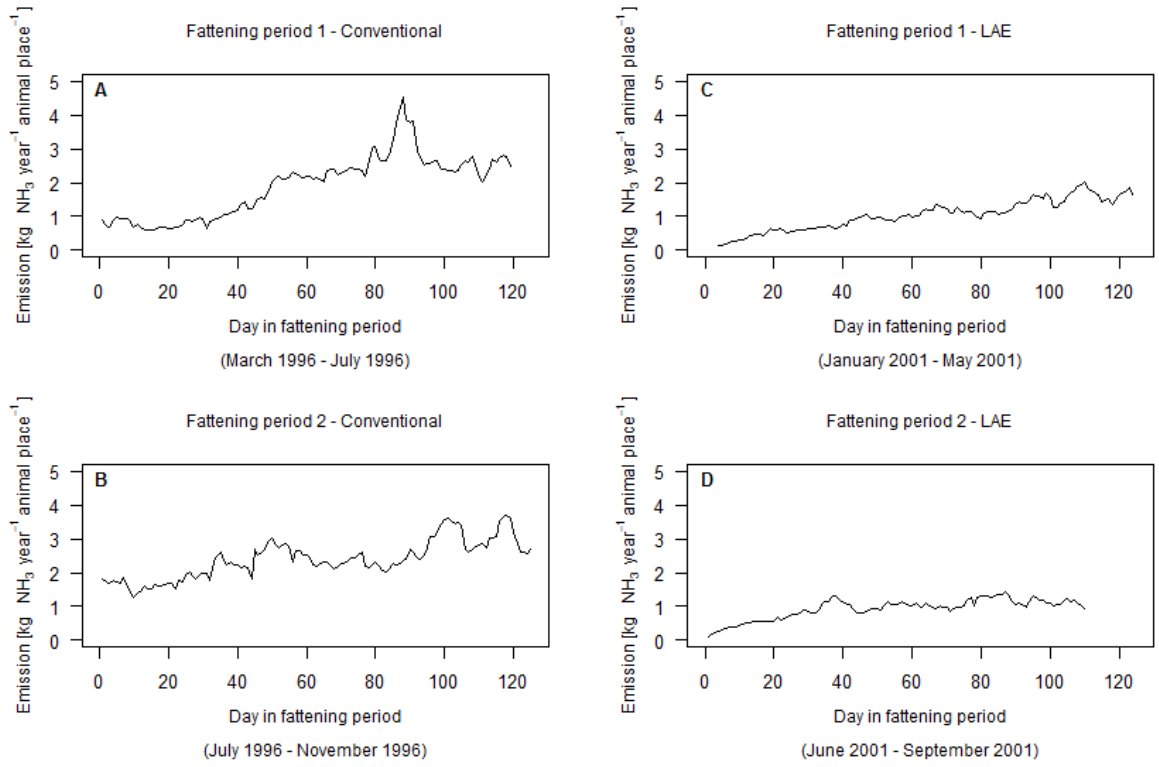

Figure 3.1. Daily NH_3 emission for both housing systems and both fattening periods.

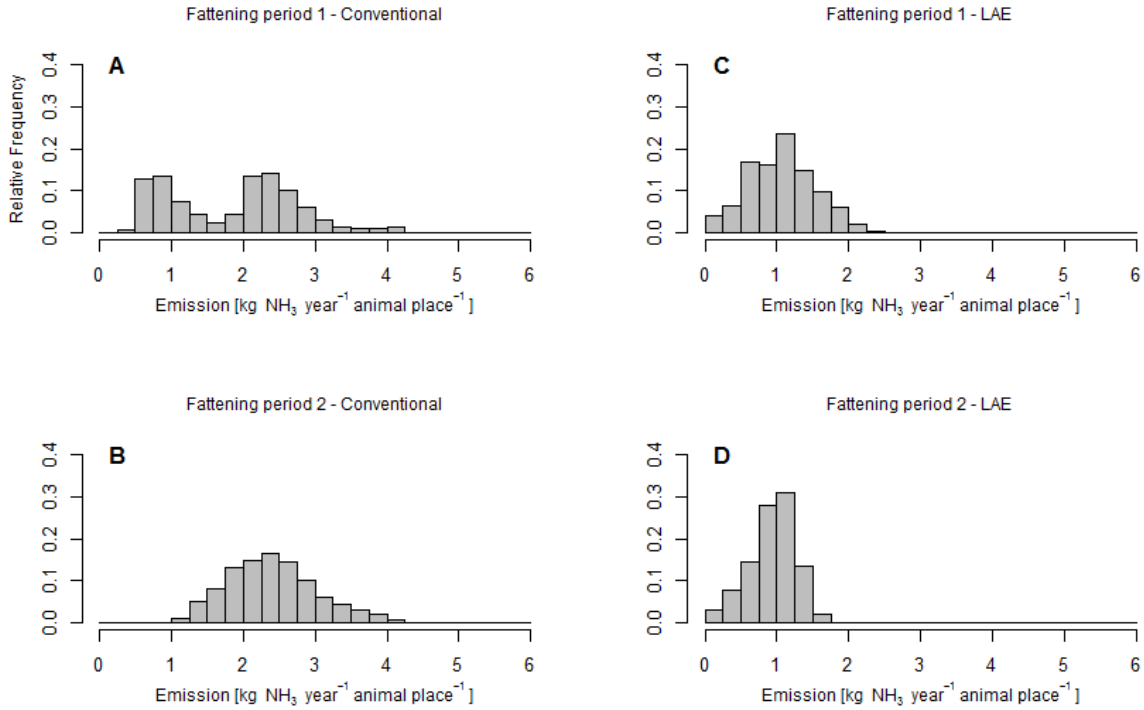

Figure 3.2. Distribution of the hourly NH_3 emission rates for both barns (conventional and LAE) and both fattening periods.

In the first dataset, for each hour, the NH_3 emission rate (ER, g $hour^{-1}$) was calculated according to Eq. 3.1. The average NH_3 EF (kg $year^{-1}$ $(animal\ place)^{-1}$) was then calculated (Eq. 3.2) by taking the mean over all the (N) NH_3 emission rates and converting this mean from g $hour^{-1}$ to kg $year^{-1}$ $(animal\ place)^{-1}$. In the second dataset, the hourly NH_3 emission rates and the average NH_3 EF were calculated in the same way, except that no correction for incoming NH_3 concentrations was made.

$$ER = Q * [C_o - C_i] * 10^{-3} \quad \text{Eq. 3.1}$$

$$EF = \frac{\sum ER}{N} * \left(\frac{10^{-3} * 24 * 365}{a}\right) \quad \text{Eq. 3.2}$$

With *ER*: emission rate (g $hour^{-1}$), *Q*: ventilation rate (m^3 $hour^{-1}$), C_o: outgoing NH_3 concentration (mg m^{-3}), C_i: incoming NH_3 concentration (mg m^{-3}), *a*: number of animal places and *N*: number of emission rates.

Based on these calculations, the pig unit in dataset 1 had an EF of 2.16 kg NH_3 $year^{-1}$ $(animal\ place)^{-1}$ and the pig unit in dataset 2 had an EF of 1.01 kg NH_3 $year^{-1}$ $(animal\ place)^{-1}$.

3.1.2.2 EVALUATION OF REDUCED SAMPLING STRATEGIES

Five different sampling strategies were investigated (Table 3.1): single grab sampling, continuous sampling (for 24 hours), continuous sampling (for 48 hours), 7 day sampling and weekly grab sampling. With these reduced sampling strategies, EFs were estimated for both datasets using the same approach and implementations as in Daelman et al. (2013). The different reduced sampling strategies were then evaluated by comparing their estimated EF with the true average EF as calculated above.

SAMPLING STRATEGIES

An estimate of the NH_3 EF was made by averaging the hourly NH_3 emission rates over one single grab sample, one continuous 24 hour period, one continuous 48 hour period or one full week respectively and converting these averages to kg $year^{-1}$ $(animal\ place)^{-1}$ (Eq. 3.2). An estimate of the NH_3 EF was also made by taking a weekly grab sample (a one hour measurement value) on a specific day of the week during several consecutive weeks, averaging the obtained grab sample results and converting this average to kg $year^{-1}$ $(animal\ place)^{-1}$ (Eq. 3.2).

For all sampling strategies, restrictions were applied, taking into account the normal working conditions in Flanders. For the single and weekly grab sampling strategy, it was assumed that a grab sample would always be taken between 9 a.m. and 5 p.m. during working days (single grab sampling strategy) or during a specific day of the week or a random working day (weekly grab sampling strategy). For the 24 hour and 48 hour continuous sampling strategies, it was presumed that measurements would not be performed during weekends and had to start between 10 and 11 a.m. For the 7 day sampling campaign it was assumed that measurements could only start on working days between 10 and 11 a.m.

Table 3.1. Overview of evaluated reduced sampling strategies.

Reduced sampling strategies	Protocol
Single grab sampling	One sampling period of 1 hour on a working day (Monday to Friday) between 9 a.m. and 5 p.m.
Continuous sampling (24 h)	One sampling period of 24 consecutive hours on a working day (Monday to Friday) with the start of the measurement between 10 a.m. and 11 a.m. on Monday to Thursday.
Continuous sampling (48 h)	One sampling period of 48 consecutive hours on a working day (Monday to Friday) with the start of the measurement between 10 a.m. and 11 a.m. on Monday to Wednesday.
Continuous sampling (7 day)	One sampling period of 7 consecutive days with the start of the measurement between 10 a.m. and 11 a.m. on a working day (Monday to Friday).
Long-term weekly grab sampling	Sampling for 1 hour on a fixed weekday or on a random working day between 9 a.m. and 5 p.m. during several consecutive weeks.

For all possible single grab samples, 24 hour, 48 hour or 7 day periods which fulfilled these conditions, emissions rates (ER) were calculated and then converted to kg $year^{-1}$ (animal $place)^{-1}$. For the 24 hour, 48 hour and 7 day periods, emission rates based on respectively less than 16, 32 or 112 sampling hours were not taken into account. The number of possible estimates for the single grab sampling, 24 hour, 48 hour or 7 day period campaigns and for both datasets are shown in Table 3.2. All possible estimates were collected in histograms with a bin width of 0.25 kg NH_3 $year^{-1}$ (animal $place)^{-1}$.

Table 3.2. Number of all possible estimates for the different sampling strategies from dataset 1 and 2.

Sampling strategy	dataset 1	dataset 2
Single grab	1244	1258
24 hour	115	116
48 hour	72	85
7 day	138	142

To implement the long-term weekly grab sampling strategy, hourly NH_3 emission rates (between 9 a.m. and 5 p.m.) were randomly selected for each day in the week. This resulted in seven long-term weekly grab sampling campaigns of 32, 33 or 34 weeks for dataset 1 and 31, 33 or 34 weeks for dataset 2. The number of available weeks could differ because not all days were equally represented in the initial datasets due to interruptions in the real sampling campaigns. Contrary to the previously reduced sampling strategies, the total number of possible estimates was very high. Therefore, the procedure was repeated 1000 times for each week day in order to get a representative sample of the full number of possible estimates. Doing so, 1000 estimated NH_3 EFs were obtained for each weekday. This procedure was also applied to the scenario where a weekly sample was taken on a random working day instead of on a fixed day, again repeated 1000 times. For each of the fixed and random weekday campaigns, the 1000 values were possible outcomes for the respective sampling campaign. All possible estimates were collected in histograms with a bin width of 0.25 kg NH_3 $year^{-1}$ (animal $place)^{-1}$.

3.1.2.3 ACCURACY AS FUNCTION OF SAMPLING FREQUENCY

The influence of taking multiple (*n*) single grab samples, 24 hour periods, 48 hour periods, 7 day periods or long-term weekly grab samples during *n* consecutive weeks on the estimated EF was evaluated. These evaluations started by calculating all possible single grab samples, 24 hour periods, 48 hour periods, 7 day periods or long-term weekly grab sample campaigns that fulfil the requirements as mentioned in section 3.1.2.2. Thereafter 1 to *n* single grab samples, 24 hour periods, 48 hour periods, 7 day periods or long-term weekly grab sample campaigns for 1 to *n* consecutive weeks were randomly taken for all possible periods. This was done by making use of script in R 3.1.1 for Windows (R Core Team, 2013). Finally, for all 1 to *n* sampling cases, the relative error between the true EF (as determined in 3.1.2.1 and based on the whole dataset) and the estimated EF (based on the reduced number of measurements and only representing an estimation of the true EF) was determined as a function of the number (*n*) of sampling periods for each of the investigated reduced sampling strategies (Daelman *et al.*, 2013). In the different reduced sampling strategies, these sampling periods were the number of 24 hours (or 48 hours) random periods or 7 day periods. For the single grab sampling campaigns, the number of single grab samples was determined necessary to obtain an estimate with an acceptable relative error. It was decided that the relative error had to be smaller than ± 15 % to be acceptable. Similarly, for the long-term weekly grab sampling campaign, the number of weeks necessary to obtain an estimate with a relative error smaller than ± 15 % was determined.

RELATIVE ERROR

The relative error (ε) was calculated for all sampling strategies using the following equation:

$$\varepsilon = \frac{Estimated\ emission\ factor - True\ emission\ factor}{True\ emission\ factor} \qquad \text{Eq. 3.3}$$

For the 24 hour/48 hour sampling period, this relative error could be calculated for any estimated EF, based on a number (*n*) of 24 hour/48 hour periods. These *n* 24 hour/48 hour periods were randomly picked from all possible 24 hour/48 hour periods calculated in section 3.1.2.2 and *n* could not be greater than the maximum number of possibilities calculated in section 3.1.2.2. These periods did not have to be consecutive. An example of a plot of the relative error ε against the number (*n*) of 24 hour periods for one simulation is seen in Figure 3.3.

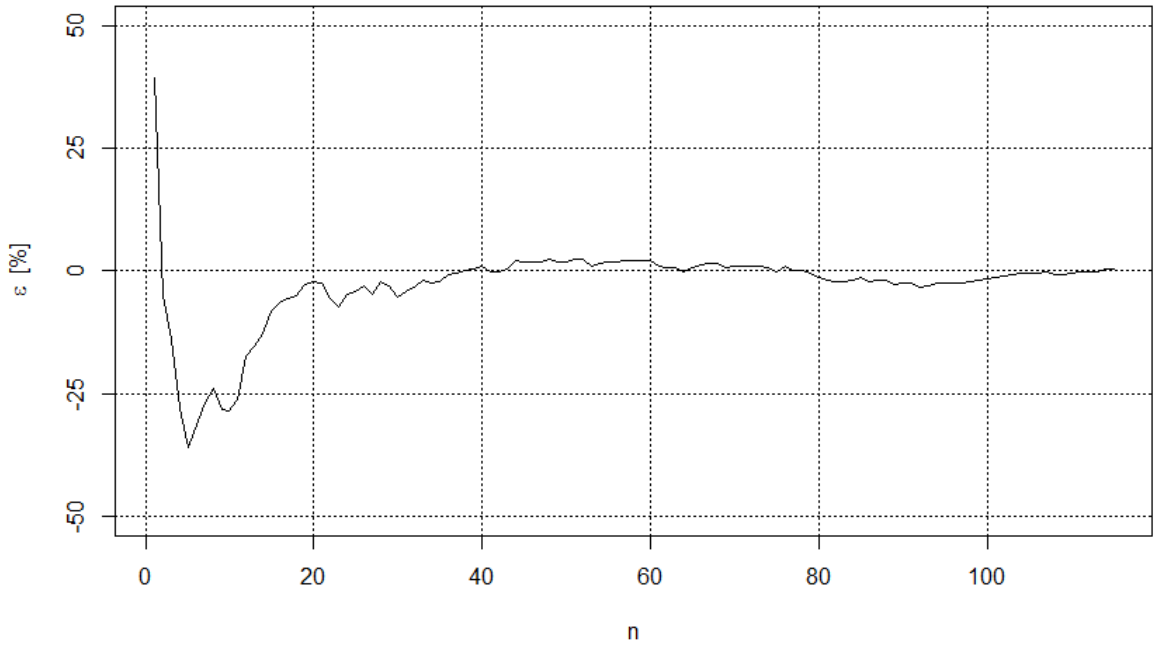

Figure 3.3. Relative error (ε) between the estimated emission factor and the true emission factor versus the number (n) of 24 h hour periods for one simulation.

The accuracy of the estimated EF as a function of the number of 7 day periods was assessed in the same way as for the number of 24 h and 48 hour sampling periods. As a result, 138 (dataset 1) and 142 (dataset 2) 7 day moving averages were calculated, representing all possible 7 consecutive measuring day periods starting between 10 and 11 a.m. and not starting in weekends. The maximum of non-overlapping periods in dataset 1 and 2 were 27 and 28, respectively. Thus, a monitoring campaign could exist of n random non-overlapping and not necessarily consecutive weeks, with a maximum of n = 27 (dataset 1) or n = 28 (dataset 2). As for the 24 hour and 48 hour sampling campaign, all of the hourly ERs over the n 7 day periods were averaged for each n.

A grab sampling campaign could consist of 1 to n grab samples. However, there were 1244 and 1258 possible single grab samples between 9 a.m. and 5 p.m. on working days in respectively dataset 1 and dataset 2. Taking a lot of grab samples is not very practical. In order to mimic a practical grab sampling campaign, 1 to 100 random grab samples were taken from the 1244 or 1258 possible grab samples. For each n, the n grab samples were

averaged to obtain an estimated ER and then converted to kg year^{-1} (animal place)$^{-1}$. This resulted in 100 estimates, one for each value of *n*.

The accuracy for the long-term weekly grab sampling strategy was determined for taking weekly grab samples on a random working day. So, this means that a grab sample would be taken on *n* consecutive weeks, each week on a random working day. This was simulated by combining the daytime values of the weekdays per week and taking *n* weekly grab samples from that combination during 1 to *n* weeks, starting on a random week. For each value of *n*, the values from the *n* weekly grab samples were averaged and then converted to kg year^{-1} (animal place)$^{-1}$. This resulted in 35 (dataset 1) and 34 (dataset 2) estimated EFs.

For each sampling strategy and for each value of *n*, 1000 simulations were executed, yielding a distribution of the relative errors as a function of the number of sampling periods. Subsequently, the average (μ) and the standard deviation (σ) of this distribution were calculated for each value of *n*. The uncertainty bounds for ε are given by $\mu - 2\sigma$ and $\mu + 2\sigma$. If the distribution of the relative error as a function of the number of sampling periods is normally distributed, the uncertainty bounds are the 95 % confidence interval around the mean. This was checked for each value of n using the Kolmogorov-Smirnov test.

3.1.2.4 ESTIMATION OF COSTS AND WORKING TIME FOR THE REDUCED SAMPLING STRATEGIES

The calculations of the measuring costs associated with the use of reduced sampling strategies were performed using the following cost estimations. The cost of using an Innova photoacoustic monitor was set at € 125 per day (24 hours). This included depreciation, calibration and upkeep of the equipment. The costs associated with the work of a technician were set at € 65 per hour. The overall costs were calculated for three different sampling strategies: the 24 hour continuous sampling strategy, the 7 days continuous sampling strategy and the random grab sampling strategy. The number of 24 hour samples, 7 day samples and random grab sampling periods needed to estimate an EF with a relative error below 15 % was based on the results from section 3.1.2.3. For all strategies it was assumed that it took one hour to get to the farm where the measurements are being performed (and one hour to get back to the workplace), that four hours were needed to set up the measuring system and install the sampling lines and one hour to dismantle the measuring system and the sampling lines afterwards (Table 3.6). For the short-term strategies (e.g. 24

hour and random grab sampling), no extra visits were deemed necessary. For the 7 day sampling strategy, it was assumed that the technician had to visit the farm one extra time to check the measuring setup. A comparison was also made with a complete measuring campaign as mostly used in Flanders to determine emission factors. Such a measuring campaign consists of 2 consecutive fattening periods. For the calculations, it was assumed that each fattening period lasted 135 days. Therefore, the working hours needed to sample for 270 consecutive days are also calculated. It was assumed that the technician had to visit the farm once every week to check the measuring setup. For all cases, no working time or costs were calculated for data processing and reporting. Subsequently, the maximum number of emission factors that could be estimated using the 24 hour sampling strategy, the 7 day period sampling strategy and the random grab sampling strategy was determined, based on the calculations for the measuring costs. Again, a comparison was made with a complete measuring campaign.

3.1.3 RESULTS

3.1.3.1 EFFECT OF THE DIFFERENT SAMPLING STRATEGIES

When all possible estimates (Table 3.2) were taken into account, similar outcomes were found for the single grab (Figure 3.4), the 24 hour (Figure 3.5), the 48 hour (Figure 3.6) and the 7 day sampling strategies (Figure 3.7). Except for the single grab sampling strategy in dataset 1, when taking one estimate, the chance of obtaining an estimated EF that is within 15 % of the true EF was higher than the chance of obtaining an estimated EF that is more than 15 % higher than the true EF (Table 3.3). Except for the single grab sampling strategy in dataset 2, these sampling strategies had a higher chance of estimating an EF that is more than 15 % higher than the true EF than estimating an EF that is more than 15 % lower than the true EF, based on one estimate. For the long-term weekly grab sampling strategy (Figure 3.8 & Figure 3.9), the chance of obtaining an estimated EF that is within 15 % of the true EF was 100 % for both datasets and for all days (data not shown).

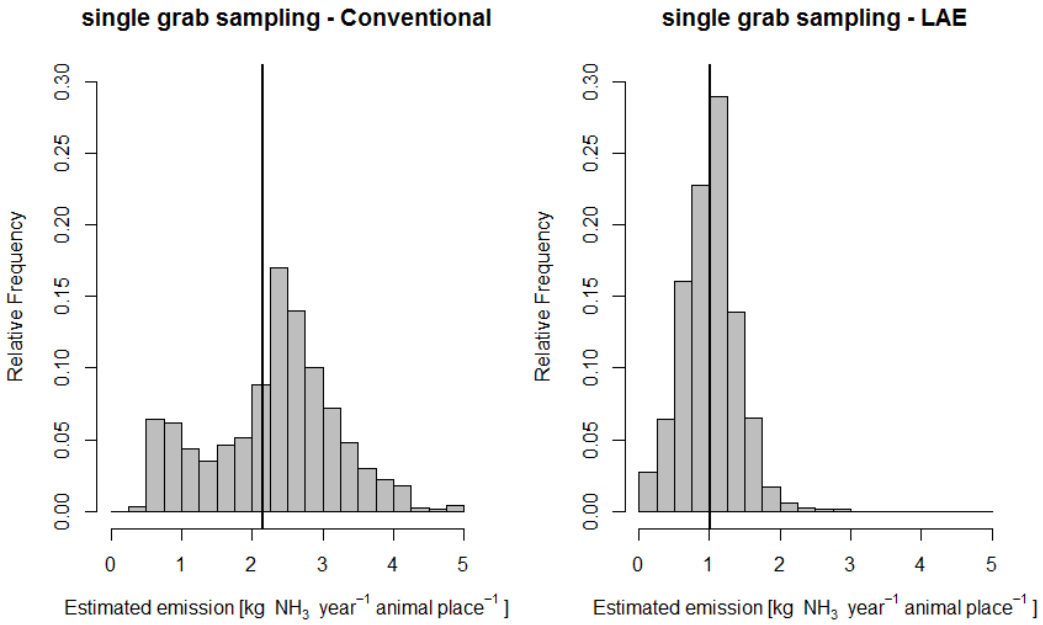

Figure 3.4. Histograms of all possible outcomes from the single grab sampling strategy for dataset 1 (left) and dataset 2 (right). The vertical lines indicate the true NH_3 EF.

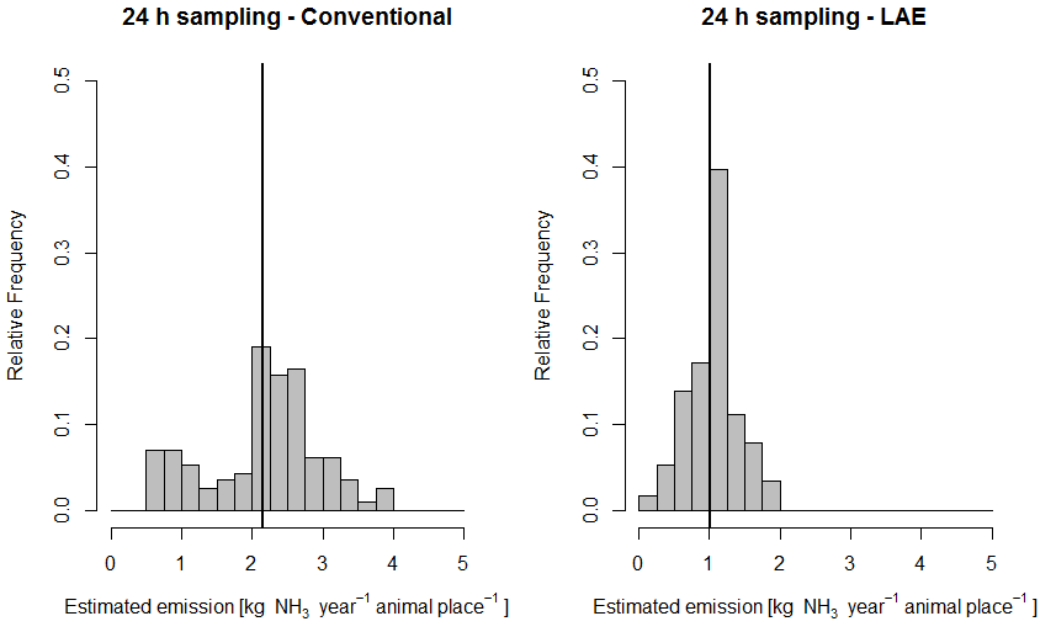

Figure 3.5. Histograms of all possible outcomes from the 24 h sampling strategy for dataset 1 (left) and dataset 2 (right). The vertical lines indicate the true NH_3 EF.

For the single grab, the 24 hour, the 48 hour and the 7 day sampling strategy, the precision (defined as the dispersion of the histograms) for the results from dataset 2 was higher than for the results from dataset 1. For the long-term weekly grab sampling strategy, no apparent difference in precision could be found between both datasets.

Table 3.3. Percentage of values lower than 85 % of the true EF, between 85 % and 115 % of the true EF and larger than 115 % of the true EF for all possible estimates for the different sampling strategies.

Sampling protocol		< 0.85 x True EF	>= 0.85 x True EF & <= 1.15 x True EF	> 1.15 x True EF
single grab sampling	dataset 1	26.8	28.6	44.6
	dataset 2	32.8	35.6	31.6
24 hour sampling	dataset 1	25.2	38.3	36.5
	dataset 2	25.0	44.0	31.0
48 hour sampling	dataset 1	22.2	40.3	37.5
	dataset 2	22.4	45.9	31.8
7 day sampling	dataset 1	25.4	41.3	33.3
	dataset 2	23.2	43.0	33.8

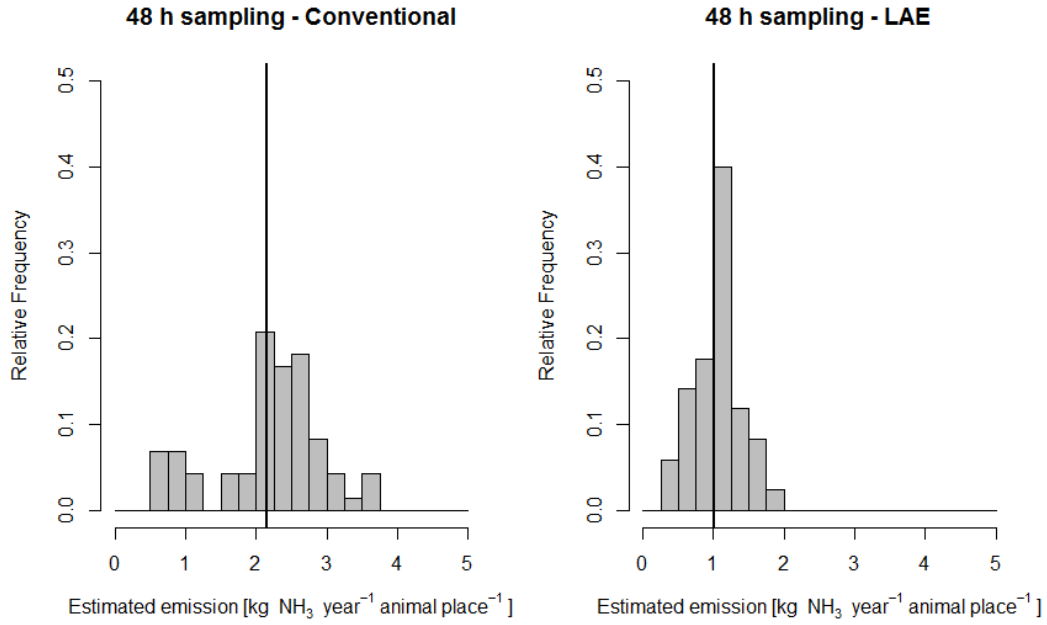

Figure 3.6. Histograms of all possible outcomes from the 48 h sampling strategy for dataset 1 (left) and dataset 2 (right). The vertical lines indicate the true NH_3 EF.

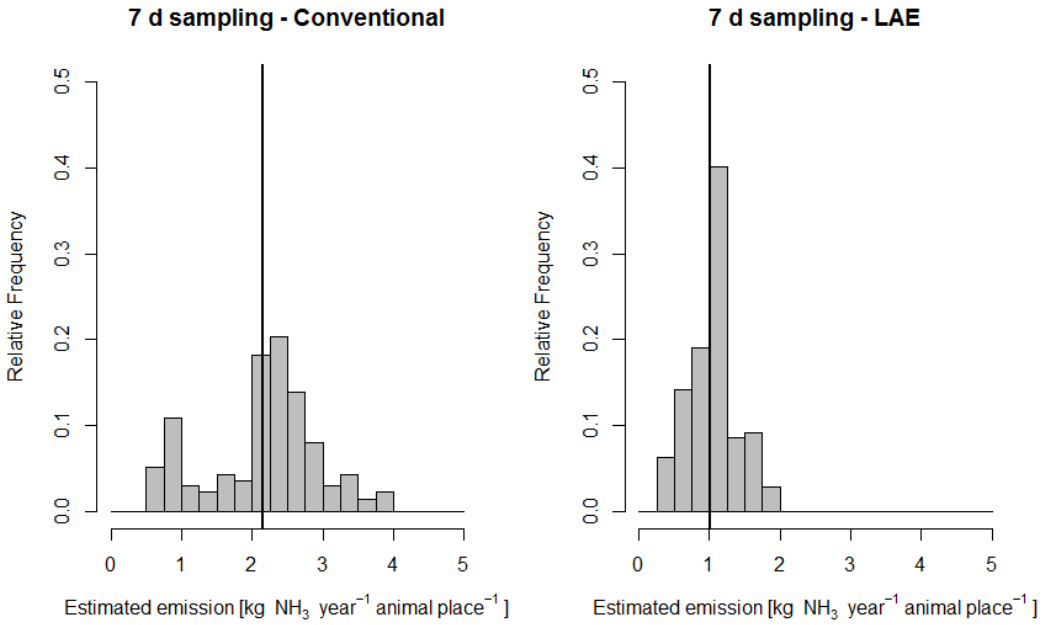

Figure 3.7. Histograms of all possible outcomes from the 7 d period sampling strategy for dataset 1 (left) and dataset 2 (right). The vertical lines indicate the true NH_3 EF.

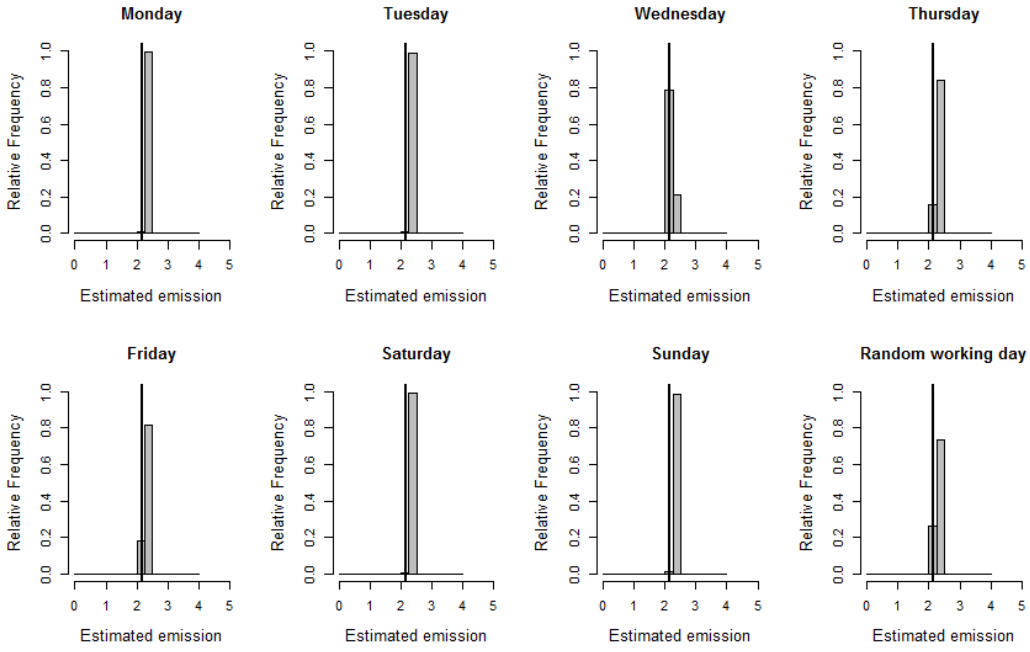

Figure 3.8. Histograms of all possible outcomes from implementing the long-term (32, 33 or 34 weeks, depending on the day of the week) weekly grab sampling strategy for dataset 1. The first seven plots show the outcome per day of the week, the last plot shows the result for a random working day. The vertical lines indicate the true NH_3 EF.

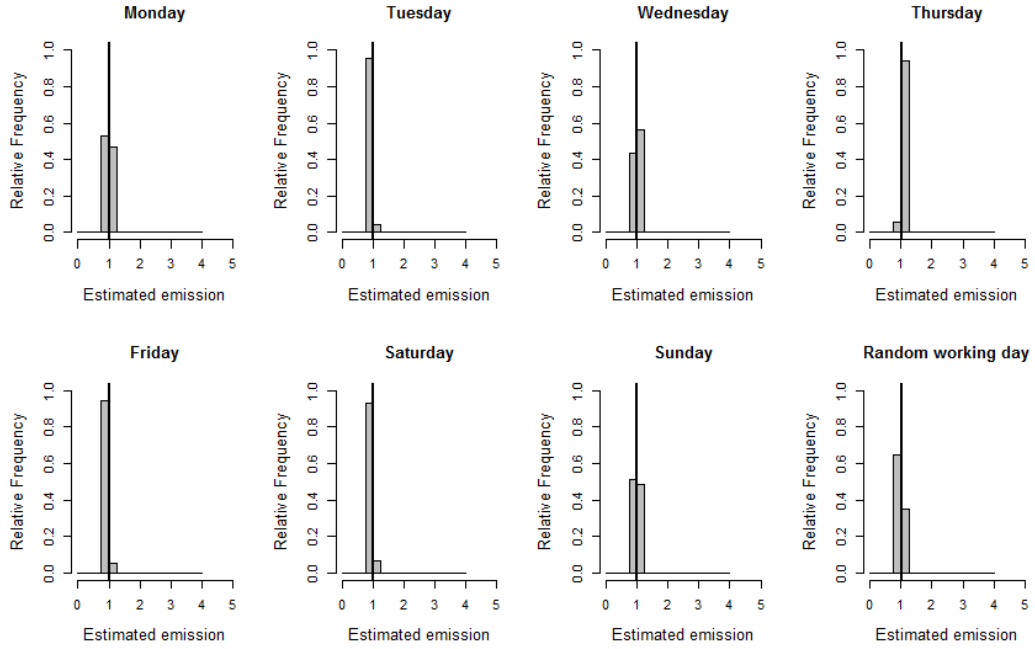

Figure 3.9. Histograms of all possible outcomes from implementing the long-term (31, 33 or 34 weeks, depending on the day of the week) weekly grab sampling strategy for dataset 2. The first seven plots show the outcome per day of the week, the last plot shows the result for a random working day. The vertical lines indicate the true NH_3 EF.

3.1.3.2 ACCURACY AS FUNCTION OF SAMPLING FREQUENCY

For each sampling strategy, it was checked for which values of *n*, the 1000 relative errors (ε) per value of *n* were normally distributed (Table 3.4). If so, for these values of *n*, the uncertainty bounds were the bounds of the 95 % confidence interval around the average.

Table 3.4. Values of *n* for which the 1000 relative errors (ε) per value of *n* are normally distributed.

	dataset 1	dataset 2
Single grab	$n>1$	all *n*
24 hour	$2<n<113$	$1<n<114$
48 hour	$3<n<70$	$1<n<83$
7 day	$n>2$	$1<n<27$
long term weekly	$n>32$	$n<5$ & $n>22$

Performing a sampling campaign based on 10 random grab samples (n = 10), 24 hour, 48 hour or 7 day sampling periods or weekly grab samples for 10 consecutive weeks yielded an estimate of the EF with the uncertainty bounds between -39 % and 48 % (Table 3.5). If the

number of random 24 hour, 48 hour or 7 day sampling periods was increased, the relative error decreased (Figure 3.10, Figure 3.11 & Figure 3.12).

Table 3.5. Relative error intervals of an estimated EF, based on 10 (*n*=10) random grab samples, 24 hour, 48 hour or 7 day sampling periods or weekly grab samples for 10 consecutive weeks.

	dataset 1	dataset 2
Single grab	between -20 % and 33 %*	between -24 % and 23 %*
24 hour	between -21 % and 23 %*	between -19 % and 24 %*
48 hour	between -19 % and 21 %*	between -16 % and 24 %*
7 day	between -15 % and 18 %*	between -12 % and 20 %*
long term weekly	between -37 % and 48 %	between -39 % and 35 %

* The relative error has a 95 % chance of lying between these values.

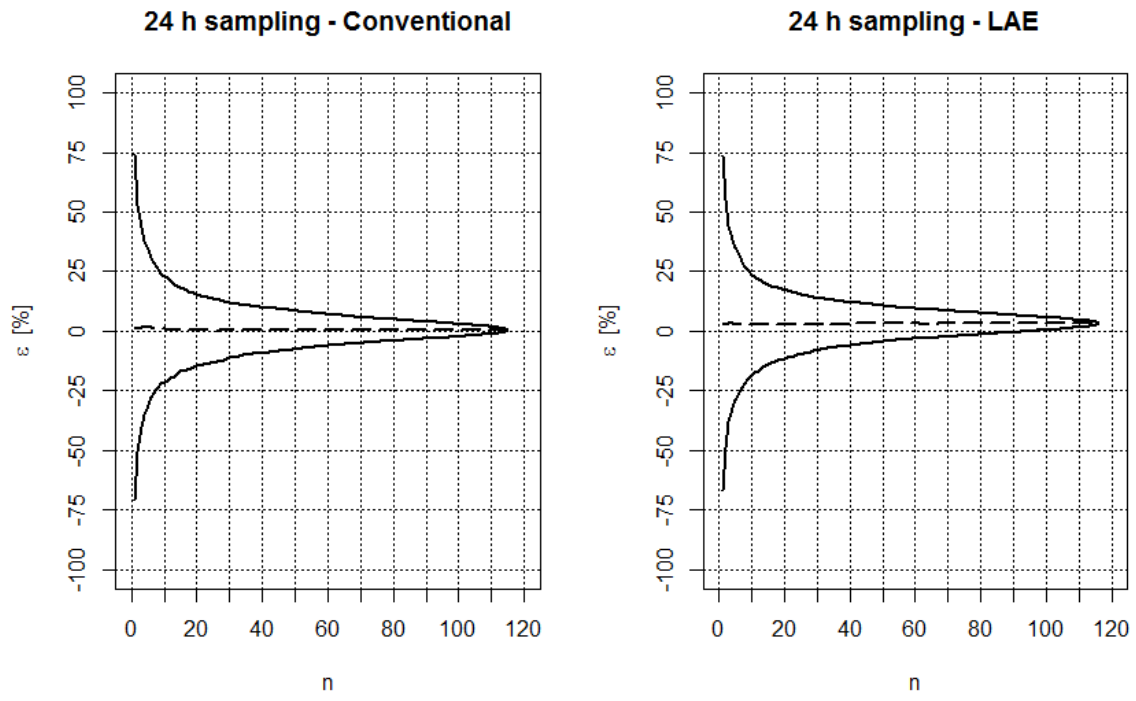

Figure 3.10. Uncertainty bounds of the relative error (ε) as a function of the number (n) of 24 hour sampling periods, based on 1000 iterations. The bold dotted line represents the mean relative error as a function of the number (n) of 24 hour sampling periods

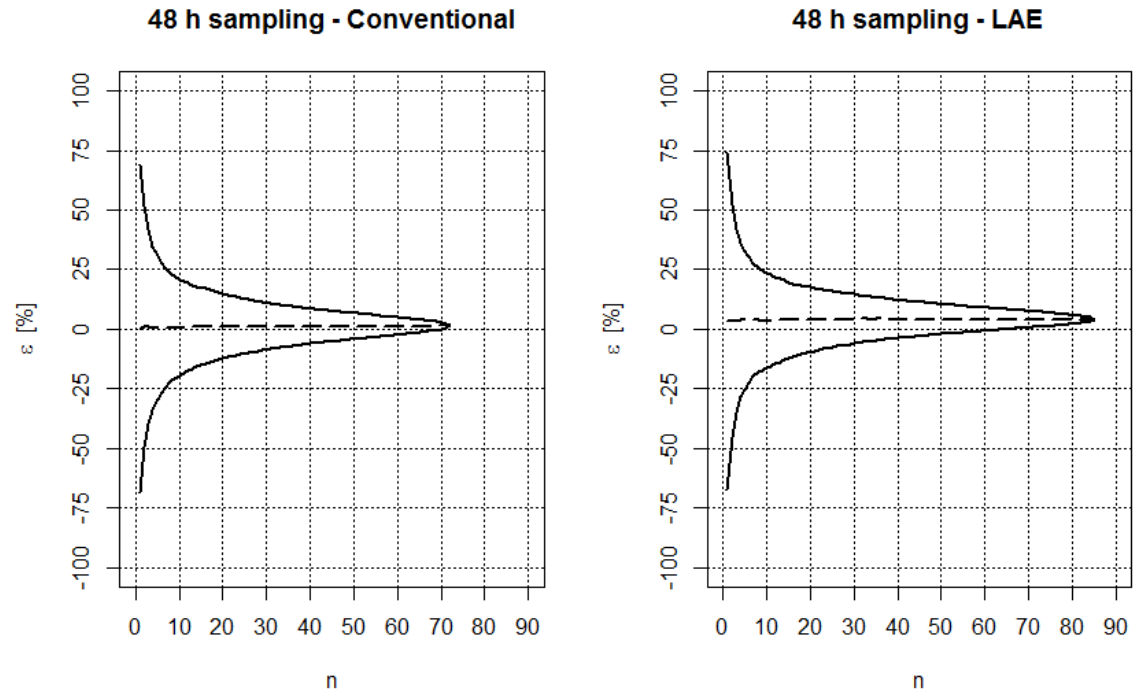

Figure 3.11. Uncertainty bounds of the relative error (ε) as a function of the number (n) of 48 hour sampling periods, based on 1000 iterations. The bold dotted line represents the mean relative error as a function of the number (n) of 48 hour sampling periods

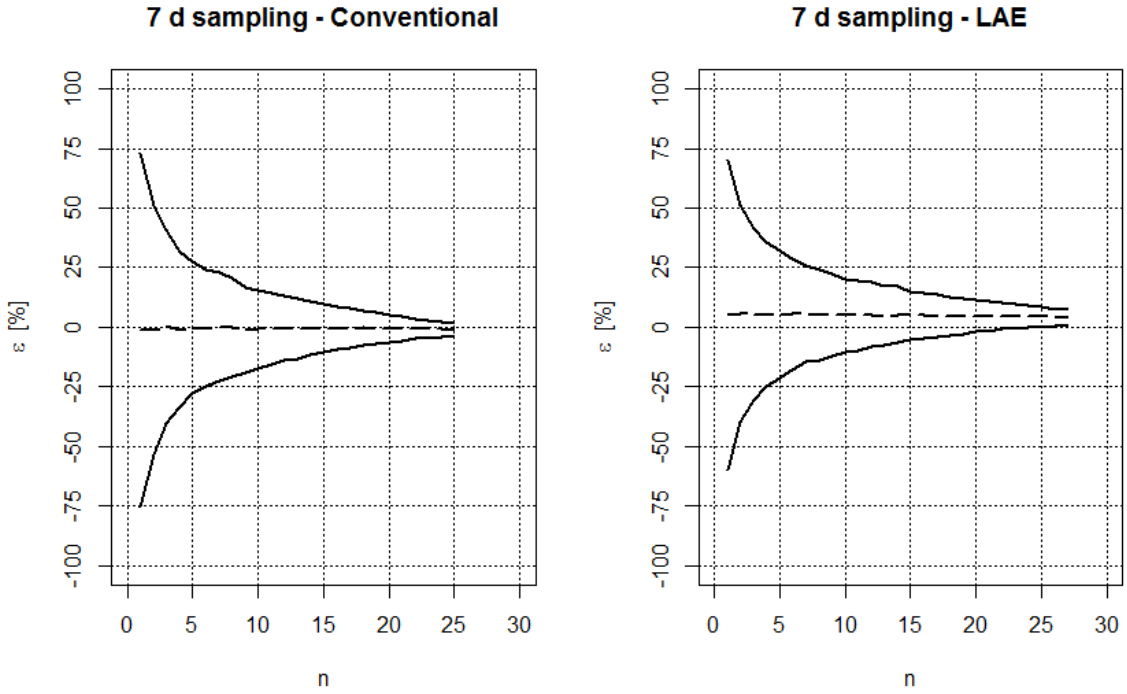

Figure 3.12. Uncertainty bounds of the relative error (ε) as a function of the number (n) of 7 day sampling periods, based on 1000 iterations. The bold dotted line represents the mean relative error as a function of the number (n) of 7 day sampling periods

In order to reduce the relative error to maximum ± 15 % for the single grab sampling strategy (Figure 3.13), 84 (dataset 1) and 27 (dataset 2) random grab samples needed to be taken. Taking 100 random grab samples reduced the relative error further down to -1 % and 14 % (dataset 1) and -8 % and 7 % (dataset 2). If more grab samples were taken, the accuracy only improved slightly. In order to reduce the relative error to maximum ± 15 % for the long-term weekly grab sampling strategy (Figure 3.14), weekly grab samples had to be taken during 32 (dataset 1) and 28 (dataset 2) consecutive weeks.

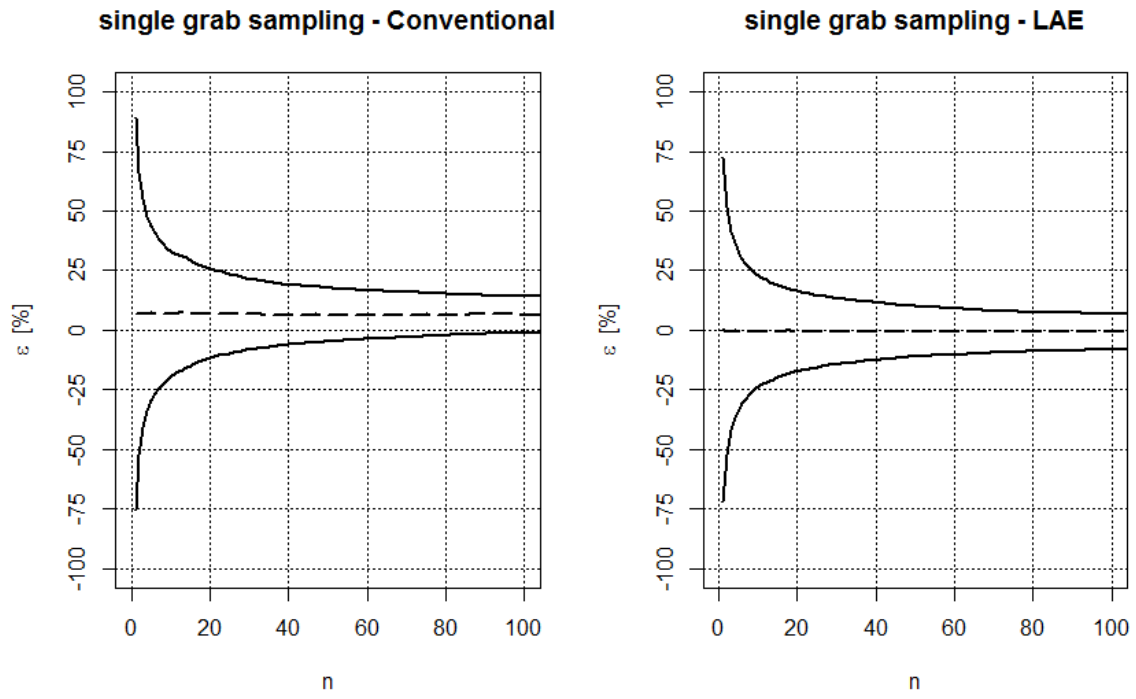

Figure 3.13. Uncertainty bounds of the relative error (ε) as a function of the number (n) of random grab samples, based on 1000 iterations. The bold dotted line represents the mean relative error as a function of the number (n) of random grab samples.

For the 24 hour and 48 hour sampling strategies (Figure 3.10 & Figure 3.11), all 4 graphs showed a small positive bias. This bias is due to the fact that these sampling campaigns only take into account hour measurements started on specific days, while the true EF is based on continuous data over all days of the week. Furthermore, the lower and upper uncertainty bounds coincided for all 4 graphs. This is because, for each of the 1000 iterations, there is only one way to sample the maximum of n 24/48 hour sampling periods. In contrast to the previous sampling strategies, the lower and upper uncertainty bounds did not coincide for both graphs (Figure 3.12) when the maximum number of 7 day periods was reached. This is

because a 27 or 28 week sampling campaign can start on each working day of the week. Therefore, the estimates between mimicked campaigns will differ depending on which day of the week the campaign started. If the maximum number of single grab samples would be taken into account, the lower and upper uncertainty bounds would coincide for both graphs (Figure 3.13). As for the 7 day sampling strategy, the lower and upper uncertainty bounds for the weekly grab sampling strategy (Figure 3.14) did not coincide since the number of possible ways to conduct a weekly grab sampling campaign of 35 (dataset 1) or 34 (dataset 2) consecutive weeks is almost infinite.

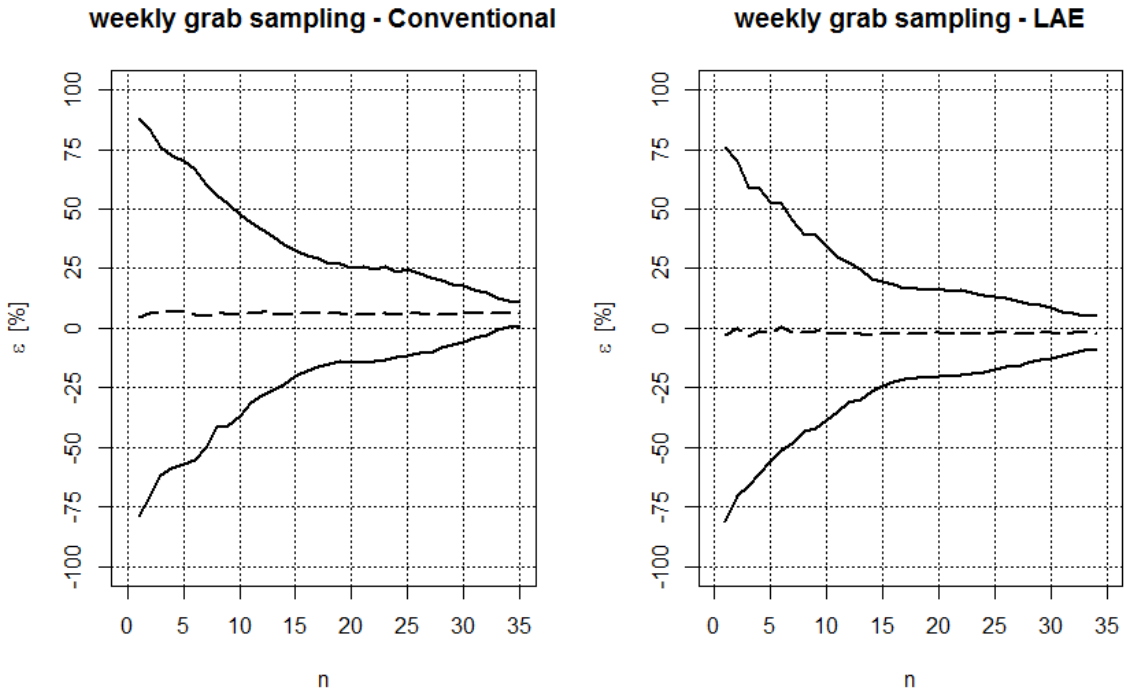

Figure 3.14. Uncertainty bounds of the relative error (ε) as a function of the number (n) of weekly grab samples, based on 1000 iterations. The bold dotted line represents the mean relative error as a function of the number (n) of weekly grab samples.

3.1.3.3 ESTIMATION OF COSTS AND WORKING TIME FOR THE REDUCED SAMPLING STRATEGIES

The working time needed to take one 24 hour sample, one 7 day sample, one random grab sample and one complete measuring campaign, taking into account transport, set-up, (control) and dismantle, was calculated as well as the number of days an Innova monitor is needed (Table 3.6).

Table 3.6. Work and measurement hours needed for one 24 hour period, one 7 day period, one random grab sample and one full measuring campaign.

	Hours needed			
	24 hour sampling strategy	7 day sampling strategy	Random grab sampling strategy	Complete measuring campaign[1]
Technician				
Set-up	4	4	4	4
Control	0	2	0	76
Dismantle	2	2	2	2
Transport	4	6	2	80
Innova				
Days needed[2]	1,33	7,33	0,38	270,33

[1] It was assumed that a complete measuring campaign consisted of 240 days.

[2] Not only the measuring period, but also the time during transport and set-up/dismantling is taken into account.

Based on Table 3.6, the costs were calculated for the different sampling strategies, taking into account the number of periods needed to obtain a good estimate (i.e. an estimate of the emission factor with a relative error below 15 %), calculated in section 3.1.3.2 (Table 3.7). The 24 hour sampling strategy was the most cost-effective one to assess the EF of the conventional housing system (dataset 1, *n = 21*) in this study. The use of this strategy gave a 61 % reduction in costs compared to a complete measuring campaign. Using the 7 day sampling strategy for dataset 1 (*n = 13*) would result in higher costs as compared to the 24 hour sampling strategy, but still would give a 46 % reduction in comparison with the complete measuring campaign. Since the number (*n* = 84) of grab samples that had to be taken to get a good estimate was high, performing a random grab sampling strategy was more expensive than a complete measuring campaign for this dataset. However, for the LAE housing system (dataset 2), the number (*n* = 27) of grab samples, needed to obtain a good estimate, was much lower in this study. Therefore, this sampling strategy was the most cost-effective one and gave a 65 % reduction in costs compared to a complete measuring campaign. The use of the 24 hour sampling strategy (*n = 24*) or the 7 day period strategy (*n = 15*) also reduced the costs of the sampling campaign considerably.

Table 3.7. Estimated costs associated with one 24 hour period, one 7 day period, one random grab sample and one full measuring campaign and with a measuring campaign with a good1 estimate.

	Costs (€)			
	24 hour sampling strategy	7 day sampling strategy	Random grab sampling strategy	Complete measuring campaign[2]
Technician				
Set-up	260	260	260	260
Control	0	130	0	4940
Dismantle	130	130	130	130
Transport	260	390	130	5200
Innova[3]	166	916	47	33791
Total for 1 period	816	1826	567	44321
Total for good[1] estimate				
Dataset 1	17141	23741	47618	44321
Dataset 2	19590	27394	15306	44321

[1] A good estimate was defined as an estimate of the emission factor with a relative error below ± 15 %.

[2] It was assumed that a complete measuring campaign consisted of 240 days.

[3] Not only the measuring period, but also the time during transport and set-up/dismantling is taken into account.

The maximum number of barns for which an emission factors could be estimated with one Innova monitor, based on Table 3.6 and the number of periods needed to estimate an emission factor with a relative error below 15 %, was 1.35 for a complete measuring campaign of 240 days. For the 24 hour sampling strategy, there are 208 possible 24 hour periods in a year, fulfilling the criteria as determined in section 2.1. Depending on the dataset, the Innova is needed 42 (dataset 1) or 48 (dataset 2) days to estimate an emission factor with a relative error below 15 %. When time for repairs, maintenance and for calibration is not taken into account, 5 (dataset 1) or 4.3 (dataset 2) emission factors can be estimated each year with one Innova monitor following the 24 hour period sampling strategy. For the 7 day period sampling strategy and when assuming that no new 7 day period could start on the same day as another 7 day period ended, there are 43 possible 7 day periods for the year 2014. Therefore, 3.3 (dataset 1) or 2.9 (dataset 2) emission factors

can be estimated each year with one Innova monitor following the 7 day period sampling strategy. Finally, for the random grab sampling, assuming that maximum one grab sample can be taken each day, there are 260 possible days in a year to take grab samples. Therefore, 3.1 (dataset 1) or 9.6 (dataset 2) emission factors can be estimated each year with one Innova monitor following the grab sampling strategy.

3.1.4 DISCUSSION

3.1.4.1 EVALUATION OF THE DIFFERENT SAMPLING STRATEGIES

Estimations of the NH_3 EF based on just one single grab sample, one 24 hour period, one 48 hour period or one 7 day period exhibited a relatively high chance (> 30 %) to overestimate the true EF. At first glance it seems striking that generally, with the exception of the single grab sampling strategy in dataset 2, there was a higher chance to overestimate the true EF than to underestimate it.

When taking a closer look at the initial dataset, multiple explanations for this outcome can be found. It can be seen in Figure 3.1 that the NH_3 emission increased more drastically towards the end of the fattening periods for both housing systems (although this was not equally pronounced in all fattening periods). For example, the NH_3 emission in fattening period 1 for the conventional housing system (Figure 3.1A) increased rather abruptly between approximately day 30 and day 50. When only one 24 hour or 48 hour sample is taken during that fattening period, there is a higher chance for that sample to be taken in a period with relatively higher NH_3 emission since there are more periods with relatively higher NH_3 emission as compared to periods with relatively lower NH_3 emission. If an EF would be estimated only on the basis of that sample, the estimated EF has, consequently, a higher chance to be larger than the true EF.

The same explanation applies to the one week sampling strategy. Since there is a higher chance for a one week period to be taken in a period with higher NH_3 emission, the estimated EF during that period has a higher chance to overestimate the true EF. This explanation is also valid for the single grab sampling strategy. However, for this reduced sampling strategy, the limited time period when the sampling can be performed (between 9 a.m. and 5 p.m.) also plays a role. During these hours, the NH_3 emissions are higher than during night hours (Figure 3.15). Higher NH_3 emissions during daytime as compared to night

time have also been found by other researchers (Aarnink *et al.*, 1996; Blanes-Vidal *et al.*, 2008; Ngwabie *et al.*, 2011) and probably originate from increased urination behaviour of the pigs (Aarnink *et al.*, 1996) and higher air movements over the manure surface during daytime (Blanes-Vidal *et al.*, 2008). Since the true EF takes into account both (higher) day and (lower) night values, it is not surprising that, depending on the dataset, approximately 30 % or 45 % of the grab samples overestimated the true EF with more than 15 %.

The absolute difference between day and night values for both systems (Figure 3.15) may explain the different results between the two datasets for all short-term sampling strategies. The smaller difference between day and night values for dataset 2 (LAE) gave rise to a better precision (defined as the dispersion of the histograms) as compared to dataset 1 (conventional).

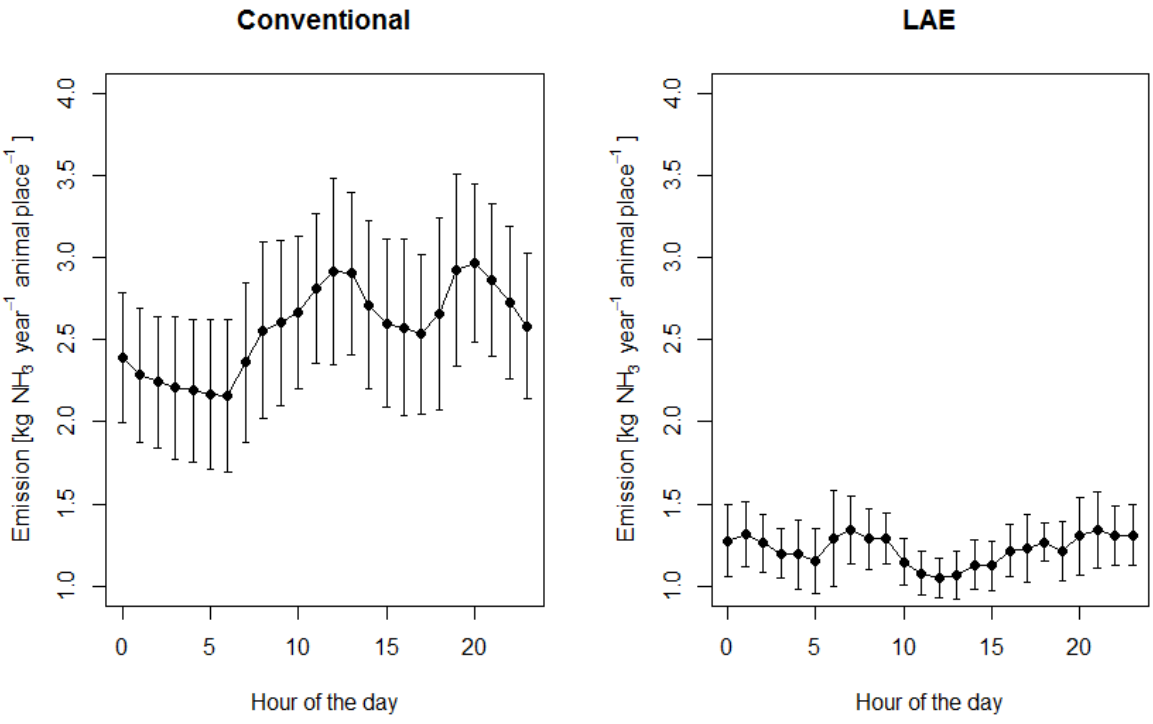

Figure 3.15. Mean emission rate for days 80 to 100 in fattening period 2 for the conventional (left) and LAE (right) housing system. Error bars indicate the standard deviations.

The long-term weekly grab sampling strategy takes a grab sample every week and is therefore better equipped to capture the increase in NH_3 emissions during a fattening period. However, as with the single grab sampling strategy, samples for the long-term weekly grab sampling strategy were only taken during daytime (between 9 a.m. and 5 p.m.).

Nevertheless, for all days and both datasets, all of the 1000 simulated long-term weekly grab sampling campaigns estimated an EF that is within 15 % of the true EF. This good accuracy for the long-term weekly grab sampling campaign probably originates from the fact that the increase in NH_3 emissions during a fattening period is included in the data here. This indicates also that taking grab samples on a regular basis (on a specific day and/or once every week) during daytime can be enough to obtain an estimate of the EF that is within 15 % of the true value.

After evaluating all sampling strategies, one can decide that the use of only one grab sample, one 24 hour sample, one 48 hour sample or one 7 day sample has a relatively high chance of overestimating the true EF. So, much caution has to be taken when estimating an EF based on only one sample of a short-term sampling strategy. Furthermore, since the spread on the estimated EFs was larger for dataset 1, the use of only one sample is certainly not advisable in situations where large differences in NH_3 emissions between days and between hours exist. Of course, in practice, no NH_3 emissions are determined on such short periods. Therefore, in section 3.1.3.2, the accuracy as a function of the number of samples was determined. The estimated EF based on one long-term weekly grab sample campaign was, in contrast to the other sampling strategies, much more precise and accurate. With one single long-term weekly grab sampling campaign, the estimated EF is within 15 % of the true EF.

3.1.4.2 ACCURACY AS FUNCTION OF SAMPLING FREQUENCY

It was shown that the long-term weekly grab sampling strategy yielded the best estimate of the true EF. However, this sampling strategy took the maximum number of weeks into account. Since sampling for such a long period would be costly, simulations were made to determine if a shorter sampling frequency (i.e. taking grab samples for fewer weeks) would also be sufficient. Since weekly grab sampling on a random working day yielded equally good results as weekly grab sampling on a specific day of the week, it was decided to test the shorter sampling frequency for a random working day only. Decreasing the number of consecutive sampling weeks increased the relative error gradually. For 15 weeks of sampling, the relative error was between -25 % and 25 % (Figure 3.14). With less than 15 sampling weeks, the relative error increased rapidly. If grab sampling was performed for 25 consecutive weeks on a random working day, an EF was estimated with a the relative error

between -12 % and 25 % for dataset 1 and a relative error which has 95 % chance of lying between -17 % and 13 % for dataset 2. If the same number of grab samples was taken completely at random (i.e. not systematically one measurement every week), the resulting estimated EF had a relative error which has 95 % chance of lying between -10 % and 24 % (dataset 1) and between -16 % and 15 % (dataset 2). So, a similar accuracy was obtained with completely random grab samples as with grab samples every week. If the number of random grab samples or the number of consecutive weeks decreased even further, the random grab sampling strategy had a better accuracy as compared to the grab sampling each week (Figure 3.13 & Figure 3.14).

If more information on the diurnal patterns is desirable, strategies based on grab samples cannot be used. Instead, the 24 hour, 48 hour or 7 day sampling strategy has to be used. These strategies can also be used to estimate an EF if multiple 24 hour, 48 hour or 7 day periods are taken. To get an estimated EF with a relative error below 15 %, 21 (dataset 1) or 27 (dataset 2) 24 hour periods, 20 (dataset 1) or 29 (dataset 2) 48 hour periods or 13 (dataset 1) or 15 (dataset 2) 7 day periods were necessary. It can be concluded that there is no big difference in the number of 24 or 48 hour periods that have to be sampled. The number of 7 day periods needed to get a good estimate of the EF is about 40 % lower than the number of 24 or 48 hour periods.

For all the sampling strategies, investigated in this study, the mean relative error for each value of *n*, which can also be seen as the bias (an estimate of the systematic measurement error), was quite low. It was noticeable that the 24 hour, 48 hour and 7 day sampling strategies gave a higher bias for dataset 1 as compared to dataset 2. Similarly, the single grab and long-term weekly grab sampling strategies gave a higher bias for dataset 2 compared to dataset 1. The latter can be explained by the smaller differences between the day and night values for the pig unit in dataset 2, as already mentioned in section 3.1.4.1. The reason why the 24 hour, 48 hour and 7 day sampling strategies gave a lower bias in dataset 2 is less clear. Since the bias remained almost constant for each sampling strategy, the precision of these sampling strategies increased with the number of sampling cases (*n*). This is because the relative error decreased when the number of sampling cases (*n*) increased.

3.1.4.3 ESTIMATION OF COSTS AND WORKING TIME FOR THE REDUCED SAMPLING STRATEGIES

When deciding which strategy to use, the efforts and time needed to install and dismantle the measuring setup should also be taken into account. The most cost-effective method to estimate an EF with a relative error below 15 % was different for both datasets: 24 hour sampling strategy (twenty-one 24 hour periods) for dataset 1 and random grab sampling strategy (27 grab samples) for dataset 2. Since only two housing systems and two datasets were compared, it is not possible to decide if this difference is caused by the housing system or is rather a consequence of the smaller variations in NH_3 emission obtained for the second dataset.

Besides the costs benefits for the farmers and companies, reducing the sampling time also ensures a higher availability of the measuring equipment. This allows performing multiple measuring campaigns and assessments of mitigation techniques during a year. Again, different results were obtained for both datasets. For dataset 1, five emission factors could be estimated in one year using the 24 hour sampling strategy, whereas nearly double the number of emission factors could be estimated in dataset 2 using the random grab sampling strategy.

3.1.4.4 GENERAL REMARKS

All of the simulated sampling strategies in this study were entirely based on only two datasets. Therefore, the results and conclusions drawn in this study should be further tested with other datasets including other buildings. This could be verified in a follow up study where the same sampling strategies are applied to other long-term continuous datasets.

Another very important remark to make is that the sampling strategies, proposed here, are only valid to estimate an EF for a certain barn and cannot be generalised to all barns of the same housing system. This in contrast to the reduced sampling strategies proposed by Ogink *et al.* (2011) or Mosquera and Ogink (2011). These authors determined a sampling strategy based on a limited number of measurements at multiple farms, in order to take into account the between-farm variance. Furthermore, these measurements had to be spread over time.

3.1.5 CONCLUSIONS

Based on all simulations carried out in this study, one can conclude that the best strategy to get an estimated EF with a relative error below 15 % for dataset 1 is to sample during 21 periods of 24 hour. Estimates with a relative error below ± 15 % for the long-term weekly grab sample strategy are only obtained when grab samples are taken during 32 consecutive weeks. For dataset 2, only 27 random grab samples are needed to obtain an estimated EF with a relative error below ± 15 %, which is equal to the number of 24 hour sampling periods needed.

4 TECHNICAL CHALLENGES FOR ENVIRONMENTAL RESEARCH IN LIVESTOCK BUILDINGS

When performing gas, PM and ventilation experiments in animal houses, technical challenges need to be overcome and still technical problems can arise during measurements. This was also the case for the trials performed during this thesis. These challenges and problems can be the consequence of instrument-specific limitations of the measuring equipment (e.g. interfering gases or too narrow measuring range) or agriculture related challenges (e.g. presence of rats). Some problems can be easily fixed once the source of problem is found. For example, after repair of our data logger, ventilation rates were registered correctly. Other problems are far more challenging to solve. The focus of this chapter will be primarily on the limitations related to the measurement equipment used for the gas and PM measurements respectively.

4.1 GAS MEASUREMENTS

All gas measurements in this thesis were performed using an Innova photoacoustic gas monitor 1314 connected to a multipoint sampler (CBISS, A1-Envirosciences ltd., Wirral, Merseyside, UK). The photoacoustic system (PAS) used in the experiments described in this thesis is equipped to measure NH_3, CO_2, N_2O, CH_4 and H_2O and was cross-calibrated for the interference of all these gases onto the respective filters.

The overall standard uncertainty[v] on emissions in mechanically ventilated buildings, under very controlled circumstances, is reported to be between 5 to 12 % (Calvet *et al.*, 2010; Gates *et al.*, 2009). However, it is accepted that measurements of building emissions on a short time basis have an uncertainty in the range of 10 to 20 % (Calvet *et al.*, 2013). Although the precision of photoacoustic techniques to measure ammonia concentrations is estimated to be around 2.5 % (Hinz & Linke, 1998), photoacoustic infrared spectrometers can suffer from inference from other (non)-measured gases and water vapour, leading to higher uncertainties (Calvet *et al.*, 2013). It is believed that the uncertainties can even be higher than the proposed reduction targets (Hassouna *et al.*, 2013). The narrow absorption bands of the optical filters give rise to a good selectivity. However, if a gas, not intended to measure with the PAS (non-target gas), absorbs infrared (IR) light at the same wavelength as the gas, intended to measure (target gas), the total IR absorbance at that wavelength will be higher and will be totally attributed to the target gas. Consequently, the concentration of the target gas will be overestimated (= interference). If the interfering gas is measured with a separate filter, cross-compensation can be performed, correcting the interference of the non-target gas on the target gas. However, absorption of IR light of another non-target gas on the filter of the interfering gas (leading to an apparent increase in concentration of the interfering gas) would ultimately lead to underestimation of the target gas (= cascade effect) (Hassouna *et al.*, 2013). The interference between two target gases can be fully eliminated

[v] According to Calvet *et al.* (2013) uncertainty of measurement is defined as "a parameter that is associated with the result of that measurement. It characterizes the dispersion of the values that could reasonably be attributed to the quantity measured and thus has an inherent statistical basis. The parameter can be a standard deviation (*standard uncertainty*), or a confidence interval which is expected to encompass a certain fraction of the distribution of values (*expanded uncertainty*)".

by the internal cross-compensation. However, the compensation of the cascade effect cannot be addressed by the internal cross-compensation. The cascade effect can potentially be estimated by a mathematical simulation (Zhao *et al.*, 2012). Both under laboratory conditions and in practise in poultry and cattle barns, it has already been demonstrated that non-target gases can cause both overestimation through interference and underestimation through the cascade effect (Hassouna *et al.*, 2013; Zhao *et al.*, 2012). A visual example of interference and cascade effects is shown in Figure 4.1.

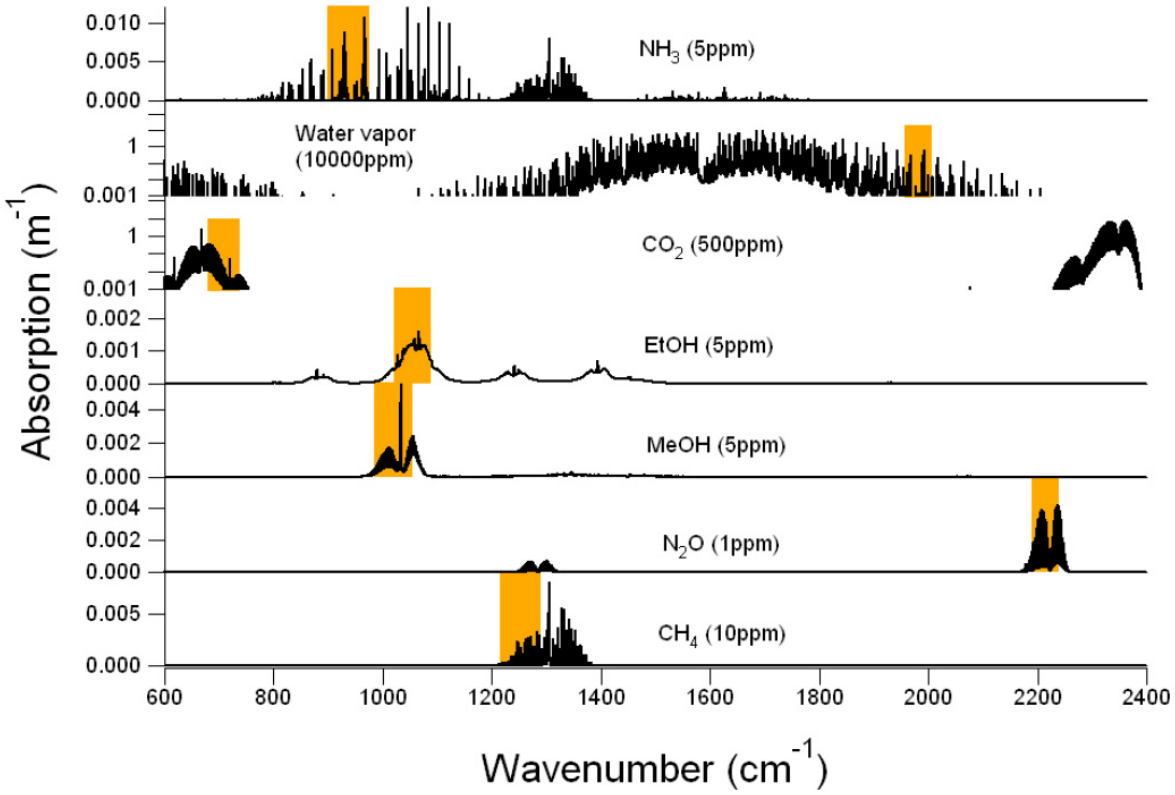

Figure 4.1. From top to bottom are the IR absorption spectra of NH_3, H_2O, CO_2, ethanol (EtOH), methanol (MeOH), N_2O and CH_4 with in orange boxes the filter centre and bandwidth for (from top to bottom) filter UA0976, SB0527, UA0982, UA0974, UA0936, UA0985 and UA0976. Situation 1: if a gas monitor is only equipped with filter UA0976 (NH_3), the presence of MeOH (in this situation a non-target gas) in the air will cause additional absorption of IR at that filter regardless of the NH_3 concentration in the air. The NH_3 concentration will be overestimated and this process is called interference. Situation 2: the gas monitor is equipped with filter UA0976 (NH_3) and filter UA0936 (MeOH) and both filters are cross-compensated for each other. If EtOH (non-target gas) is present in the air, it will cause additional absorption of IR on the MeOH filter and the concentration of MeOH will be overestimated (= interference). Because the MeOH filter is cross-compensated for NH_3, the (apparent!) contribution of MeOH to the NH_3 concentration will be deducted. However, since the MeOH concentration is overestimated (due to interference of EtOH), too much will be deducted and the NH_3 concentration will be underestimated. This is called the cascade effect (Figure adapted from Zhao *et al.* (2012)).

The interference with water vapour has previously been described by other researchers and increases linearly with increasing water vapour concentrations in a range between 0 and 30 000 ppmv for CO_2 and CH_4 and between 5 000 and 30 000 for N_2O (Yamulki & Jarvis, 1999). Since relative humidity inside livestock buildings is usually higher than 5 000 ppmv,

the non-linear relation between water vapour and N_2O below 5 000 ppmv is not really an issue.

To cope with these problems of over- and underestimation, it is necessary to include the analyser set-up, optical filters used and different management related parameters (e.g. animal feed) when publishing data (Hassouna *et al.*, 2013). Of course, one should try to select the optimal filter configuration before performing the measurements. However, post measurement adjustments using mathematical simulations are a possible alternative to correct for non-target gas interferences (Zhao *et al.*, 2012).

Besides the problems directly associated with the measurement technique, Innova gas measurements, and especially NH_3 measurements, can also be influenced by the adsorption of gases in the sampling lines. However, Shah *et al.* (2006) did not find a significant effect of tubing on NH_3 adsorption (for 1 ppmv and 10 ppmv of NH_3). In their experiment, polytetrafluoroethylene (PTFE) tubing (the kind of tubing used in this thesis) was included. Still, the high water solubility and polarity of NH_3 can be a problem, causing it to adsorb onto various surfaces in the sampling system (such as filters, valves and pumps). This would give rise to time delays ("NH_3 lag") (Rom & Zhang, 2010). This problem can arise when sequentially measuring on multiple channels. It is of particular importance especially when the difference in NH_3 concentrations between the different channels is large. A study by Rom and Zhang (2010) indicated that a substantial time delay exists when switching between channels with high and low NH_3 concentrations. They concluded that measuring periods of 12.5 to 25 minutes are needed in order to obtain reliable NH_3 concentrations (Rom & Zhang, 2010).

Besides uncertainties and errors in measuring gas concentrations, measuring the ventilation rate, in order to calculate emissions, is also associated with errors. However, the errors associated with ventilation rate are beyond the scope of this discussion. More information, both for mechanical ventilation measurements and natural ventilation measurements, can be found in Calvet *et al.* (2013).

In this thesis, problems with the Innova gas monitor were both a consequence of instrument-specific limitations and/or problems related to livestock indoor environments. The latter were associated with e.g. broken sample tubing due to rats or water condensation

in sample tubing due to electricity blackouts or defective heating ribbons. It seems impossible to totally exclude the possibility of these kinds of events to happen. As long as measurements will be performed, these problems will arise. Attention has to be paid in order to reduce the chance that these events occur. This can be done by, for example, protecting the sample tubing against rats or by making use of an uninterruptible power source to overcome short periods of time without electricity. Problems with NH_3 lag are typically encountered in every measurement campaign. These problems can be overcome by measuring multiple times on the same channel. The other instrument-specific limitations were mainly encountered during the trial in the pig nursery (chapter 2.3). During this trial, the Innova reported negative NH_3 concentrations. This is of course physically not possible. In an attempt to exclude as much potential causes as possible the dust filters, sampling tubes, length of the sample tubes and Innova gas monitor were replaced or changed between compartments with negative NH_3 concentrations and compartments with normal NH_3 concentrations. However, the problem remained for some specific compartments. This was an indication that the problems were not Innova specific but were rather compartment specific. Together with the negative NH_3 concentrations, unusually high indoor CH_4 concentrations were measured. During this trial, concentrations up to 2500 ppmv were obtained when measuring at animal height (0.8 m above the slatted floor). Such concentrations exceed the measuring range of the Innova or more specifically the range of CH_4 concentrations at which the cross-compensation with the other gases is linear. In order to exclude the possibility on a non-target gas absorbing IR light at the same wavelength of CH_4 and causing a cascade effect on NH_3 concentrations, aerial samples were taken and sent for gas chromatography (GC) analysis. The comparison between the CH_4 concentrations, measured above the manure drain and at animal height, obtained with the Innova (average over 20 minutes, approximately 6 measurements) and with GC are shown in Table 4.1.

In general, methane concentrations measured with the Innova or GC corresponded well. However, a tendency of the Innova to overestimate CH_4 concentrations at high concentrations and to underestimate CH_4 at low concentrations can be seen. This overestimation has also been reported by other authors (Osada *et al.*, 1998). The results from this small experiment show that the high CH_4 indoor concentrations as reported by the Innova are not caused by interference from a non-target gas. This also means that the

negative NH_3 indoor concentrations are not due to a cascade effect caused by interference of a non-target gas on the CH_4 filter. It is possible that the cross-compensation between CH_4 and the other gases is no longer linear at high CH_4 concentrations, leading to overcompensation of the influence of CH_4 on the other gas concentrations. This is illustrated in Figure 4.2 where the CH_4 indoor concentrations over one day are compared with the other indoor gas concentrations. From this figure, it can be seen that, when CH_4 concentration increases to very high values, the NH_3 and N_2O concentrations decrease (up to even physically impossible negative values). The effect on CO_2 concentrations is less clear.

Table 4.1. Comparison between the CH_4 concentrations obtained with the Innova and with GC.

			CH_4 concentration (ppmv)	
			Measuring device	
Sample	Compartment	Location[1]	Innova[2]	GC
1	A	manure drain	3550 ± 230	2948
2	B	manure drain	4210 ± 582	3856
3	A	animal height	810 ± 141	1229
4	B	animal height	1060 ± 148	1318

[1] animal height: 0.8 m above the slatted floor.

[2] average over 20 minutes, approximately 6 measurements.

The source of the high CH_4 indoor concentrations was found to be the exterior underground manure pit, which was in direct air contact with the compartments. This direct contact was caused by the lack of a siphon in the piping between the manure drain in the compartment and the exterior manure pit. Because it was impossible to correct the aberrant NH_3 and N_2O concentrations and there were also doubts about the correctness of the CO_2 concentrations, gas indoor concentrations were not taken into account when comparing different feed forms and grinding intensities (chapter 2.3).

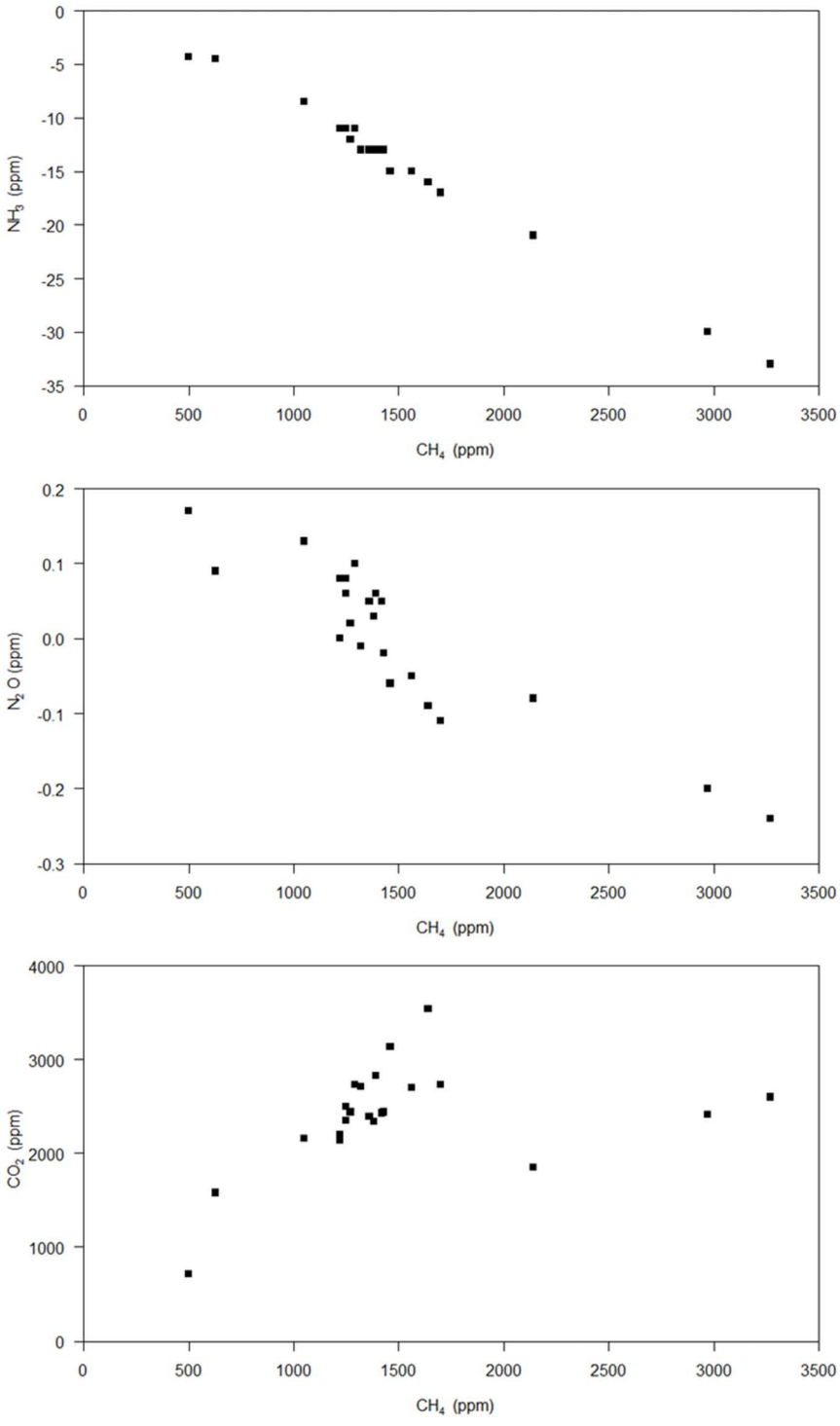

Figure 4.2. Correlation of the observed CH_4 indoor concentrations with the observed indoor concentrations of NH_3, N_2O and CO_2 at high CH_4 indoor concentrations. Data from August 8, 2013 10 a.m. till August 9, 2013 9 a.m.

From the experiences gained during our measuring campaigns, one can conclude that it is very important to minimise the chance of problems related to agricultural settings to happen. Furthermore, further research should be performed on determining the measuring

ranges of the different gases, measured by the Innova, and the ranges in which the cross-compensation between these different gases is still performed correctly. Ideally, to measure in an agricultural environment, the gas detection systems should be capable of measuring continuously and fast, chemically inert and durable, portable and remotely accessible, preferably at a low investment and operating cost. As long as such a method does not exist, control measurements will have to be performed in order to check the proper functioning of the measuring devices. An overview of the capabilities, limitations and applicability to NH_3 and GHGs in an agricultural environment of the currently available measurement methods can be found in a recent review by Hu *et al.* (2014).

4.2 PM MEASUREMENTS

The main problem associated with PM measurement inside livestock buildings, are the high PM concentrations encountered inside livestock buildings as compared to ambient air. This makes it almost impossible to get good results with a gravimetric sampling instrument with an impaction plate pre-separator (IPS), the reference instrument for measuring PM in ambient air. High PM concentrations lead to overloading of the IPS, causing larger particles to bounce off onto the filter, resulting in an overestimation of the PM concentrations (Zhao *et al.*, 2009). Buser *et al.* (2001) suggested maximum PM_{10} (138.3 µg m^{-3}) and $PM_{2.5}$ (11.9 µg m^{-3}) concentrations at which the reference instrument could be used in an agricultural environment. Zhao *et al.* (2009) showed that a $PM_{2.5}$ IPS in a poultry house gets overloaded in less than an hour, while a PM_{10} IPS did not get overloaded after 24 hours. Switching to another type of pre-separator (e.g. a cyclone pre-separator) could potentially solve this problem (Zhao *et al.*, 2009). The lack of equivalence between a reference instrument and the Grimm spectrometers (Van Ransbeeck *et al.*, 2013b) for $PM_{2.5}$ and PM_1 concentrations from livestock buildings can be explained by the fact that the PM indoor concentrations are usually higher than the values proposed by Buser *et al.* (2001). This leads to overloading of a $PM_{2.5}$ IPS at a very short time. Van Ransbeeck *et al.* (2013b) did find equivalence between these two devices for PM_{10} concentrations from livestock buildings although slightly higher PM_{10} concentrations were measured with the spectrometers (Van Ransbeeck *et al.*, 2013b). Comparison of a PM_{10} gravimetric cyclone sampler and another

light scattering device (DustTrak aerosol monitor, TSI Incorporated, Shoreview, MN, U.S.A.) showed much lower PM_{10} concentrations measured by the light scattering device (Cambra-López *et al.*, 2012).

Because of the many advantages (real-time and continuous measurements, particle size distribution, less overloading problems and portable) associated with the use of spectrometers, we used Grimm spectrometers and GrayWolf Particle Counters in our experiments. The GrayWolf Particle Counters were only used in the first trial (chapter 2.1). The latter showed problems due to overloading of the protective filters. Therefore the measuring interval was increased from 1 to 15 minutes and the instruments were no longer used in future trials. The main problem associated with the use of the Grimm spectrometers, was the breakdown of the pump (due to the high PM concentrations). The silicon beads also had to be frequently replaced (due to high relative humidity). Therefore, this device could not measure autonomously for a long time. It is also important to notice that spectrometers (or light scattering devices) are factory calibrated with a "standard" type of PM. This "standard" type can differ from PM found in livestock buildings. In particular, differences in particle shape, size and density are expected. Therefore, sampling bias may occur (Cambra-López *et al.*, 2012).

There is still a need for a reference method for sampling indoor PM concentrations and probably even more specific for sampling in an agricultural environment. As long as this method is not available, results from different studies should be compared with caution.

5 GENERAL DISCUSSION

5.1 INTRODUCTION

Intensification of agriculture and more specifically livestock farming, has led to increased pollutant (NH_3, CO_2, CH_4, N_2O and PM) emissions at a local level, thus increasing the impact. This has increased the need for mitigation techniques in order to decrease the adverse effects on the environment and human health. In an ideal world, these mitigation techniques should take into account the relations among the different pollutants and the way the different pollutants can affect each other. This could then lead to mitigation techniques which reduce a wide range of pollutants. Furthermore, preference should be given to affordable (source-oriented) mitigation techniques inside the barn which also improve the quality of the indoor climate. However, mitigation techniques mainly focus at a single pollutant and, since the reduction potential of (source-oriented) mitigation techniques inside the barn is typically lower, more expensive end-of-pipe techniques are frequently used.

Therefore, the aim of this thesis was to investigate the influence of three source-oriented techniques on a wide range of pollutants by a multi-pollutant research approach and to examine the possible relations between the different pollutants. The assessment of reduced sampling strategies for gaseous emissions was also part of this thesis. This chapter starts with an assessment of the effectiveness of the tested source-oriented techniques and a number of reasons why source-oriented techniques should be further investigated in the future. Next, the advantages, both in time and money, of reduced sampling strategies are discussed. At the end of this chapter, an overview is given of possible topics for further research with a focus on source-oriented techniques and reduced sampling strategies.

5.2 EFFECTIVENESS AND RELEVANCE OF SOURCE-ORIENTED TECHNIQUES

In this dissertation, we could not find a positive effect of the studied cleaning protocols, housing systems (chapter 2.1) and diets (chapter 2.3) in reducing pollutant concentrations in pig barns. A more elaborate cleaning protocol did not result in significantly lower indoor pollutant concentrations and therefore, in this case, cannot be seen as an effective management strategy to reduce indoor concentrations (and most probably also emissions). Nevertheless, it can be expected that the extra steps in the more elaborate cleaning protocol, especially the disinfection step, reduces the number of bacteria inside livestock houses. However, other studies with daily fogging of disinfectants in a pig farrowing - weaning room did show a reduction of fungal spores, but not a reduction in airborne aerobic bacteria (Costa *et al.*, 2014). Conversely, another study by Mannion *et al.* (2007) has shown that the application of cleaning and disinfection at pig farms can reduce the level of *Enterobacteriaceae* on the floor of the pens although the feeders and drinkers contained as many, or even higher numbers of *Enterobacteriaceae* after cleaning as compared to before cleaning. This could be the consequence rather of careless disinfection practices or due to the difficult accessibility of some edges of the feeders and drinkers. It was also possible that the use of pressure washers on the floor led to splashing of contaminated water and faeces onto the feeders and drinkers. Furthermore, cleaning practices increased the *Salmonella* contamination on farms with already high *Salmonella* prevalence (serological *Salmonella* prevalence of more than 50 % as determined by meat juice ELISA on finishing pigs at slaughter), but reduced the contamination of farms with low *Salmonella* contamination (serological *Salmonella* prevalence of less than 10 % as determined by meat juice ELISA on finishing pigs at slaughter). The authors did not give a further explanation for this remarkable finding (Mannion *et al.*, 2007). Recent research also pinpointed the drinking-nipples on a broiler farm as critical locations for cleaning and disinfection (K. Luyckx, ILVO, personal communication; Luyckx *et al.*, 2013).

In this dissertation, the use of an officially approved low-ammonia-emission housing system did not result in significantly lower NH_3 indoor concentrations as compared to a conventional housing system. This indicates that it is not solely the housing system that determines the NH_3 concentrations, but that other factors also play an important role. In this respect, management is probably important although the influence of management

practices on the emission factor of a given housing system was not investigated in this dissertation. The lack of effect on the other pollutants, except for the obtained reduction in CH_4 indoor concentrations, could have been expected since this housing system was not designed to reduce these pollutants. However, if future legislation requires a reduction of PM, it seems unlikely that this housing system will provide solutions.

Surprisingly, feeding pellets instead of meal to piglets dramatically increased indoor PM concentrations in the study conducted in this dissertation. This is in contradiction with previous research (Bundy & Hazen, 1975; Li *et al.*, 1993; Zeitler *et al.*, 1987), which was however conducted more than 20 years ago. Questions can be raised regarding the comparability of the dust monitors in the present and the older studies and their ensuing results. Furthermore, the results from the other studies are not always unambiguous. Zeitler *et al*. (1987) for example found differences in PM for floor feeding, but not for feeding with self-feeders, which was the type of feeders used in the study in this dissertation. With the exception of this study, recently no studies reported the influence of feed form on indoor PM concentrations. The feed efficiency for piglets on the pelleted diets however was much better as compared to the piglets on the meal diets. This makes it difficult to give proper advice to the livestock farmers. As long as no specific legislation regarding PM emissions or PM indoor concentrations in an agricultural environment or, more specifically, inside livestock houses are in force, farmers probably will favour the use of pellets for economic reasons. Because of the better feed efficiency (G:F = 0.77), growing a piglet from 8 to 25 kg required in our study 22.1 kg of pelleted feed compared to 26.2 kg of meal feed (G:F = 0.65). With a feed cost of 341 €/tonne (situation May 2014 in Flanders (Landbouw en Visserij,)) and a pelleting cost of approximately 7 €/tonne (I. Peeters, Aveve, personal communication), feeding pellets to a piglet (from 8 till 25 kg) would reduce the feed cost with 1.24 € per pig. It must be noted that the improvement in feed efficiency (+ 18 %) for pelleted diets as compared to meal diets found in this thesis and the G:F of both the meal and pelleted diets are high. The G:F of meal and pelleted diets are lower in other studies and improvements in G:F for pelleted diets are mostly around 5 % (Table 5.1). However, these improvements are almost never statistically significant. The observed differences can be a consequence of differences in ingredients of the diets, way of pelleting, genetics of the animals, farm management or other factors. This also implies that the differences found in PM indoor

concentrations between the diets can be different for diets with for example other ingredients or other pelleting techniques. Further research will be needed to validate the high difference in PM indoor concentrations for pelleted versus meal diets.

Table 5.1. Effect on feed form on G:F ratio in different studies.

Diet	G:F[1]	Improvement	Animals	Reference
Coarse meal	0.55		Piglets	(Grosse Liesner *et al.*, 2009)
Very coarse Pellet	0.58	+ 5.8 %		
Coarse pellet	0.60	+ 8.9 %		
Fine pellet	0.58	+ 6.4 %		
Very fine pellet	0.63	+ 15.8 %		
Meal	0.39[a]		Growing pigs	(O'Doherty *et al.*, 2001)
Pellet	0.41[b]	+ 4.9 %	(30 kg - 60 kg)	
Meal	0.33		Finishing pigs	(O'Doherty *et al.*, 2001)
Pellet	0.35	+ 4.6 %	(60 kg - 88 kg)	
Meal	0.52		Starting pigs (20 - 40 kg)	(Vande Ginste & De Schrijver, 1998)
Pellet	0.51	- 2 %		
Meal	0.39		Growing pigs (40 - 70 kg)	(Vande Ginste & De Schrijver, 1998)
Pellet	0.41	+ 5.8 %		
Meal	0.30	+ 4.7 %	Finishing pigs (70 - 100 kg)	(Vande Ginste & De Schrijver, 1998)
Pellet	0.31			
Meal	0.65[a]		Piglets	Chapter 2.3
Pellet	0.77[b]	+ 18.5 %	(8 - 23 kg)	

[1] G:F ratios without a superscript are not significantly different (P > 0.05).

As already discussed in chapter 2.3, the health of the farmer and workers in livestock buildings can be affected when exposed to high PM concentrations. The effects of high PM concentrations on the performance and the health of pigs are at the moment not totally clear (Chiba *et al.*, 1985; Michiels *et al.*, 2015; Takai *et al.*, 1995; Wathes *et al.*, 2004), but it

can be expected that lower PM concentrations are beneficial for the pigs. If the findings of our study are confirmed on other feeds, a difficult balance between economic reasons on the one hand and health and indoor air quality (and associated emissions) on the other hand exists for the choice of feeding pellets or meal diets to the pigs. Although the use of pellets gave rise to higher PM concentrations (and emissions), less pelleted feed was needed by the pigs to gain weight. Since the production of animal feed also has an environmental impact, this is another important parameter to include. Ideally, modifications of the pelleted feeds could lead to reduced PM generation while retaining good feed efficiency.

The tested source-oriented techniques in this thesis did not result in significant reductions of pollutant concentrations. However, as described in chapter 1, other source-oriented techniques, and more general mitigation techniques inside the barn, have shown to be effective in the reduction of pollutant concentrations and/or emissions. One of the great advantages of the source-oriented techniques which are based on management (e.g. choice of feed, manure management or cleaning protocol), is that most of these techniques can be applied to a large group of farms, regardless the housing system. For example, reducing the crude protein level in the feed can be accomplished on virtually every farm, while end-of-pipe techniques (e.g. wet air scrubbers) can only be used for farms with a central extraction point. This can be a very important aspect in Flanders, where upcoming legislation (e.g. PAN, see 1.3 Policy and legislation) will force a considerable group of farmers to reduce their pollutant emissions (in the case of PAN: NH_3 emissions). Since the number of end-of-pipe techniques that can be implemented at an existing farm is rather limited (mainly air scrubbers) due to technical limitations, it is very important that farmers can choose from (a combination of) source-oriented techniques in order to keep up with legislation. Second, another advantage of management-based source-oriented techniques is that they can be combined at one farm at a relatively low cost. For example, the reduction of the crude protein level could be combined with another management-based source-oriented technique (e.g. frequent removal of manure from the pit). In that way, the relatively small individual reduction percentages (as compared with end-of-pipe techniques) from each individual management-based technique can be combined to get an overall acceptable

reduction percentage. However, in practice it is not always easy to quantify those individual reduction percentages. This is partly due to the fact that the application of source-oriented techniques tends to result in small reduction percentages which can be masked by other influencing parameters. Therefore, when assessing the overall reduction percentage of combined source-oriented techniques, it will be very difficult to determine the individual contribution of each technique. Third, as already stated several times before, source-oriented techniques have a positive influence on the indoor climate. This can improve the working conditions for the farmer and living circumstances for the animals. Furthermore, if the hypothesis of the formation of secondary PM inside livestock buildings is confirmed, reducing indoor NH_3 concentrations will also have a positive impact on indoor PM concentrations. With all these advantages in mind, it remains useful to investigate the effectiveness of source-oriented techniques in the future.

5.3 REDUCED SAMPLING STRATEGIES

Measuring NH_3 concentrations and ventilation rates to determine emission factors is an essential process, both from a regulatory as from a scientific point of view. However, performing these measurements can be costly. The high costs associated with these measurements might be a hurdle for farmers and companies to develop innovative housing systems. Therefore, reduced sampling strategies, which estimate an emission factor on a reduced number of sampling days while keeping the error on the estimated emission factor at acceptable levels, could be a good alternative. In this dissertation (chapter 3), a number of reduced sampling strategies were tested and evaluated. These reduced sampling strategies determined the number of random periods needed to estimate an emission factor which had a relative error below 15 % without taking into account parameters that influence NH_3 emissions (e.g. temperature or total pig weight). It is possible that less 24 hour periods, 7 day periods or random grab samples could be used if these periods were taken at regular intervals or at predefined time points. Instead of measuring on one farm, it would also be possible to measure at multiple farms. Doing so, the between-farm variance would be taken into account and the resulting estimated EF would then be valid for all barns of the same housing system.

The estimations made in chapter 3 show that the use of reduced sampling strategies can considerably reduce the cost of the measurements as well as the time needed to perform them. Hence, it becomes possible to estimate multiple emission factors in one year with the same equipment. However, these calculations were based on simulations, performed on only two datasets. More simulations on multiple datasets should be performed in order to determine whether the obtained results can be more widely applicable. Extra simulations on new datasets could also give more insight in the differences found between dataset 1 and dataset 2. Are these linked with a specific housing system or rather the consequence of the smaller variations in NH_3 emission obtained for the second dataset?

5.4 FUTURE RESEARCH

At the end of this thesis, plenty of challenges and opportunities remain to be solved.

In order to perform correct and accurate measurements of gases and PM, it is necessary to keep searching for **new techniques and reference methods**. The **applicability** of already available instruments (e.g. Innova gas monitors) should be further investigated in order to determine the circumstances in which these instruments can be used. For example, a lab experiment with known NH_3 and CH_4 concentrations could give more information about the range in which the cross-compensation between those two gases is linear and correctly implemented by the Innova. Additionally, it is always highly recommended to perform additional measurements with an **alternative measuring method** at regular intervals to check the proper functioning of the used measuring devices. Furthermore, extra research should be conducted on the development of easy to maintain **low-cost continuous measuring systems** (e.g. electrochemical sensors or adapter laser systems). **Reduced sampling strategies**, together with low-cost continuous measuring systems, could significantly reduce the costs to measure the effectiveness of new low emission housing systems and techniques and consequently stimulate innovations in housing systems and techniques. Therefore, simulations on new datasets should be performed in order to validate the results obtained in chapter 3. Obtaining a **reference method and sampling instrument** for indoor measurements **at high PM concentrations** remains a challenge which needs to be tackled in the future. Apart from the mass or number concentrations of PM, future research will have to focus on the **PSD of PM in agriculture** and the nature (e.g. density, composition or source) of PM in agriculture. The latter could be accomplished by making use of techniques such as **electron microscopy** or other imaging techniques. The **composition of secondary PM** inside and in the vicinity of livestock buildings is another important aspect which has to be investigated in the future.

Further research is also required in order to find **the optimum** between the **feed efficiency** of different diets and the **PM** (partly) generated by these different diets. In order to reduce the need for costly experimental trials inside pig buildings, the laboratory tests (drop and shake tests), as discussed in chapter 2.3, could be further modified and fine-tuned until a good correlation between the PM values obtained inside the barn and the PM values from

the laboratory tests is achieved. This would make it possible to test several different forms and compositions of diets in a standardised and cost efficient way.

It remains useful to explore the **effectiveness of source-oriented techniques**. These techniques can reduce both the indoor pollutant concentrations as pollutant emissions, often at a lower cost than end-of-pipe techniques. Furthermore, it might be possible to combine these techniques to reach higher reduction percentages. However, care must be taken that, when combining different techniques, these techniques do not counteract each other. Therefore, the working principles of these techniques should be well documented and jointly evaluated. This evaluation should further take into account several pollutants and other important parameters, e.g. installation and operational costs. The reduction percentage, obtained with **combined source-oriented techniques** will also have to be established.

The experience gained in this dissertation has also shown that it is not straightforward to **combine different research goals into one measuring campaign**. Therefore, it is strongly recommended to list the different needs of each research goal separately and to identify a common research approach which still corresponds to **the individual needs of each project**. However, financial and time-bound parameters will ensure that this task remains a difficult exercise in the future.

Although this thesis already combined different measuring and analysis techniques and had a multidisciplinary approach, still a large number of relations between pollutants and other important aspects could not be investigated. Further research could, for example, investigate the link between PM emissions and the **transmission of pathogens** or the link between PM (and gas) emissions and **odour issues**.

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APPENDIX A: LIST OF LAE HOUSING SYSTEMS FOR FATTENING PIGS IN FLANDERS

The following LAE housing systems for fattening pigs were approved in the Ministerial Decree of May 31th, 2011 and have a maximal emission factor of 1.4 kg NH_3 $year^{-1}$ animal $place^{-1}$:

System V4.1: Manure reception in aerated liquid manure and flushing with aerated liquid manure - pen area of 0.65 m² to 0.8 m²

System V4.2: Manure reception in aerated manure and replacement of the aerated manure via a sewage system or another drainage which can be secluded from the air - pen area of 0.65 m² to 0.8 m²

System V4.3: Manure surface cooling system with 170 % cooling area and floors with increased manure passage

System V4.4: Manure surface cooling system with 200 % cooling area and floors with increased manure passage, up to 0.8 m² emitting manure surface area per animal place

System V4.5: Manure surface cooling system with 200 % cooling area and not with floors with increased manure passage, up to 0.6 m² emitting manure surface area per animal place

System V4.6: Manure pit with (water) and manure channel with an overflow, eventually with sloped pit wall(s) and with floors with increased manure passage, up to 0.27 m² emitting manure surface area per animal place

System V4.7: Manure pit with (water) and manure channel with an overflow, the latter with sloped pit wall(s) and not with floors with increased manure passage, up to 0.18 m² emitting manure surface area per animal place

An eighth system was added to this list in the Ministerial Decree of March 26th, 2012:

System V4.8: separate discharge of manure and urine by means of a manure and liquid manure gutter with manure scraper

CURRICULUM VITAE

EDUCATION

2011 – present	Doctoral Schools of Bioscience Engineering, Ghent University, Ghent
2007 – 2009	MSc in Biochemistry and Biotechnology, option Research, Faculty of Science, KU Leuven, Leuven
2004 – 2007	BSc in Biochemistry and Biotechnology, option Informatics, Faculty of Science, KU Leuven, Leuven
2002 – 2004	Secondary school, Science – Mathematics, KOL Sint-Gertrudis, Landen
1998 – 2000	Secondary school, Latin – Mathematics, KOL Sint-Gertrudis, Landen

PROFESSIONAL CAREER

2011 – present	Researcher at the Institute for Agricultural and Fisheries Research (ILVO), Technology and Food Science Unit, Agricultural Engineering Animal Sciences Unit, Pig Husbandry Research subject: 'Source-oriented mitigation techniques for air emissions from pig husbandry: a multi-pollutant approach.'
2009 – 2010	Researcher at the KU Leuven, Department of Chemistry, , Division of Biochemistry ,Molecular and Structural Biology, Laboratory for Biomolecular Modelling Research subject: 'Structural research and drug design on the interaction between the ant-apoptotic protein Bcl-2 and the IP3-receptor.'

PUBLICATIONS

INTERNATIONAL JOURNALS WITH PEER REVIEW

Ulens, T., Millet, S., Van Weyenberg, S., Van der Meeren, P., Van Langenhove, H., & Demeyer, P. (2015). Correlations between aerial pollutants and particle size distributions of particulate matter inside a pig fattening facility (Submitted).

Michiels, A., Piepers, S., Ulens, T., Van Ransbeeck, N., Del Pozo Sacristán, R., Sierens, A., Haesebrouck, F., Demeyer, P., & Maes, D.. (2015). Impact of particulate matter and ammonia on average daily weight gain, mortality and lung lesions in pigs (Submitted).

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Voet, A., Callewaert, L., Ulens, T., Vanderkelen, L., Vanherreweghe, J. M., Michiels, C. W., & De Maeyer, M. (2011). Structure based discovery of small molecule suppressors targeting bacterial lysozyme inhibitors. *Biochemical and biophysical research communications, **405**,* 527-532.

CONFERENCE PROCEEDINGS (FIRST AUTHOR)

Ulens, T., Millet, S., Van Ransbeeck, N., Van Weyenberg, S., Van Langenhove, H., & Demeyer, P. (2014)**.** Indoor concentrations of particulate matter in a pig fattening facility: effect of different pen cleaning techniques and housing systems. In: Book of Abstracts of the International Conference on Atmospheric Dust, Castellaneta Marina, Italy.

Ulens, T., Van Ransbeeck, N., Van Langenhove, H., & Demeyer, P. (2014)**.** Preliminary research on the ammonium content of particulate matter (PM) from indoor air of pig housing systems. In: Book of Abstracts of the International Conference on Atmospheric Dust, Castellaneta Marina, Italy.

NATIONAL JOURNALS

De Smet, S, Ulens, T., & De Sutter, R. (2014). (Fijn) stof tot nadenken. *Management en Techniek, 21, 34-35.*

Brusselman, E., Van Ransbeeck, N., Ulens, T., Hove, N., & Demeyer, P. (2012). Luchtemissies uit Vlaamse stallen: Onderzoek ter ondersteuning van de sector en de regelgever. *MilieuTechnologie, 9, 2-7*.

SCIENTIFIC ACTIVITIES

ORAL PRESENTATIONS AT CONFERENCES

June 1-6, 2014 — Indoor concentrations of PM in a pig fattening facility: effect of different pen cleaning techniques and housing.
International Conference on Atmospheric Dust, Castellaneta Marina, Italy

November 6-8, 2012 — Preliminary research on the presence of ammonium salts in PM emissions from piggeries.
11th NH_3-workshop, Hannover, Germany

April 23th, 2010 — Discovery of an inhibitor for a lysozyme – lysozyme inhibitor interaction using a virtual screening approach.
PhD interaction day BioSCENTer, Leuven, Belgium

POSTER PRESENTATIONS AT CONFERENCES

June 1-6, 2013 — Indoor concentrations of PM in a pig fattening facility: effect of different pen cleaning techniques and housing.
International Conference on Atmospheric Dust, Castellaneta Marina, Italy

April 23th, 2010 — Discovery of an inhibitor for a lysozyme – lysozyme inhibitor interaction using a virtual screening approach.
PhD interaction day BioSCENTer, Leuven, Belgium

March 1-2, 2010 — Discovery of an inhibitor for a lysozyme – lysozyme inhibitor interaction using a virtual screening approach.
10th Flemish Youth Conference of Chemistry, Two day conference of the youth section of the Royal Flemish Society of Chemistry, Blankenberge, Belgium

November 6th, 2009 — Discovery of an inhibitor for a lysozyme – lysozyme inhibitor interaction using a virtual screening approach**.**
Does size matter? Beyond small molecule therapeutics: challenges and
success stories, Annual One-Day Meeting on Medicinal Chemistry of SRC & KVCV, ULB Brussels, Belgium

AWARDS

June 6, 2014 — Award for best oral presentation 'Indoor concentrations of PM in a pig fattening facility: effect of different pen cleaning techniques and housing. *International Conference on Atmospheric Dust, Castellaneta Marina, Italy*