

# Contribution of agriculture to Air Quality problems in cities and in rural areas in Europe



**ETC/ACM Technical Paper 2013/10**  
**August 2013**

*Susana López-Aparicio, Cristina Guerreiro, Mar Viana,  
Cristina Reche, Xavier Querol*



The European Topic Centre on Air Pollution and Climate Change Mitigation (ETC/ACM) is a consortium of European institutes under contract of the European Environment Agency  
RIVM UBA-V ÖKO AEAT EMISIA CHMI NILU VITO INERIS 4Sfera PBL CSIC

**Front page picture:**

*Livestock production and manure spreading; Photo Source: S. Lopez-Aparicio and Wikipedia Commons licensed under the Creative Commons Attribution Share Alike 2.0 Generic Licence.*

**Author affiliation:**

*Susana López-Aparicio, Cristina Guerreiro: NILU – Norwegian Institute for Air Research, Norway*

*Mar Viana, Cristina Reche, Xavier Querol: IDAEA-CSIC – Institute for Environmental Assessment and Water Research, Spain*

**DISCLAIMER**

This ETC/ACM Technical Paper has not been subjected to European Environment Agency (EEA) member country review. It does not represent the formal views of the EEA.

© ETC/ACM, 2013.

*ETC/ACM Technical Paper 2013/10*

*European Topic Centre on Air Pollution and Climate Change Mitigation*

*PO Box 1*

*3720 BA Bilthoven*

*The Netherlands*

*Phone +31 30 2748562*

*Fax +31 30 2744433*

*Email [etcacm@rivm.nl](mailto:etcacm@rivm.nl)*

*Website <http://acm.eionet.europa.eu/>*

## Table of Contents

<b>1. Introduction.....</b>	<b>1</b>
1.1. Background.....	1
1.2. Secondary Inorganic Aerosol (SIA) formation.....	2
1.3. Agricultural sources of pollutants.....	3
Livestock production.....	3
Application of Fertilizers and Pesticides.....	3
Land preparation.....	3
Harvesting.....	3
Field burning of agricultural waste.....	4
1.4. Objectives.....	4
<b>2. Methodology.....</b>	<b>4</b>
<b>3. Agricultural Emissions and Contribution to EU-27 Emissions.....</b>	<b>5</b>
<b>4. Contribution of Agricultural Emissions to Particulate Matter Concentrations.....</b>	<b>8</b>
<b>5. NH<sub>3</sub> Emission Control and Mitigation Measures.....</b>	<b>13</b>
Nitrogen management.....	14
Animal feeding strategies.....	16
Animal housing.....	16
Manure storage.....	17
Manure application.....	18
Fertilizer application.....	19
<b>6. Conclusions.....</b>	<b>20</b>
<b>7. References.....</b>	<b>21</b>

## 1. Introduction

This report is part of the air pollution assessment carried out to support the Air Quality Directives revision. The current work is a literature review focusing mainly on the analysis of agricultural emissions, especially relevant for the future negotiations of the National Emission Ceiling (NEC) directive and to address the recurrent particulate matter (PM) exceedances in Europe, and the impact of agricultural emissions on air quality and particularly on PM.

### 1.1. Background

Air pollutant emissions from agriculture are main contributors to air pollution, and imply both local and regional problems, such as PM exposure, eutrophication and acidification, and contribution to greenhouse gas (GHG) emissions. Air pollutants emitted from the agricultural sector are mainly methane ( $\text{CH}_4$ ), nitrous oxide ( $\text{N}_2\text{O}$ ) and ammonia ( $\text{NH}_3$ ). Agriculture is also a main source of PM, both primary and secondary in origin. Figure 1 shows the contribution from agricultural emissions to the EU-15 emissions of specific atmospheric pollutants.  $\text{NH}_3$  is a main concern as emissions contribute notably to acidification, eutrophication and PM formation. Thereby agricultural emissions are of great concern, as  $\text{NH}_3$  from agriculture contributed to 94% of total  $\text{NH}_3$  emissions in 2010 within the EU-27 (Chapter 3).

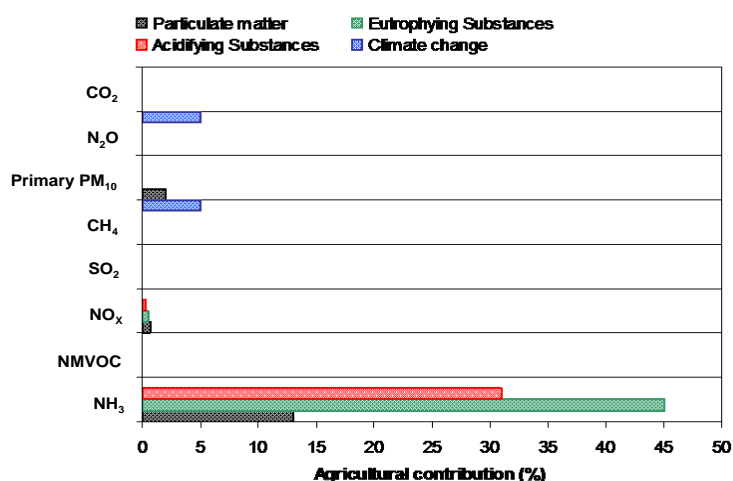


Figure 1: Emission-based assessment of agriculture contribution to different environmental issues in EU-15 (After Erisman et al., 2008).

Policies have been developed to reduce agricultural emissions to the air with certain success in some countries. In the case of particulate matter (PM), these policies focus mainly on primary PM (as opposed to secondary). The slowly decreasing agricultural  $\text{NH}_3$  emissions are expected to contribute to a baseline PM concentration that is only declining slowly. Some European countries (e.g. France, The Netherlands) experience a substantial number of exceedances of PM limit values where secondary inorganic aerosol (SIA) formed from agricultural emissions of  $\text{NH}_3$  play a major role (Weijers et al., 2010). In parts of Europe agricultural biomass burning is common practice, and besides the release of primary PM, emissions of carbonaceous species occur leading to secondary organic aerosol (SOA) formation.

This report focuses on the contribution of agricultural emissions to total EU emissions (Chapter 3) and air quality problems (Chapter 4), with special focus on  $\text{NH}_3$  emissions and PM concentration levels. The assessment relates to the analysis of the agricultural emission reduction strategies in view

of the future negotiations of the National Emission Ceilings (NEC) directive for certain pollutants. A literature review has been carried out to determine the contribution from a quantitative point of view and based on the latest studies. Additionally, based on the literature review the best available control measures for mitigation of NH<sub>3</sub> emissions from agriculture are summarized and discussed (Chapter 5).

## 1.2. Secondary Inorganic Aerosol (SIA) formation

In Europe, secondary particulate matter (both SIA and SOA) contributes about 70% or more of PM<sub>2.5</sub> levels in the air (Putaud et al 2010). Therefore, to reduce PM concentrations it is necessary to lower precursor emissions. The precursors of secondary PM are SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub>, anthropogenic and biogenic volatile organic compounds (VOCs); secondary PM components are sulphate, nitrate, ammonium and compounds of organic carbon. While European NH<sub>3</sub> emissions are known to stem mainly from agriculture, SO<sub>2</sub> and NO<sub>x</sub> emissions have an industrial and traffic-related origin. It is relevant to consider here the successful abatement of SO<sub>2</sub> emissions which has been taking place in recent years, resulting in NH<sub>4</sub>NO<sub>3</sub> as the main contributor to secondary inorganic aerosols.

Sulphate (SO<sub>4</sub><sup>2-</sup>), nitrate (NO<sub>3</sub><sup>-</sup>) and ammonium (NH<sub>4</sub><sup>+</sup>) are the main SIA components in PM, occurring mainly as ammonium sulphate ((NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>) and ammonium nitrate (NH<sub>4</sub>NO<sub>3</sub>). These salts are result from the neutralization of sulphuric acid (H<sub>2</sub>SO<sub>4</sub>) and nitric acid (HNO<sub>3</sub>) with ammonia (NH<sub>3</sub>), and they exist in thermodynamic equilibrium with the precursor gases.

The equilibrium between gaseous ammonia and nitric acid, and ammonium nitrate aerosol is written as:



Ammonium nitrate is semi-volatile and the equilibrium can shift to the gas phase under conditions of high temperature and/or low relative humidity. On the other hand, the formation of ammonium bisulphate and sulphate follows the reactions 2 and 3, respectively, which occurs at higher velocities with increasing temperature (mainly during the summer months):



Both reactions, to form ammonium sulphate from H<sub>2</sub>SO<sub>4</sub> and ammonium nitrate from HNO<sub>3</sub>, arising from SO<sub>2</sub> and NO<sub>x</sub> oxidation, compete with each other for 1) the available OH-radicals to form sulphuric acid and nitric acid, respectively, and 2) for the availability of NH<sub>3</sub>. Ammonia tends to be trapped to preferably form ammonium sulphate, thus the formation of ammonium nitrate will depend on the availability of NH<sub>3</sub>. It has been established that the amount of NH<sub>3</sub> needed to lead to the formation of ammonium nitrate exceeds the amount of sulphate by a factor of two (Schaap, 2003). While ammonium sulphate is a well-known tracer of long-range transport due to its stability and consequent long atmospheric residence time, ammonium nitrate is unstable at temperatures >25-30°C.

### 1.3. Agricultural sources of pollutants

A brief characterization of the most frequent agricultural sources of pollutants is presented in this section, with special focus on sources of both primary and secondary PM, and NH<sub>3</sub>.

#### **Livestock production**

Livestock production constitutes the main source of air pollution from agricultural activities. The most relevant emissions are CH<sub>4</sub>, which mostly occur as part of the natural digestive process of animals and the manure management, N<sub>2</sub>O, and NH<sub>3</sub>. Coarse particles (PM<sub>10-2.5</sub>) are also produced within a livestock farming environment, they are primary in origin and consist of up to 90% organic matter (Aarnink et al. 1999), which provides opportunities for the adherence of bacteria and odorous components to the particles. The extent to which PM from livestock houses can adsorb and contain irritating gases such as NH<sub>3</sub>, odorous compounds, and pathogenic and non-pathogenic micro-organisms is still uncertain. One of the main sources of NH<sub>3</sub> is the manure from livestock production, and mainly through the processes of storage and management. As described in Chapter 3, the NH<sub>3</sub> content in the manure and therefore volatilization will depend on the livestock type.

#### **Application of Fertilizers and Pesticides**

In recent years, the consumption of fertilizers has increased exponentially worldwide, causing serious environmental problems and responsible for an expected significant increase of N<sub>2</sub>O emissions by 2020 (Smith et al. 2007). Air pollution associated with the application of fertilizers is dominated by nitrogen oxides (i.e. NO, N<sub>2</sub>O, NO<sub>2</sub>) and NH<sub>3</sub> emissions. The application of ammonium fertilizers to the soil surface may result in ammonium loss to the atmosphere by volatilization, especially soils with a high pH (i.e. pH>7). In European countries, NH<sub>3</sub> volatilization from field-applied fresh manure, a commonly used organic fertilizer, is a major contributor to the overall NH<sub>3</sub> load from the agricultural sector (Huijsmans et al., 2003). These emissions also lead to secondary inorganic aerosol formation and therefore increase in the PM load.

#### **Land preparation**

The preparation of the land includes the activities carried out before cultivation and after harvesting. This activity may account for most of PM emissions from agricultural operations, being tillage one of the most important causes of release of PM from soils. Measurements in Europe indicate that emission of dust by tillage operations is many times higher than by natural wind erosion (Funk et al. 2008).

#### **Harvesting**

Harvesting includes three different types of operations; crop handling by the harvest machine, loading of the harvested crop into trailers or trucks, and transport by trailers or trucks in the field. Emissions from these operations are in the form of solid particulates composed mainly of raw plant material and soil dust that is entrained into the air. These emissions may simply be due to soil resuspension, the mechanical processing of the plant material and underlying soil, or due to blowing or sweeping the crop to remove waste materials (e.g. almonds, Faulkner et al., 2011). Defoliant and/or desiccants may constitute an additional source of PM. They are used in some crops weeks before harvesting and PM emissions are due to the drifting of the chemicals, which has been estimated to be about 1% of the product applied on the crop (USEPA, 2006).

### Field burning of agricultural waste

Agricultural residue burning is the main source of primary PM<sub>2.5</sub> emissions in the agricultural sector, as well as a major source of secondary organic aerosols (SOA). PM resulting from burning of crop residues is made up of a large variety of carbonaceous compounds including sugars, polycyclic aromatic hydrocarbons (PAHs), alkanes and alkenes (Lioussé et al., 1996; Turn et al., 1997). Additionally a large variety of volatile organic compounds (VOCs), including PAHs, have been identified as products from the burning of agricultural waste residues (Keshtkar et al. 2007). Inorganic compounds such as KCl are also generated by this activity (Niemi et al., 2004; Hays et al., 2005). These studies evidence the large variability of emissions as a function of the agricultural residue being combusted (e.g., rice or wheat residuals, pruning residues, etc.).

### 1.4. Objectives

The aim of this report is to collect and review the outcomes from the latest studies about the contribution of agriculture to air quality problems, with special interest in the contribution to PM concentration levels, and with focus on the contribution of NH<sub>3</sub> emissions to the formation of SIA. In addition, the best available NH<sub>3</sub> mitigation measures will be presented, based on the latest work done to support the review and amendment of the Gothenburg Protocol under the Convention on Long-range Transboundary Air Pollution (UNECE, 2012a).

The objective of this report is to shed light in the understanding of the contribution of agriculture to air quality in urban and rural areas in Europe through answering the following questions:

- How much does agriculture contribute to air quality problems and in particular to PM levels in the air?
- What are the best available control measures to abate agricultural air pollution, with special focus on NH<sub>3</sub> emission reductions.

## 2. Methodology

A literature search was carried out in different phases, and focussing mainly in European studies. A first phase focussed on peer-reviewed journals and issues published in the last 5 years (i.e. 2008-2012), thereafter the search was extended to older issues as few relevant results were found. The search was primary carried out via search engines such as “ISI Web of Knowledge”, “Science Direct” and “Springer Link” and based on combinations of the following keywords; “ammonia”, “ammonium”, “secondary inorganic aerosol (SIA)”, “particulate matter” “agriculture”, “agricultural contribution”, “mitigation”, “agricultural emissions”, among others. A second phase of the search focussed on the so called grey literature, which include reports, communications and databases. The grey literature has been mainly obtained from national/regional air quality assessment programmes (e.g. Netherland, Ile-de-France region), through EU programs such as “The European Nitrogen Assessment” (ENA) from the NinE programme (Nitrogen in Europe, current problems and future solutions), reports published by the European Environmental Agency, the Convention of Long-range Transboundary Air Pollution (LRTAP) and in particular documents related with the Gothenburg Protocol. The information concerning emission at European levels have been obtained mainly from official reports to the LRTAP, and later on it has been supported by information from the scientific literature.

### 3. Agricultural Emissions and Contribution to EU-27 Emissions

As pointed out at the beginning of this report, agriculture is largely responsible for emissions of major greenhouse gases (i.e.  $\text{CH}_4$  and  $\text{N}_2\text{O}$ ) and ammonia, and it is a significant source of PM, both primary and secondary. In 2002 agriculture contributed around 10% of total greenhouse gases (GHG) and 94% of ammonia emissions (EEA, 2005; Figure 2). Nitrous oxide ( $\text{N}_2\text{O}$ ) is emitted from synthetic fertilizers and from soil and livestock manure/urine management, while methane ( $\text{CH}_4$ ) is mainly emitted from enteric fermentation, manure management, burning of agricultural residues and rice cultivation. Ammonia emissions occur mostly as a result of volatilization from livestock excretion (e.g. manure, slurry), thus the most common sources of ammonia are livestock buildings, open feedlots, manure storage facilities, and activities related with the handling, management and treatment of the manure and when that is applied on land. Regarding primary PM, tillage and harvesting are the main contributors along with the burning of agricultural residues, which especially contributes to  $\text{PM}_{2.5}$  emissions.

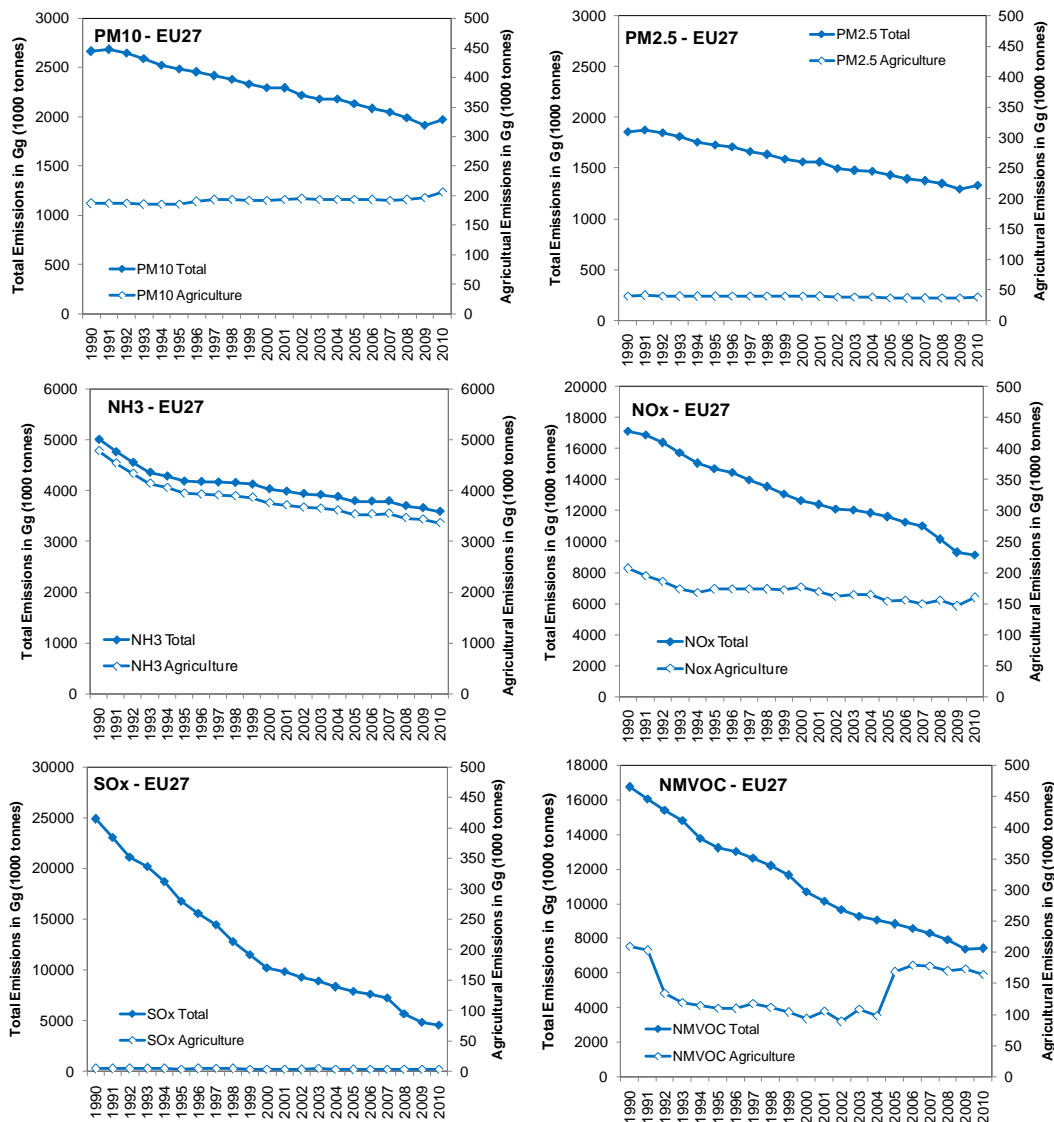


Figure 2: Emissions of primary particulate matter ( $\text{PM}_{2.5}$  and  $\text{PM}_{10}$ ), and secondary particulate matter precursors ( $\text{NH}_3$ ,  $\text{NO}_x$ ,  $\text{SO}_x$ , and NMVOC) for total (all sectors) and for agricultural sector in EU27 in 2010. National emissions reported to the Convention on Long-range Transboundary Air Pollution (Source: EEA 2013).



Figure 2 shows the contribution of agriculture to total EU-27 emissions of particulate matter (PM) and secondary PM precursors from 1990 to 2010. Agricultural emissions contribute about 2.5 and 8.5% of total emissions of primary PM<sub>2.5</sub> and PM<sub>10</sub>, respectively, whereas it is responsible of 94% of the total NH<sub>3</sub> emissions (EEA 2005). Total emissions changes are noteworthy; significant decrease is observed for the precursors, SO<sub>x</sub>, NMVOC and NO<sub>x</sub> from 1990 to 2010, with a decrease of about 44, 125 and 87%, respectively. Primary emissions of PM<sub>2.5</sub> and PM<sub>10</sub> decrease by around 40 and 35%, respectively, and NH<sub>3</sub> emissions by around 40% from 1990 to 2010. Emissions from the agricultural sector have also experienced reductions, for instance, emissions of PM<sub>2.5</sub> and PM<sub>10</sub> decreased by 5 and 10% from 1990 to 2010, respectively, and NO<sub>x</sub> and NH<sub>3</sub> emissions decreased by 30 and 42%, respectively.

Agricultural emission changes and specifically those related to ammonia emission are slightly different between European countries. Agriculture is estimated to contribute to around 97% of the anthropogenic ammonia emissions in France and Denmark (CITEPA, 2010; NERI, 2011), with about 40% due to land fertilization. In Greece livestock production is estimated to contribute by around 57% to national total NH<sub>3</sub> emissions (Sidiropoulos and Tsilingiridis, 2008).

Figure 3 shows reported changes in ammonia emissions for European country and the comparison with the emission ceiling directive and 2020 Gothenburg protocol target. Countries such as Netherlands and Lithuania reported ammonia emissions reductions above 60% from 1990 to 2010, whereas Norway and Spain reported ammonia emission increases (Figure 3). All EU Member States except Spain and Denmark reported national NH<sub>3</sub> emissions levels for 2010 below the emission ceilings set by the National Emission Ceilings Directive (NECD).

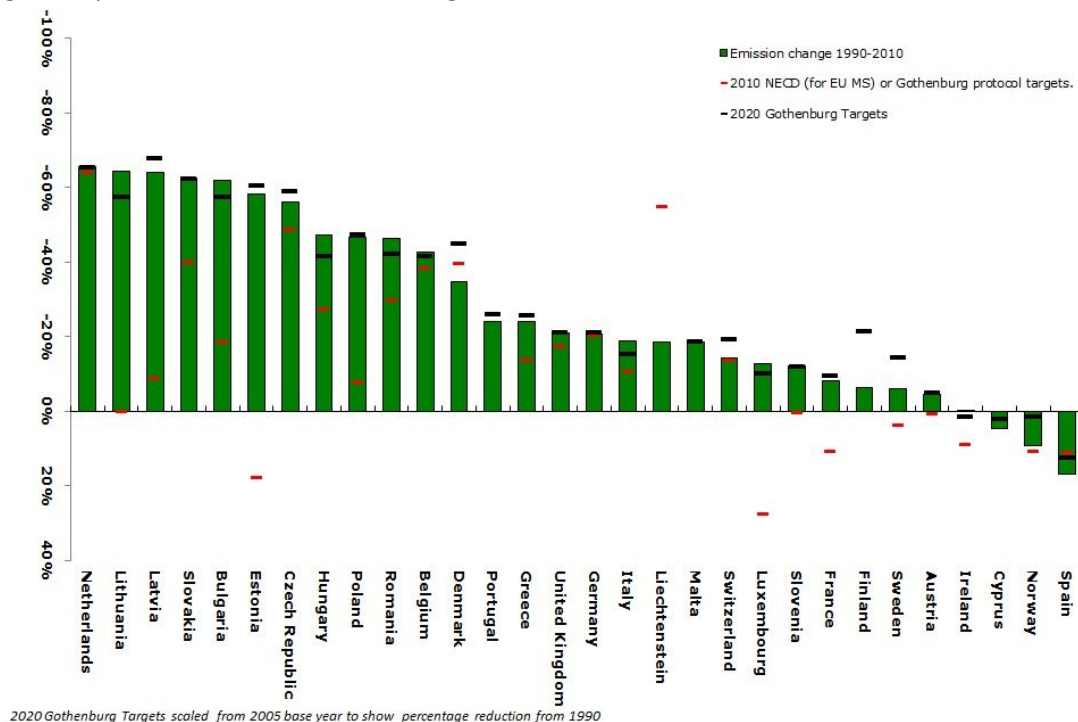


Figure 3: Reported changes in ammonia (NH<sub>3</sub>) emissions for each country from 1990 to 2010 in comparison with the 2010 National Emission Ceilings Directive (NECD) and 2020 Gothenburg protocol targets (EEA/ACM, 2013).

European figures for PM emissions may also slightly differ from those at national level and especially for those countries with intense agriculture. For instance in The Netherlands, the contribution of agriculture to PM emissions has been estimated to be of about 25% in 2002 (Chardon and van der

Hoek, 2002) and of about 17% contribution to emissions of PM<sub>10</sub> in 2010 (Dutch Emission Registration, 2013). In Denmark, 29% of the national Total Suspended Particulate (TSP), 18% of PM<sub>10</sub> and 5% of PM<sub>2.5</sub> emissions is attributed to agriculture (NERI, 2011), in France, the percentage was estimated to be up to 30% of the national emission of PM<sub>10</sub> (CITEPA, 2010) and in the UK, the estimated national agricultural contribution to total PM<sub>10</sub> emissions was between 5-15% (HPA, 2006).

Differences in emissions have also been reported for different agricultural activities or types of productions; for instance in livestock production, poultry and pig houses are the main sources of PM emissions, contributing to about 50 and 30%, respectively, of total PM emissions from agriculture in Europe (EMEP-CORINAIR, 2007). In the United Kingdom the spatial distribution of NH<sub>3</sub> emissions showed high levels at areas characterized by intensive agricultural activity, specially pig and poultry farming (Hellsten et al., 2008). Figure 4 shows the emissions from different types of livestock production at European levels in 2010. Swine, poultry and cattle are the main emission sources for PM and NH<sub>3</sub>. Emissions from poultry and swine are mainly of PM<sub>10</sub>, and cattle and swine are the main sources of NH<sub>3</sub>.

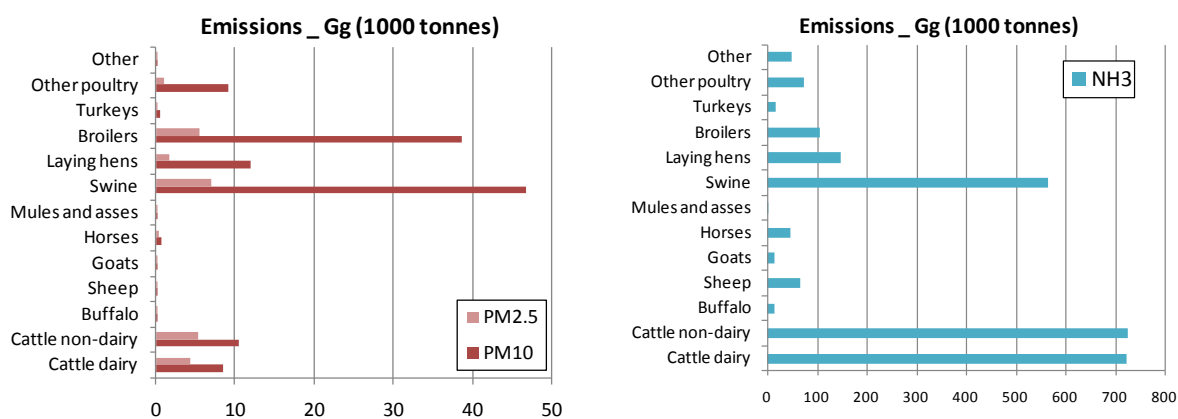


Figure 4: Emissions from livestock production sectors in Europe (2010). National emissions reported to the Convention on Long-range Transboundary Air Pollution (Source: EEA, 2013).

Other activities need to be taken into account as part of the agricultural sector. Figure 5 shows NH<sub>3</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> emissions from different activities of the agricultural sectors; livestock production (i.e. Cattle dairy and non-dairy, buffalo, sheep, goats, horses, mules and asses, swine and poultry), synthetic N-fertilizers, agricultural operations both at farm level and off farm (e.g. plowing, harrowing, disking and cultivating), N-excretion, burning of agricultural wastes, and others. Comparing the different activities, livestock production is the main emission source of NH<sub>3</sub>, PM<sub>10</sub> and PM<sub>2.5</sub>. Synthetic N-fertilizers contribute in addition to NH<sub>3</sub> emissions, and agricultural operations at farm levels contribute to PM, especially PM<sub>10</sub>. For instance, Bogman et al. (2007) assessed the particle emission from farming operations in Belgium and they established emissions of about 10.1 kton (7.5 kg ha<sup>-1</sup>) of TSP per year, and from 2.0 to 3.1 kton (1.5 to 2.3 kg ha<sup>-1</sup>) of PM<sub>10</sub>, accounting for 35% of total TSP emission and 24% of total PM<sub>10</sub> emission. It is relevant to highlight here that estimates of emissions from burning of agricultural waste may not be fully accurate, given the complexity of this issue. On the one hand, obtaining precise emission factors is highly complex because of the largely varied types of agricultural residues which are burnt in the different regions in Europe. On the other hand, biomass burning activities are mostly regulated to be carried out during specific annual periods with favourable meteorological conditions, but these regulations are not

always observed and the degree of un-regulated biomass burning activities may vary across the different European regions.

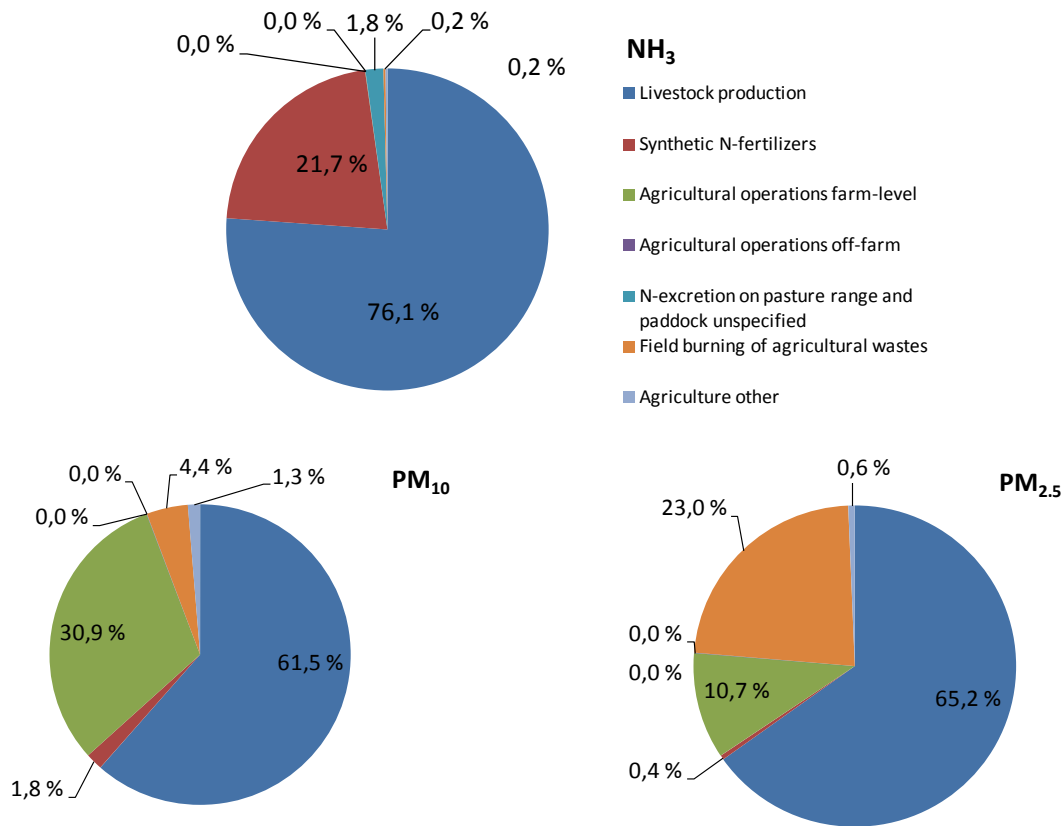


Figure 5: NH<sub>3</sub>, PM<sub>10</sub> and PM<sub>2.5</sub> emissions from agricultural sectors in Europe (2010). National emissions reported to the Convention on Long-range Transboundary Air Pollution (Source: EEA, 2013).

#### 4. Contribution of Agricultural Emissions to Particulate Matter Concentrations

Agricultural emissions can contribute to PM levels directly (primary particles) or indirectly through the formation of SIA and SOA (secondary particles). Primary emissions of PM (PM<sub>2.5</sub> and PM<sub>10</sub>) from agriculture are below 10% of total emissions at European levels, and studies about the contribution of agricultural emissions to PM concentration through primary emissions are scarce. Agricultural activities sources of primary PM range from those associated with livestock production, harvesting, land preparation to agricultural waste burning. In addition, factors such as housing, feeding, type of animal and environmental will influence the formation of PM and the concentration levels. Cambra-López et al. (2010) reviewed the state of the art concerning particulate matter from livestock production emphasizing the need for further characterization of both PM and the sources. Although it is known that 90% of the PM from livestock production is organic matter, the contribution from primary or secondary sources is still uncertain.

The burning of agricultural residues is a significant source of primary PM, especially PM<sub>2.5</sub> as it contributes to about 23% of primary PM<sub>2.5</sub> emissions from the agricultural sector (Figure 5). It is also a major source of secondary organic aerosols (de Gouw & Jiménez, 2009). Several studies in literature have focused on the characterization of the source profiles of PM emitted from a specific

kind of crop burning. In Zhang et al. (2007), authors investigated on the profiles of particulate organic matters emitted from cereal straw burnings, finding a high contribution of levoglucosan and methyloxylated phenols to the mass of fine particulates. In Hays et al. (2005) they investigated on the chemical characterization of PM<sub>2.5</sub> from simulated agricultural fires of surface residuals of rice and wheat in China, finding that combustion-derived PM emissions from wheat were enriched in K and Cl, whereas the PM emissions from rice were largely carbonaceous. Organic matter was identified to account for as much as 18% of the PM mass emissions. In Viana et al. (2008) it was shown that PM<sub>10</sub> levels during open burning events of rice straw residues were especially enriched in oxalate, fluoranthene, C31 n-alkane, levoglucosan, K, water-soluble organic carbon (WSOC), oleic acid, Cl<sup>-</sup>, Na, NO<sup>3-</sup> and V. Furthermore, high enrichments were observed for Zn, Pb, Cr, and Cd, probably due to bioaccumulation of these metals in rice straw. In Lai et al. (2009), rice-straw burning was identified as a predominant source of atmospheric PAHs during burning periods at a rural and at an urban sites in central Taiwan, especially of low-weights PAHs.

Despite the availability in the literature of studies focusing on the characterisation of emissions, there are very few studies that provide quantitative information about agricultural waste burning contribution to PM levels at both rural and urban areas. Reche et al. (2012a) and Viana et al. (2013) identified the transport of agricultural wastes burning emissions from the regional scale to the urban environment (i.e. Barcelona, Spain) and established a contribution of about 7±4% of PM<sub>2.5</sub> urban aerosols. Rural sites in Germany are one of the best characterized regarding this concern within Europe, as Germany is among the largest agricultural producers in the European Union, for example in Goossens et al. (2001) agricultural activities have been defined as a serious degradation process in rural environments in north-western Germany. In this study, authors estimated the contribution of agriculture to total transported dust levels, accounting for around 25-30%. PM source apportionment studies in rural areas generally identify biomass burning as one of the most important atmospheric emission sources, especially for the organic fraction (Puxbaum et al., 2006; Gelencsér et al., 2007; Sandradewi et al., 2008; Yttri et al., 2011). According to Puxbaum et al. (2006), biomass burning within Europe contributes in around 10-30% to organic matter in the PM<sub>2.5</sub> fraction. However, studies rarely discriminate between the contribution of wild and agricultural fires. Within Europe, open burning of rice field residues (rice straw) in a rural site in Spain was estimated to increase ambient PM<sub>10</sub> levels on the regional scale by 10–15 µg m<sup>-3</sup> on average, reaching 30 µg m<sup>-3</sup> during peak episodes, these concentrations may represent up to 40% of the PM<sub>10</sub> mass during peak episodes (Viana et al., 2008). Regarding biomass burning of agricultural residues, in an urban background site in northern Spain, a transport of agricultural wastes burning emissions from the regional scale towards the city was identified and quantified to contribute with 7±4% of PM<sub>2.5</sub> urban aerosols (Reche et al., 2012a).

Scarce information is available regarding the contribution of agricultural emissions to the urban environments. An estimation of this contribution is highly uncertain due to the variability of agricultural emissions together with the complexity of urban pollutants. Agricultural activities were estimated to increase annual PM<sub>2.5</sub> levels by 1% in Paris, although in this study it is highlighted that the effect of ammonia through formation of secondary particle was not investigated (AirParif, 2012).

The formation of SIA due partly to agricultural emissions of ammonia is of concern, as PM concentration in Europe remains high over large regions. It is essential to remember that NH<sub>3</sub> emissions originate mainly from agricultural sources, but that this is not the case for NO<sub>x</sub> and SO<sub>2</sub>. As

it was shown in previous chapters, total emissions of ammonia have an extraordinary contribution from the agricultural sector (i.e.  $\approx 94\%$ ), thus control strategies to reduce ammonia emissions will have a considerable influence on PM concentrations through reduction of formation of secondary PM (i.e. ammonium sulphate and ammonium nitrate). For instance, Erismann and Schaap (2004) addressed the role of ammonia in the formation of SIA and they pointed out how SIA concentration can only be effectively reduced if ammonia emissions are decreased. The EURODELTA II study (Thunis et al., 2008) shows that reductions of  $\text{NH}_3$  emissions may be more effective in terms of reduced PM mass concentrations per kt of gas emitted than reductions of  $\text{NO}_x$  emissions in some countries, like the UK. This is nevertheless not always the case, and the effectiveness of  $\text{NH}_3$  versus  $\text{NO}_x$  and  $\text{SO}_x$  emissions reduction, vary depending on the emission country. The same study shows that reducing primary PM emissions is considerably more effective to reduce PM concentrations, than reducing emissions of any precursor gas (about 3 to 5 times more effective per reduced emitted kt).

Urban ammonia emissions are also of concern in this respect and should thus also be addressed by control strategies (Reche et al., 2012b). Even if urban  $\text{NH}_3$  emissions are low in comparison to agricultural emissions, they are especially relevant because they are produced in areas (the urban environment) with relatively high  $\text{NO}_x$  emissions (potentially also  $\text{SO}_2$ ) and high population exposure. As a result, urban ammonia emissions have a high potential for SIA formation in the urban scale, as ammonia is a pollutant that deposits more rapidly than e.g.  $\text{NO}_x$  or  $\text{SO}_2$ . Significant urban  $\text{NH}_3$  sources have been identified in several cities (Perrino et al., 2002; Vogt et al., 2005).

SIA originated from agricultural but also from urban and industrial sources constitutes a significant part of PM, and especially of the fine fraction ( $\text{PM}_{2.5}$ ). In their review of 2010, Putaud et al. concluded that the sum of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  ranges between 19-20% of the  $\text{PM}_{10}$  mass from rural to kerbside environments in N-western Europe, is around 20% of  $\text{PM}_{10}$  in Southern Europe, and ranges between 17-22% of  $\text{PM}_{10}$  in Central Europe (Table 1). Values around 40% of SIA in  $\text{PM}_{10}$  have been reported in urban backgrounds in Germany (Quass et al., 2004) and in six European cities (i.e. Duisburg, Prague, Amsterdam, Helsinki, Barcelona and Athens; Sillanpaa et al., 2006), and SIA of about 20-30% are reported at European level (WHO, 2003) and of about 30 and 50% average in the Netherlands and associated with  $\text{PM}_{10}$  concentration levels below and above  $40 \mu\text{g m}^{-3}$ , respectively (Table 1). It is noteworthy to point out that in some European areas such as in The Netherlands the highest SIA levels frequently coincide with exceedances of the  $\text{PM}_{10}$  daily limit value (Weijers et al., 2010).

SIA are mainly composed of nitrate, sulphate and ammonium, thus the contribution of ammonium to SIA and total PM ( $\text{PM}_{2.5}$  and  $\text{PM}_{10}$ ) is relevant for the evaluation of the contribution of ammonia emissions, and thus agricultural emissions, to air quality and particularly to PM concentration levels. Table 1 shows a summary of studies which provide quantitative information about SIA and their constituents in  $\text{PM}_{2.5}$  and/or  $\text{PM}_{10}$ , at different scales and based on both measurements and modelling. The values reported for the contribution of ammonium to the PM are similar between studies and they range between 7 and 10% in most of the cases. In certain European regions such as Spain, clear spatial gradients for  $\text{NH}_4^+$  have been detected (Querol et al., 2008, Figure 6), with higher ammonium levels along the Eastern coast of Spain where the highest  $\text{NH}_3$  agricultural emissions are recorded but also where the relative humidity is highest, thus implying a higher atmospheric stability of the ammonium nitrate salt. Putaud et al. (2010) evaluated the data of aerosol concentration and composition obtained at >60 natural background, rural, near-city, urban, and kerbside sites across Europe. They concluded that the main constituents of both  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  are generally organic

matter, sulphate and nitrate, and that there is a clear decreasing gradient in  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  contribution to  $\text{PM}_{10}$  when moving from rural to urban to kerbside sites. In contrast, the total carbon/ $\text{PM}_{10}$  ratio increases from rural to kerbside sites. Significant gradients in PM chemistry were observed when moving from North-western, to Southern to Central Europe. In a review of European cities by Querol et al (2004), it was concluded that the contribution of SIA (from traffic, industrial emissions including power generation and agriculture) ranged from 3 to 9  $\mu\text{g m}^{-3}$  for  $\text{PM}_{10}$  and from 3 to 8  $\mu\text{g m}^{-3}$  for  $\text{PM}_{2.5}$  at regional sites. These concentrations were somewhat lower (3–5  $\mu\text{g m}^{-3}$ ) in Sweden. SIA levels were very similar in urban areas, but in intensively industrialised regions or heavily polluted urban areas, an input of secondary inorganic aerosols from 2 to 5.5  $\mu\text{g m}^{-3}$  was detected for  $\text{PM}_{10}$  and from 1 to 5  $\mu\text{g m}^{-3}$  for  $\text{PM}_{2.5}$  on top of the above contributions. Figure 7 shows the relative contribution of SIA components to the PM mass in three categories. Both sulphate and ammonium contribute mainly to the fine PM fraction ( $\text{PM}_{2.5}$ ), whereas nitrate contributes in addition to the coarse PM fraction ( $\text{PM}_{10-2.5}$ ) (Putaud et al. 2004).

Table 1: Contribution of SIA, sulphate, nitrate and ammonium reported in the literature. Studies listed in chronological order.

Study	Location - Scale	Contribution	Notes
Schaap, 2003	Europe	Sulphate: 25-50% $\text{PM}_{2.5}$ Nitrate: 5-35% $\text{PM}_{2.5}$	Modelling
Putaud et al. 2004	24 European sites; natural, rural, near city, urban and kerbside	<b>Ammonium</b> to $\text{PM}_{10}$ : 6% natural/rural background, 7% near-city/urban background, 4% kerbside <b>Ammonium</b> to $\text{PM}_{2.5}$ : 8% natural/rural background, 10% near-city/urban background, 7% kerbside	Measurements
Putaud et al. 2010	60 European sites; natural, rural, near city, urban and kerbside	Sulphate + Nitrate to $\text{PM}_{10}$ : 19-20% N-W Europe, 20% S-Europe and 17-22% C-Europe	Measurements
Querol et al. 2004	7 European site; regional background, roadside and urban background	SIA to $\text{PM}_{10}$ : 27-42% regional background, 10-33% roadside, 17-40% Urban Background SIA to $\text{PM}_{2.5}$ : 36-44% regional background, 23-45% roadside, 32-45% Urban Background	Measurements
Quass et al. 2004	Germany, urban background	SIA: 40% $\text{PM}_{10}$ (17% Nitrate; 16% Sulphate; 7% <b>Ammonium</b> ) SIA: 44% $\text{PM}_{2.5}$ (17% Nitrate; 18% Sulphate; 9% <b>Ammonium</b> )	Measurements
Sillanpaa et al. 2006	Duisburg, Prague, Amsterdam, Helsinki, Barcelona and Athens; Urban Background	SIA: 40% $\text{PM}_{2.5}$  Sulphate: 14-31% $\text{PM}_{2.5}$ <b>Ammonium</b> : 7-10% $\text{PM}_{2.5}$	Measurements
WHO, 2006	Europe	SIA: 20-30% $\text{PM}_{10}$ SIA: 30-40% $\text{PM}_{2.5}$	Modelling
Forfs et al. 2007	Belgium	Sulphate: 18% $\text{PM}_{2.5}$ Nitrate: 10% $\text{PM}_{2.5}$ <b>Ammonium</b> : 8% $\text{PM}_{2.5}$	Measurements
Whyatt et al. 2007	UK, regional	<b>Ammonium</b> : 14.4% $\text{PM}_{10}$ (measured) <b>Ammonium</b> : 8.4% $\text{PM}_{10}$ (modelled)	Measurements and Modelling
EMEP, 2008	Europe, rural stations	Nitrate: 6-19% $\text{PM}_{10}$ <b>Ammonium</b> : 5-9% $\text{PM}_{10}$	
Weijers et al., 2010	The Netherlands	SIA: 30-40% $\text{PM}_{10}$ (average) SIA: 25-35% $\text{PM}_{10}$ ( $\text{PM}_{10} < 40 \mu\text{g m}^{-3}$ ) SIA: 45-55% $\text{PM}_{10}$ ( $\text{PM}_{10} > 40 \mu\text{g m}^{-3}$ )	measurements and modelling
Hristov 2011	US, different regions	<b>PM<sub>2.5</sub> from ammonia</b> : 5-11% total $\text{PM}_{2.5}$	
Moldanová et al. 2011	Europe	<b>Ammonia</b> : 20% SPM	Modelling
AirParif, 2012	Paris; roadside, urban background, suburban background, rural	SIA: 25% $\text{PM}_{2.5}$ (25% traffic, 40% urban, >45% rural) SIA: 30% $\text{PM}_{10}$	Measurements

The comparison between stations carried out by Putaud et al. (2004) does not seem to show a clear trend in ammonium contribution (Figure 7), being slightly lower in kerbside, and similar at rural and urban backgrounds. However, a higher proportion of  $\text{NH}_4\text{NO}_3$  may be formed in urban atmospheres even at the local-scale (Amato et al., 2011) in Southern European regions. The levels of SIA at pairs of regional background and urban background stations in Southern Europe (applying the Lenschow approach) show important differences (Querol et al., 2004). Concerning the contribution of SIA to  $\text{PM}_{2.5}$ , it was 37% at urban environments in N-western Europe, 22% in Southern Europe and 32% in

Central Europe. It is estimated that the contribution from ammonium sulphate is mainly linked to long-range transport, whereas that of ammonium nitrate has also meso-scale and urban-scale components.

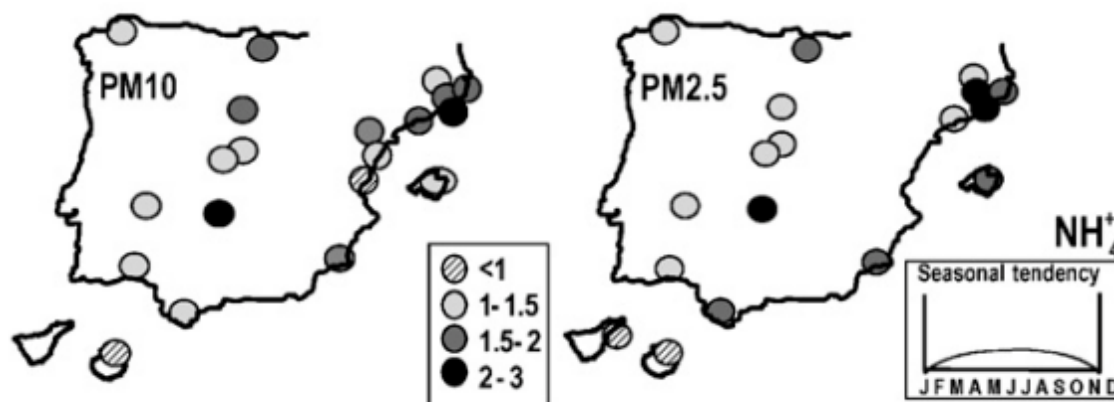


Figure 6: Distribution of mean annual  $\text{NH}_4^+$  levels ( $\mu\text{g m}^{-3}$ ) across Spain for both PM<sub>10</sub> (left) and PM<sub>2.5</sub> (right) fractions. Seasonal trends are also shown (Querol et al. 2008).

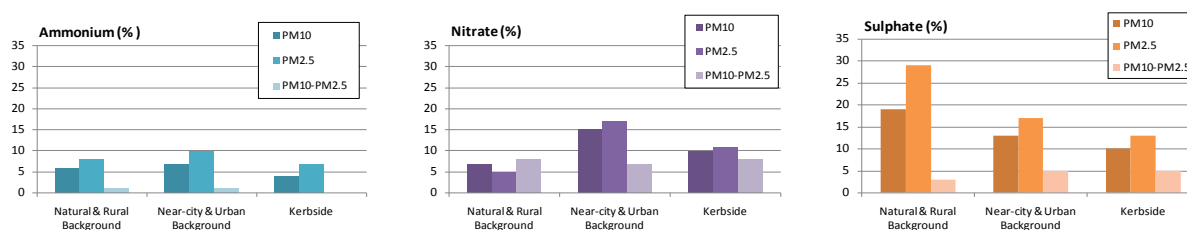


Figure 7: Relative contribution (%) of ammonium, nitrate and sulphate to PM mass in three site categories (Data from Putaud et al., 2004).

Ammonium contribution to PM is similar between studies, ranging between 4 to 10% in most of them (Table 1). For instance, Putaud et al. (2004) reported an ammonium contribution of about 4 - 7% to PM<sub>10</sub> and of about 7 - 10% to PM<sub>2.5</sub> (Figure 7), confirming the main contribution to the fine PM fraction. Similar ranges were obtained in Spain (Querol et al., 2008) and Germany (Quass et al., 2004) (Table 1), being also sulphate the main component of the SIA. Higher ammonium values than 10% have been reported at regional level in the UK ( $\approx 14\%$ ; Whyatt et al., 2007), and of about 20% of the suspended particulate matter at European level (Moldanová et al. 2011).

As discussed above, ammonium salt aerosols formed by the neutralization of sulphuric acid and nitric acid with ammonia originate partly from agricultural emissions. Agriculture extraordinarily contributes to total  $\text{NH}_3$  emissions (i.e. 94-97%) and it has been reported as the main source of ammonia. In addition and based on previous studies, ammonium is transported from the rural to the urban areas (Putaud et al. 2004), and even ammonium originating in the Netherlands has been established to contribute considerably to the particle mass in a German urban area (i.e. Münster; Vogt et al., 2005). Some studies established other urban sources as additional contributor to  $\text{NH}_3$  emissions in urban areas; such as traffic (e.g. Roma; Perrino et al., 2002), or waste containers and sewage systems (e.g. Barcelona; Reche et al., 2012b; Pandolfi et al., 2012).

Certain uncertainties in the assessment of agricultural emissions and thus formation of SIA exist. Recent investigations show that PM emissions from agriculture in regions of intensive ammonia emission may have been previously underestimated, and a large part of the gap between modelled and measured PM concentrations might be explained by previously underestimated agricultural sources (Wu et al., 2008). Thereby, uncertainties have been associated with measurement data, as volatilization loss has been identified in some studies (Keck and Wittmaack, 2005). Furthermore, large uncertainties still remain regarding primary emissions of mineral aerosols from land preparation activities, due to the large variability in emission factors across Europe as a consequence of the differences in aridity of the soils and in land preparation practices. Finally, SOA formation processes are well-known to be linked to agricultural biomass burning emissions, but studies suggest that previous estimates underestimated the relevance of this emission source (de Gouw & Jiménez, 2009). All of this results in the relatively high uncertainty of the estimates of the impact of agricultural emissions on air quality in Europe.

## 5. NH<sub>3</sub> Emission Control and Mitigation Measures

As indicated in previous chapters, the agricultural sector is responsible for more than 90% of the NH<sub>3</sub> emissions in Europe. Moreover, some studies pointed out about reducing NH<sub>3</sub> emissions as an effective way of reducing SIA concentration levels, thus PM concentration levels, and the number of PM<sub>10</sub> daily limit exceedances (Erisman and Schaap, 2004; Putaud et al., 2010). The most important sources of NH<sub>3</sub> emissions in the EU-27 are livestock production, especially animal manures, followed by the application of synthetic nitrogen fertilizers.

Animals utilize only a fraction (5 to 45%) of the available N in the feeding for the production of meat, milk, eggs and offspring. The greater part is excreted via urine and faeces, which is either stored and managed for some time in manure storage systems, or deposited directly on land and allowed to decompose. Following storage, manure is applied to agricultural land to fertilize crops including grasslands. However, only about 30 to 60% of the manure N will be utilized by growing crops for the production of plant protein, and only the protein in the harvested fraction of the crop will feed people or livestock. Therefore, in a livestock farming system less than 10% of the N from manure is exported from the farm as animal products (i.e. meat, milk and eggs), and the greater part is dissipated into the environment (Oenema and Velhof, 2007).

Significant losses of gaseous N compounds occur via volatilization of NH<sub>3</sub>. Emissions of NH<sub>3</sub> occur during various stages of the animal production and manure management:

- 1) Animal feed,
- 2) Animal manure excreted in housing systems and in pasture,
- 3) Animal manure in storage systems, and
- 4) From animal manure applied to crop land.

In addition, NH<sub>3</sub> emissions occur from the application of synthetic nitrogen fertilizers, especially urea- and ammonium-based nitrogen fertilizers. Figure # schematically shows the key sources of ammonia from livestock and crop production.



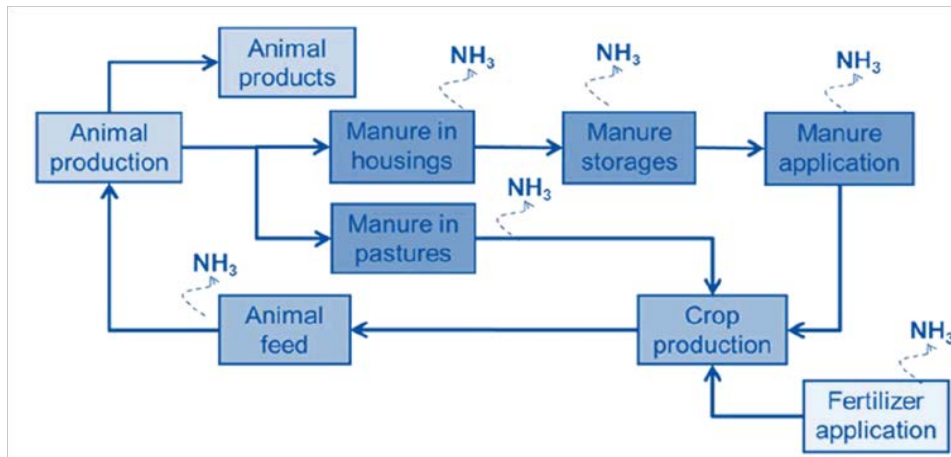


Figure 8: Key sources of ammonia from livestock and crop production (Source: Oenema and Velthof, 2012).

Methods to control and mitigate ammonia emissions from agriculture have been developed and tested for several decades. A range of emission control options is now proven in practice in more and more countries for the major sources of agricultural ammonia emissions (e.g. animal manure and urea fertilizer application). Furthermore and because of learning effects, the practical functioning of these techniques has been improved and costs have declined. This chapter summarizes a review of the main control and mitigation measures for  $\text{NH}_3$  emissions from agriculture reported in Oenema et al (2007), Oenema and Velthof (2007), Witzke and Oenema (2007), Döhler et al (2011), Oenema et al (2011), and most recently in Oenema and Velthof (2012) and UNECE (2012b).

It is important to note that emission reduction efficiencies of the different available abatement measures depend on the chosen reference system and on local factors, such as climate and soil conditions, differences in management practices and in the technical performances of abatement measures. Further, these categories of measures may affect each other's effectiveness and efficiency. The applicability of these measures and their costs will also vary across countries, depending on types of farms, their size, methods commercially available and experience. Costs tend to be higher on small farms in countries with little experience, and lower on large farms in countries with lots of experience.

Annex IX of the Gothenburg Protocol (UNECE, 1999) categorizes ammonia emission abatement measures as follows:

- Nitrogen management, taking account of the whole nitrogen cycle;
- Livestock feeding strategies;
- Low-emission animal housing systems;
- Low-emission manure storage systems;
- Low-emission manure spreading techniques;
- Possibilities for limiting ammonia emissions from the use of mineral fertilizers.

### Nitrogen management

Nitrogen management is seen as an integral measure of the whole nitrogen cycle, which may affect all sources of  $\text{NH}_3$  emissions and help to prevent pollution swapping between different sources, nitrogen compounds and environmental compartments. It is based on the premise that decreasing the nitrogen surplus and increasing nitrogen use efficiency contributes to lower emissions of  $\text{NH}_3$ . On

mixed livestock farms, between 10 to 40% of the nitrogen surplus is related to NH<sub>3</sub> emissions, while the remaining part will be lost through N leaching and denitrification (Oenema and Velthof, 2012; UNECE, 2012b).

Oenema et al (2011) provides a conceptual framework for developing integrated approaches to N management. The 'optimum' level of integration depends on many factors (e.g. farm type), and it remains a challenge to define such optima for various situations and cases. The integral approach can be seen as an extension to known abatement options that would allow to avoid losses and to better provide agricultural production (animals, plants) with the needed nitrogen amounts. The principles in the nitrogen management to decrease emissions of N are (UNECE, 2012b):

- All N sources on the farm are fully considered in a coherent whole-farm perspective and a whole N cycle perspective;
- All N sources are stored and handled properly;
- Amounts of N used are strictly according to the needs of growing plants and animals;
- Nitrogen sources are used in a timely manner, using the appropriate techniques, in the appropriate amounts and appropriate place;
- All possible N loss pathways are considered in a coherent manner.

UNECE (2012b) summarises the indicative ranges for target nitrogen (N) surplus and N use efficiency (NUE) as function of farming system, crop species and animal categories as follows:

<i>Farming systems</i>	<i>Species/categories</i>	<i>NUE, kg/kg</i>	<i>N surplus, kg/ha/yr</i>	<i>Comments</i>
Specialized cropping systems	Arable crops	0.6-0.9	0-50	Cereals have high, root crops low NUE
	Vegetables	0.4-0.8	50-100	Leafy vegetables have low NUE
	Fruits	0.6-0.9	0-50	
Grassland-based ruminant systems	Dairy cattle	0.3-0.5	100-150	High milk yield, high NUE; Low stocking density, low N surplus
	Beef cattle	0.2-0.4	50-150	Veal production, high NUE; 2 year old beef cattle, low NUE
	Sheep & goats	0.2-0.3	50-150	Sheep & goats
Mixed crop-animal systems	Dairy cattle	0.4-0.6	50-150	High milk yield, high NUE; Concentrate feeding, high NUE
	Beef cattle	0.3-0.5	50-150	
	Pigs	0.3-0.6	50-150	
	Poultry	0.3-0.6	50-150	
	Other animals	0.3-0.6	50-150	
Landless systems	Dairy cattle	0.8-0.9	n.a.*	N Output via milk, animals, manure + N-loss ~equals N input; N surplus is gaseous N losses from housing and storages.
	Beef cattle	0.8-0.9	n.a.	
	Pigs	0.7-0.9	n.a.	
	Poultry	0.6-0.9	n.a.	
	Other animals	0.7-0.9	n.a.	

\* Not applicable, as these farms have essentially no land. However, the N surplus can be expressed in kg per farm per year. In the case that all animal products, including animal manure and all residues and wastes, are exported, the target N surplus can be between 0 - 1000 kg per farm per year, depending on farm size and gaseous N losses.

## Animal feeding strategies

The quantity of nitrogen excretion in animal faeces and urine is linearly dependent on the intake of nitrogen in food (protein). For example, about 65% of the nitrogen ingested by pigs is not used for growth and is excreted. Feeding according to the protein requirements of the animals reduces the oversupply of protein, leading to a reduction in nitrogen excretions and thus a decrease in ammonia emissions.

Feeding strategies are implemented through:

- phase feeding,
- low-protein feeding, with or without supplementation of specific synthetic amino acids and ruminal bypass protein,
- increasing the non-starch polysaccharide content of the feed,
- supplementation of pH-lowering substances, such as benzoic acid.

Phase feeding is an effective and economically attractive measure even if requiring additional installations. These strategies decrease ammonia emissions from manure in housing, storage and during application to crop land (UNECE, 2012b).

Phase feeding implies adapting the amount of proteins in the food to the needs of the animals. For instance young animals and high-productive animals require more protein than older, less-productive animals. In average this measure leads to a NH<sub>3</sub> emission reduction of 10% for a 1% reduction in the mean protein content in the diet, but efficiencies depend strongly on the animal categories.

Low-protein animal feeding is one of the most cost-effective ways to reduce NH<sub>3</sub> emissions. It has no implications on animal health as long as the requirements for all amino-acids are met. It is most applicable to housed animals, as experts have indicated that the practical applicability of feeding strategies to grazing animals is limited (UNECE, 2011). Never the less, emissions from pastures are low and grazing itself is indicated as a measure. For example, total annual emissions (from housing, storage and spreading) from dairy systems may decrease by up to 50% with nearly all-day grazing, as compared to animals that are fully confined (UNECE, 2012b).

## Animal housing

The available techniques to reduce NH<sub>3</sub> emissions from animal housing have been well known for decades and apply one or more of the following principles:

Principles	NH <sub>3</sub> emission reduction*
• Decrease the surface area fouled by manure	15-25% in pig housing
• Rapid removal of urine and rapid separation of faeces and urine	25-46%
• Decrease air temperature and velocity above the manure	up to 20%
• Reduce PH of the manure	up to 60% in pig and cattle housing systems
• Reduce temperature of the manure	45-75%
• Dry the manure (e.g. poultry litter)	up to 70%
• Scrubbing ammonia from exhaust air	70-95%
• Decrease housing time by increasing grazing time	10-50%, but some emission swapping

\* Oenema and Velthof (2012)

All of these principles are scientifically sound and practically proven. Different housing systems and environmental conditions are required by different animal categories, requiring therefore different techniques for the application of the above mentioned principles with different results in NH<sub>3</sub> emission reduction. UNECE (2012b) lists the NH<sub>3</sub> emission reduction techniques for animal housing in different categories and their emission reduction levels as follows:

Category	Emission reduction*
• Existing pig and poultry housing on farms with >2000 fattening pigs or >750 sows or >40 000 poultry	20%
• New or largely rebuilt cattle housing	0-70%
• New or largely rebuilt pig housing	20 to 90%
• New and largely rebuilt broiler housing	20-90%
• New and largely rebuilt layer housing	20 to 90%
• New and largely rebuilt animal housing on farms for animals other than those already listed in this table	0 to 90%

\* Compared with reference as defined in UNECE (2012b)

### Manure storage

Measures to reduce NH<sub>3</sub> emissions from manure storage systems apply one or more of the following principles: a) Decreasing the surface area where emissions can take place, i.e. through covering of the storage, encouraging crusting and increasing depth of storages; b) Reducing the pH and temperature of the manure; and c) Minimizing disturbances such as aeration (UNECE, 2012b).

The Nitrates directive requires modified storage conditions and storage times, which has implications on NH<sub>3</sub> emissions and mitigation potentials. The directive demands leak-tight manure storages and sufficient storage capacity, related to longer prohibition periods for manure application. However, the Nitrates directive does not require covered manure storages. Ammonia losses from slurry stores can be minimized by covering open stores. Döhler et al (2011) and Oenema and Velthof (2012) list the main natural and artificial covers solutions and their efficiencies as follows:

- **Natural floating covers**, by allowing the formation of a natural crust, are the simplest and most inexpensive form of slurry store covering. These covers primarily form on cattle slurry, but they also develop on pig slurry rich in fibre and dry matter, when mixing is reduced and manure input is below the surface. The reduction potential ranges between 30 and 80% for cattle slurry and between 20 and 70% for pig slurry.
- **Artificial floating covers** with chopped straw, peat, bark, etc, can have a higher reduction effect of 40% to 90%. However, the straw cover must be at least 10 cm thick. The effectiveness of both cover variants (natural floating cover and chopped straw) is limited on farms with frequent slurry distribution because the natural floating layer or the straw layer is destroyed temporarily or permanently. Straw covers must be replaced after stirring. It is applicable to large earth-banked lagoons and concrete or steel tanks.
- **Granule covers** have lower material losses than in the case of straw. They float again shortly after the slurry has been stirred. Therefore, only a small amount of the granules is spread with the slurry. However, it is necessary to replace the lost material. Emission losses are reduced by 60 to 90%.

- **Floating sheets** also have a reduction potential from 60 to 90%. Their advantage lies in low maintenance requirements. Precipitation water must be led or pumped into the slurry lying underneath.
- **Floating bodies and solid covers**, such as a concrete cover, a tent roof, or a plastic cover, have the highest reduction potential of up to 95% (60-95%). Floating bodies are only suitable for liquid pig slurry without a natural floating layer. Solid covers have the longest service life and low maintenance requirements, and avoid rainwater input.

## Manure application

Low-emission manure application techniques involve machinery that (i) decreases the exposed surface area of slurries applied to surface soil through band application, injection, incorporation, and/or (ii) decreases the time that emissions can take place, i.e. buries slurry or solid manures through injection or incorporation into the soil; and (iii) decreases the source strength of the emitting surface, i.e., through lowering the pH and  $\text{NH}_4$  concentration of the manure (through dilution). Examples of such application techniques are:

Measures	Emission reduction*
<ul style="list-style-type: none"> <li>• The band-spreading of slurry at the soil surface, which deposits the slurry on the soil in parallel bands using <b>trailing hose</b>, suitable for arable grassland.</li> </ul>	30-35%
<ul style="list-style-type: none"> <li>• The band-spreading of slurry at the soil surface, which deposits the slurry on the soil in parallel bands using <b>trailing shoe</b> methods, suitable for arable grassland.</li> </ul>	30-60%
<ul style="list-style-type: none"> <li>• <b>Slurry injection - open slots</b>, suitable for application on grassland and in growing crop stands, with slopes &lt; 15% and low stone content.</li> </ul>	60-80% (Oenema and Velthof, 2012) 70% (UNECE, 2012b)
<ul style="list-style-type: none"> <li>• <b>Slurry injection - closed slots</b>, where the slurry is fully covered after injection by closing the slots with press wheels or rollers fitted behind the injection tines. Deeper injection is required when greater volumes of manure are injected to avoid manure oozing to the surface</li> </ul>	70-90% (Oenema and Velthof, 2012). 80% (shallow slot 5-10 cm); 90% (deep injection >15cm) (UNECE, 2012b)
<ul style="list-style-type: none"> <li>• <b>Incorporation of surface-applied solid manure and slurry into soil</b>. Applicable in arable soils without vegetation (in pre-sowing season)</li> </ul>	When the manure is completely buried within the soil immediately after application: 80-90%. Incorporation within 4 h.: 45-65%. Incorporation within 24 h.: 30% for slurry, 35% for solid manure from cattle and pig and 55% from poultry
<ul style="list-style-type: none"> <li>• <b>Dilution of slurry</b> by at least 50% in low pressure water irrigation systems, suitable for arable grassland.</li> </ul>	30%

\* Based on Oenema and Velthof (2012), UNECE (2007) and UNECE (2012b).

Known techniques to reduce  $\text{NH}_3$  emissions from manure application are now applied at a much larger scale and in more countries, lowering costs, especially for larger farms. The up-scaling has

taken place mainly through contractors as many animal farmers in e.g. the Netherlands and Denmark have outsourced manure application to specialized contractors.

The timing of manure application has changed during the last decade as a result of the Nitrates directive. The directive imposes longer prohibition periods for manure application to decrease the risk of nitrate leaching. Ammonia emissions might however increase with longer prohibition periods because manure application is then more concentrated during the growing seasons with higher temperatures and less precipitation (Oenema and Velthof, 2012).

### Fertilizer application

Measures to reduce emissions of NH<sub>3</sub> from the application of urea and ammonium based fertilizers are based on one or more of the following principles (UNECE, 2012b):

- decrease the surface area where emissions may occur, i.e. through band application, injection, incorporation (but a rapid increase in pH in concentrated bands of urea, especially where there is high crop residue, may lead to high emissions due to rise in pH);
- decrease the period emissions may occur, i.e. through rapid incorporation of fertilizers into the soil or via irrigation;
- decrease the source strength of the emitting surface, i.e., through urease inhibitors, blending and acidifying substances;
- ban its use (as in the case of ammonium (bi)carbonate).

UNECE (2012b) summarized the ammonia emission reduction techniques for application of urea and ammonium-based fertilizers and their emission reduction levels as follows:

Fertilizer type	Application techniques	Emission reduction* %
Urea	Injection	> 80
	Urease inhibitors	> 30
	Incorporation following surface application	> 50
	Surface spreading with irrigation	> 40
Ammonium carbonate	Ban	~100
Ammonium-based fertilizers	Injection	> 80
	Incorporation following surface application	> 50
	Surface spreading with irrigation	> 40

\* Compared to the broadcast application of the urea- and ammonium based fertilizers

## 6. Conclusions

The main outcomes from the literature review are summarized in the following points:

- Burning of agricultural waste and agricultural operations at farm level (e.g. plowing, harrowing, disking and cultivating) are main sources of primary and secondary PM<sub>2.5</sub> and PM<sub>10</sub>. However, studies about their contribution to air quality and PM concentration are scarce, and thus a significant knowledge gap exists.
- Agricultural emissions contribute to about 2.5 and 8.5% of the total emissions of primary PM<sub>2.5</sub> and PM<sub>10</sub>, respectively. In addition, agriculture is responsible for 94% of the total NH<sub>3</sub> emissions, which is one of the precursors of SIA.
- Based on the studies published in the literature, the contribution of SIA to PM is mainly to the fine fraction (PM<sub>2.5</sub>), contributing around 30-45% of the PM<sub>2.5</sub>, reaching values above 40% for high PM<sub>10</sub> concentration levels or exceedances days (i.e. PM<sub>10</sub> > 50 µgm<sup>-3</sup>).
- The contribution of total ammonium to PM has been established in different studies ranging from around 7% to 10% of PM<sub>2.5</sub> in urban background, and below 10% of PM<sub>10</sub>. This contribution is essential in the sense that ammonia levels contribute to SIA formation (the presence of NH<sub>4</sub><sup>+</sup> being the limiting factor for the formation of ammonium sulphate and nitrate). Thus, ammonia emissions favour the increase of the whole SIA fraction in PM<sub>10</sub> and PM<sub>2.5</sub>, contributing with high percentages to the total PM load.
- Differences in SIA concentration levels between locations (e.g. rural vs. urban) have been reported to be small, and interpreted as a confirmation that SIA compounds are mainly long-range transported from outside the urban environment, even though there are also urban sources of NH<sub>3</sub>.
- Several studies pointed out about reducing NH<sub>3</sub> emissions as an effective way of reducing SIA concentration levels, thus PM concentration levels, and the number of PM<sub>10</sub> daily limit exceedances (Erisman and Schaap, 2004; Putaud et al., 2010). A reduction in primary PM (PPM) emissions is, nevertheless, the most effective way of reducing PM in terms of reduced PM concentration mass per kT emission reduction of PPM or precursor gas.
- The most important abatement techniques and strategies to reduce NH<sub>3</sub> emissions from agriculture are scientifically sound and practically proven. Their description, applicability and emission reduction potential is well described in recent literature, as UNECE (2012b) and Oenema and Velthof (2012).

## 7. References

- Aarnink A.J.A., Roelofs P.F.M.M., Ellen H.H., Gunnink H. (1999) Dust sources in animal houses. In: Proceedings of International Symposium on Dust Control in Animal Production Facilities, Aarhus, Denmark.
- AirParif, (2012) Source apportionment of airborne particles in the Ile-de-France region. Final Report, Surveillance de la Qualité de l'Air en Ile-de-France. Paris.
- Amato F., Viana M., Richard A., Furger M., Prévôt A.S.H., Nava S., Lucarelli F., Bukowiecki N., Alastuey A., Reche C., Moreno T., Pandolfi, M., Pey J., Querol X. (2011) Size and time-resolved roadside enrichment of atmospheric particulate pollutants. *Atmos Chem Phys* 11, 2917-2931.
- Bogman P., Cornelis W., Rollé H., Gabriels D. (2007) Prediction of TSP and PM10 emissions from agricultural operations in Flanders, Belgium. DustConf 2007, Maastricht, Netherlands. Available at [http://www.dustconf.org/CLIENT/DUSTCONF /UPLOAD/S9/BOGMAN\\_B.PDF](http://www.dustconf.org/CLIENT/DUSTCONF /UPLOAD/S9/BOGMAN_B.PDF)
- Cambra-López M., Aarnink A.J.A., Zhao Y., Calvet S., Torres A.G. (2010) Airborne particulate matter from livestock production system: a review of an air pollution problem. *Environ Pollut*, 158, 1-17.
- CITEPA (2010) Inventaire des émissions de polluants atmosphériques en France au titre de la convention sur la pollution atmosphérique transfrontalière à longue distance et de la directive européenne relative aux plafonds d'émissions nationaux (NEC). Format CEE-NU, édition mars 2010. CITEPA, Paris. <http://citepa.org/publications/Inventaires.htm>.
- Chardon W.J., van der Hoek K.W. (2002) Berekeningsmethode voor de emissie van fijn stof vanuit de landbouw (Calculation method for emission of fine dust from agriculture) Alterra/RIVM, Wageningen, The Netherlands, 35 pp.
- de Gouw, J., Jiménez, J.L. (2009) Organic Aerosols in the Earth's Atmosphere. *Environ Sci Tech* 43, 7614–7618.
- Döhler H., Eurich-Menden B., Rößler R., Vandré R., Wulf S. (2011) UN ECE-Convention on long-range transboundary air pollution – Task Force on Reactive Nitrogen Systematic cost-benefit analysis of reduction measures for ammonia emissions in agriculture for national cost estimates. Federal Environment Agency in Germany (Umweltbundesamt). Report No. (UBA-FB) 001527/E. (<http://www.uba.de/uba-info-medien/4207.html>, Assessed February 2013).
- Dutch Emission Registration (2013) <http://www.emissieregistratie.nl/ERPUBLIEK/bumper.en.aspx> (Assessed in April 2013).
- EEA (2005) Agriculture and environment in EU-15— the IRENA indicator report. European Environmental Agency, Report 6/2005.
- EEA (2013) National emissions reported to the Convention on Long-range Transboundary Air Pollution (LRTAP Convention); <http://www.eea.europa.eu/data-and-maps/data/data-viewers/air-emissions-viewer-lrtap> (Assessed January 2013).



- EMEP-CORINAIR (2007) Atmospheric Emission Inventory Guidebook. December 2007 Update, third ed. EEA, Copenhagen, Denmark.
- Erismán J.W., Schaap M. (2004). The need for ammonia abatement with respect to secondary PM reductions in Europe. *Environ Poll* 129, 159–163.
- Erismán J.W., Bleeker A., Hensen A., Vermeulen, A. (2008) Agricultural air quality in Europe and the future perspectives. *Atmos Environ* 42; 3209-3217.
- Funk R., Reuter H.I., Hoffmann C., Engel W., Öttl D. (2008) Effect of moisture on fine dust emission from tillage operations on agricultural soils. *Earth Surf Proc Land*, 33, 1851–1863.
- Hays M., Fine P.M., Gernon C.D., Kleeman M., Gullett B.K. (2005) Open burning of agricultural biomass: Physical and chemical properties of particle-phase emissions. *Atmos Environ*, 39, 6747-6764.
- Hellsten S., Dragosits U., Place C.J., Vieno M., Dore A.J., Misselbrook T.H., Tang Y.S., Sutton M.A., (2008) Modelling the spatial distribution of ammonia emissions in the UK. *Environ Pollut*, 370-379.
- HPA (2006) Health Protection Agency, Position Statement: Intensive farming. Available at: [http://www.hpa.org.uk/webc/HPAwebFile/HPAweb\\_C/1194947378905](http://www.hpa.org.uk/webc/HPAwebFile/HPAweb_C/1194947378905).
- Hristov A.N. (2011) Technical note: Contribution of ammonia emitted from livestock to atmospheric fine particulate matter (PM<sub>2.5</sub>) in the United States. *J Dairy Sci*, 94:3330-3136.
- Huijsmans J.F.M., Hol J.M.G., Vermeulen G.D. (2003) Effect of application method, manure characteristics, weather and field conditions on ammonia volatilization from manure applied to arable land. *Atmos Environ*, 37, 3669–3680.
- Karlsson P.E., Ferm M., Tømmervik H., Hole L.R., Karlsson G.P., Ruoho-Airola T., Aas W., Hellsten S., Akselsson C., Mikkelsen T.N., Nihlgård B. (2013) Biomass burning in eastern Europe during spring 2006 caused high deposition of ammonium in northern Fennoscandia. *Environ Poll*, 176, 71-79.
- Keck L., Wittmaack K. (2005) Effect of filter type and temperature on volatilisation losses from ammonium salts in aerosol matter. *Atmos Environ* 39, 4093-4100.
- Keshtkar H., Ashbaugh, L. L. (2007) Size distribution of polycyclic aromatic hydrocarbon particulate emission factors from agricultural burning. *Atmos Environ*, 41, 2729–2739.
- Kumar A., Howard C.J., Derrick D., Malkina I.L., Mitloehner F.M., Kleeman M.J., Alaimo C.P., Flocchini R.G., Green P.G. (2011) Determination of volatile organic compound emissions and ozone formation from spraying solvent-based pesticides. *J Environ Qual*, 40, 1423-31.
- Liousse C., Penner J.E., Chuang C., Walton J.J., Eddleman H., Cachier H. (1996) A global three-dimensional model study of carbonaceous aerosols. *J Geophys Res* 101, 19411–19432.
- Moldanová J., Grennfelt P., Jonsson A., (2011). Chapter 8: Nitrogen as threat to European air quality. *The European Nitrogen Assessment*, ed. Mark A. Sutton, Clare M. Howard, Jan Willem Erismán, Gilles Billen, Albert Bleeker, Peringe Grennfelt, Hans van Grinsven and Bruna Grizzetti. Published by

Cambridge University Press. Available at: [http://www.nine-esf.org/sites/nine-esf.org/files/ena\\_doc/ENA\\_pdfs/ENA\\_c18.pdf](http://www.nine-esf.org/sites/nine-esf.org/files/ena_doc/ENA_pdfs/ENA_c18.pdf).

NERI (2011) National Environmental Research Institute, Aarhus University, Danish emission inventory for agriculture. Inventories 1985-2009. Available at: <http://www2.dmu.dk/pub/fr810.pdf>.

Niemi J.V., Tervahattu H., Vehkamäki H., Kulmala M., Koskentalo T., Sillanpää M., Rantamäki M., (2004) Characterization and source identification of a fine particle episode in Finland. *Atmos Environ* 38, 5003-5012.

Novak S.M., Fiorelli J.L., (2009) Greenhouse gases and ammonia emissions from organic mixed crop-dairy systems: a critical review of mitigation options. *Agron Sustain Dv* 30, 215-236.

Oanh K.T.N., Ly B.T., Tipayarom D., Manadhar B.R., Prapat P., Simpson C.D., Sally Liu L.-J. (2011) Characterization of particulate matter emission from open burning of rice straw. *Atmos Environ* 45, 493–502.

Oenema, O., Oudendag, D.A. , Witzke, H.P., Monteny, G.J., Velthof, G.L., Pietrzak, S., Pinto, M., Britz, W., Schwaiger, E., Erisman, J.W., de Vries, W. van Grinsven, J.J.M., and Sutton, M. (2007) Integrated measures in agriculture to reduce NH<sub>3</sub> emissions; final summary report. Alterra report. ([http://ec.europa.eu/environment/archives/cafe/activities/pdf/alterra\\_final\\_report.pdf](http://ec.europa.eu/environment/archives/cafe/activities/pdf/alterra_final_report.pdf), viewed February 2013).

Oenema, O., Salomez, J., Branquinho, C. et al (2011) Developing integrated approaches to nitrogen Management. In: *The European Nitrogen Assessment*, ed. M. A. Sutton , C. M. Howard , J. W. Erisman et al. , Cambridge University Press. ([http://www.nine-esf.org/sites/nine-esf.org/files/ena\\_doc/ENA\\_pdfs/ENA\\_c23.pdf](http://www.nine-esf.org/sites/nine-esf.org/files/ena_doc/ENA_pdfs/ENA_c23.pdf), viewed February 2013)

Oenema O. Velthof G. L. (2007) Analysis of international and European policy instruments: pollution swapping. Alterra, Wageningen, 31 May 2007. ([http://ec.europa.eu/environment/archives/cafe/activities/pdf/alterra\\_final\\_report\\_task2.pdf](http://ec.europa.eu/environment/archives/cafe/activities/pdf/alterra_final_report_task2.pdf), Assessed February 2013)

Oenema O. Velthof G.L. (2012) Emissions from agriculture and their control potentials. TSAP Report #3. IIASA, June 2012. (<http://gains.iiasa.ac.at/index.php/policyapplications/tsap/394-emissions-from-agriculture-and-their-control-potentials>, assessed February 2013)

Pandolfi M., Amato F., Reche C., Querol X., Alastuey A., (2012) Summer ammonia measurements in a densely populated Mediterranean city. *Atmos Chem Phys*, 12, 7557-7575.

Perrino C., Catrambone A., Di Menno Di Bucchianico A., Allegrini I. (2002) Gaseous ammonia in the urban area of Rome, Italy and its relationship with traffic emissions. *Atmos Environ*, 36, 5385–5394.

Putaud J.-P., Raes F., van Dingenen R., Brüggemann E., Facchini A.-C., Decesari S., Fuzzi S., Gehrig R., Hüglin C., Laj P. Lorbeer G., Maenhaut W., Mihalopoulos N., Müller K., Querol X., Rodriguez S., Schneider J., Spindler G., ten Brink H., Tørseth K., Wiedensohler A. (2004). A European aerosol phenomenology-2: chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe. *Atmos Environ*, 38, 2579 – 2595.

Putaud J.-P., van Dingenen R., Alastuey A., Bauer H., Birmili W., Cyrys J., Flentje H., Fuzzi S., Gehrig R., Hansson H.C., Harrison R.M., Herrmann H., Hitenberger R., Hüglin C., Jones A.M., Kasper-Giebl A., Kiss G., Koussa A., Kuhlbusch T.A.J., Löschan G., Maenhaut W., Molnar A., Moreno, T., Pekkanen J., Perrino C., Pitz M., Puxbaum H., Querol X., Rodriguez S., Salma I., Schwarz J., Smolik J., Schneider J., Spindler G., ten Brink H., Tursic J., Viana M., Wiedensohler A., Raes R. (2010) A European aerosol phenomenology – 3: Physical and chemical characteristics of particulate matter from 60 rural, urban and kerbside sites across Europe. *Atmos Environ*, 44, 1308-1320.

Quass U., T. Kuhlbusch, M. Koch (2004): Identifizierung von Quellgruppen für die Feinstaubfraktion. IUTA-Report, LP15.

Querol X., Alastuey A., Ruiz C.R., Artíñano B., Hansson H.C., Harrison R.M., Buringh E., Brink H.M.t., Lutz M., Bruckmann P., Straehl P., Schneider J. (2004). Speciation and origin of PM10 and PM2.5 in selected European cities. *Atmos Environ* 38, 6547-6555.

Querol, X., Alastuey, A., Moreno, T., Viana, M., Castillo, S., Pey, J., Rodríguez, S., Artíñano, B., Salvador, P., Sánchez, M., Garcia Dos Santos, S., Herce Garraleta, M.D., Fernandez-Patier, R., Moreno-Grau, S., Minguillón, M.C., Monfort, E., Sanz, M.J., Palomo-Marín, R., Pinilla-Gil, E., Cuevas, E. (2008). Spatial and temporal variations in airborne particulate matter (PM10 and PM2.5) across Spain 1999-2005. *Atmos Environ* 42, 3964–3979.

Reche C., Viana M., Amato F., Alastuey A., Moreno T., Hillamo R., Teinilä K., Saarnio K., Seco R., Peñuelas J., Mohr C., Prévôt A.S.H., Querol X., (2012a). Biomass burning contributions to urban aerosols in a coastal Mediterranean city. *Atmos Environ*, 42, 1941–1957.

Reche C., Viana M., Pandolfi M., Alastuey A., Moreno T., Amato F., Ripoll A., Querol X., (2012b) Urban NH<sub>3</sub> levels and sources in a Mediterranean environment. *Atmos Environ*, 57, 153–164.

Schaap, M. (2003): On the importance of aerosol nitrate over Europe, Thesis, University of Utrecht, December, 2003, Utrecht, the Netherlands.

Sillanpää, M., R. Hillamo, S. Saarikoski, A. Frey, A. Pennanen, U. Makkonen, Z. Spolnik, R. Van Grieken, M. Braniš, B. Brunekreef, M. Chalbot, T. Kuhlbusch, J. Sunyer, V. Kerminen, M. Kulmala and R.O. Salonen (2006): Chemical composition and mass closure of particulate matter at six urban sites in Europe. *Atmos Environ* 40 212-223.

Smith P., Martino D., Cai Z., Gwary D., Janzen H., Kumar P., McCarl B., Ogle S., O'Mara F., Rice C., Scholes B., Sirotenko O. (2007) Agriculture. p. 497–540. In B. Metz et al. (ed.) *Climate change 2007: Mitigation. Contribution of Working Group III to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*. Cambridge Univ. Press, Cambridge, UK, and New York.

Torfs, R., F. Deutsch, L. Schrooten, S. Broekx, J. Vankerkom, C. Matheeußen, E. Roekens, F. Fierens, G. Dumont and M. Bossuyt (2007): MIRA Milieurapport Vlaanderen.

Thunis P, Cuvelier C (editors), Roberts P, White L, Post L, Tarrason L, Tsyro S, Stern R, Kerschbaumer A, Rouil L, Bessagnet B, Bergström R, Schaap M, Boersen G, Boersen P (2008) EURODELTA II – Evaluation of a Sectoral Approach to Integrated Assessment Modelling Including the Mediterranean Sea. EUR 23444 EN. Luxembourg (Luxembourg): OPOCE; 2008. JRC41801.

Turn S.Q., Jenkins B.M., Chow J.C. (1997). Element characterization of particulate matter emitted from biomass burning: wind tunnel derived source profiles for herbaceous and wood fuels. *J Geophys Res* 102, 3683–3699.

UNECE (1999) Protocol to the 1979 Convention on Long-Range Transboundary Air Pollution to Abate Acidification, Eutrophication and Ground-Level Ozone. 65 S.  
(<http://www.unece.org/fileadmin/DAM/env/lrtap/full%20text/1999%20Multi.E.Amended.2005.pdf>, viewed February 2013).

UNECE (2007) Guidance Document on Control Techniques for Preventing and Abating Emissions of Ammonia. 35 S.  
(<http://www.unece.org/env/documents/2007/eb/wg5/WGSR40/ece.eb.air.wg.5.2007.13.e.pdf>; Assessed February 2013).

UNECE (2011) Options for revising the annexes to the Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone: annex IX. Forty-ninth session. Geneva, 12–16 September 2011. Item 3 (c) of the provisional agenda. ECE/EB.AIR/WG.5/2011/16.  
(<http://www.unece.org/fileadmin/DAM/env/documents/2011/eb/wg5/WGSR49/ece.eb.air.wg.5.2011.16.e.pdf>, viewed February 2013).

UNECE (2012a) The 1999 Gothenburg Protocol to abate Acidification, Eutrophication and Ground-level Ozone under the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP) agreed on a revision of the Protocol (<http://www.unece.org/index.php?id=29858>) 8 July 2012.

UNECE (2012b) Draft decision on adoption of guidance document on preventing and abating ammonia emissions from agricultural sources. Paper ECE/EB.AIR/2012/L.9, October 2, 2012, UNECE, Geneva.  
([http://www.unece.org/fileadmin/DAM/env/documents/2012/EB/ECE\\_EB.AIR.2012.L.9\\_Draft\\_decision\\_on\\_adoption\\_of\\_guidance\\_document\\_GP\\_Ammonia\\_emissions\\_from\\_agriculture.pdf](http://www.unece.org/fileadmin/DAM/env/documents/2012/EB/ECE_EB.AIR.2012.L.9_Draft_decision_on_adoption_of_guidance_document_GP_Ammonia_emissions_from_agriculture.pdf))

USEPA (2006) AP-42: Compilation of Air Pollutant Emission Factors Volume I: Stationary Point and Area Sources.

Viana M., López J.M., Querol X., Alastuey A., García-Gacío D., Blanco-Heras G., López-Mahía P., Piñeiro-Iglesias M., Sanz M.J., Sanz F., Chie X., Maenhaut W. (2008) Tracers and impact of open burning of rice straw residues on PM in Eastern Spain. *Atmos Environ*, 42, 1941-1957.

Viana, M., Reche, C., Amato, F., Alastuey, A., Querol, X., Moreno, T., Lucarelli, F., Nava, S., Cazolai, G., Chiari, M., Rico, M., 2013. Evidence of biomass burning aerosols in the Barcelona urban environment during winter time. *Atmospheric Environment*, in press.

Vogt E., Held A., Klemm O. (2005) Sources and concentrations of gaseous and particulate reduced nitrogen in the city of Münster (Germany). *Atmos Environ*, 38, 7393-7402.

WHO, (2006) Health risks of particulate matter from long-range transboundary air pollution. Available at [http://www.euro.who.int/\\_\\_data/assets/pdf\\_file/0006/78657/E88189.pdf](http://www.euro.who.int/__data/assets/pdf_file/0006/78657/E88189.pdf)

Weijers E.P., Sahan E., ten Brink H.M., Schaap M., Matthijsen J., Otjes R.P., van Arkel F., (2010) Contribution of secondary inorganic aerosols to PM<sub>10</sub> and PM<sub>2.5</sub> in the Netherlands; measurement

and modelling results; Netherlands Research Program on Particulate Matter, AH Bilthoven, The Netherlands.

Whyatt J.D., Metcalfe S.E. Nicholson J., Derwent R.G., Page T., Stedman J.R. (2007) Regional scale modelling of particulate matter in the UK, source attribution and an assessment of uncertainties. *Atmos Environ* 41, 3315–3327.

Witzke, H. P. Oenema, O. (2007) Assessment of most promising measures. Alterra, Wageningen, 31 May 2007.

([http://ec.europa.eu/environment/archives/cafe/activities/pdf/alterra\\_final\\_report\\_task3.pdf](http://ec.europa.eu/environment/archives/cafe/activities/pdf/alterra_final_report_task3.pdf), viewed February 2013).

Wu S.Y., Krishnan S., Zhang Y., Aneja V.P. (2008) Modelling atmospheric transport and fate of ammonia in southeast U.S., Part I: Evaluation of meteorological and chemical predictions. *Atmos Environ*, 42, 3419–3436.

Yates S.R., McConnell L.L., Hapeman C.J., Papiernik S.K., Gao S., Trabue S.L. (2011) Managing Agricultural Emissions to the Atmosphere: State of the Science, Fate and Mitigation, and Identifying Research Gaps. *J Environ Qual*, 40, 1347-1358.

Zhang Y.X., Shao M., Zhang Y.H., Zeng L.M., He L.Y., Zhu B., Wei Y.J., Zhu X.L. (2007) Source profiles of particulate organic matters emitted from cereal straw burnings. *J Environ Sci*, 19, 167-175.