Secondary inorganic aerosols from agriculture in Europe

ETC/ACM Technical Paper 2016/4
December 2016

Mar Viana, Xavier Querol, Andrés Alastuey,
Augustin Colette, Florian Couvidat, Jérôme Drevet,
Bertrand Bessagnet, Anke Lükewille

European Topic Centre
on Air Pollution and
Climate Change Mitigation

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 Aether CHMI CSIC EMISIA INERIS NILU ÖKO-Institut ÖKO-Recherche PBL UAB UBA-V VITO 4Sfera

Front page picture:
Source: SXC #106396, http://www.freeimages.com. Content License by FreeImages.com

Author affiliation:
Mar Viana, Xavier Querol, Andrés Alastuey: Institute of Environmental Assessment and Water Research (IDAEA-CSIC; ES)
Augustin Colette, Florian Couvidat, Jérôme Drevet, Bertrand Bessagne: INERIS (FR)
Anke Lükewille: EEA (DK)

DISCLAIMER

ETC/ACM Technical Paper 2016/4
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
Website http://acm.eionet.europa.eu/

Contents

1. Introduction

Airborne fine particulate matter ($PM_{2.5}$) is Europe's most problematic air pollutant in terms of harm to human health (Erisman et al., 2008; EEA, 2015). Thus, health benefits can be expected from all efforts to reduce the mass of $PM_{2.5}$ in the air (Brunekreef et al., 2015).

In the EU 28, agricultural emissions contribute to about 5% of the total emissions of primary $PM_{2.5}$, i.e. of particulate matter emitted directly into the air (EEA, 2016a). Burning of agricultural waste and agricultural operations at farm level (e.g. ploughing, harrowing, disking and cultivating) are main sources. However, almost 95% of all ammonia (NH_3) emissions to the air are from agricultural activities, and NH_3 contributes significantly to the formation of secondary $PM_{2.5}$ in the atmosphere.

1.1 Secondary particulate matter: Secondary Inorganic Aerosol (SIA) formation

In Europe, secondary PM (both inorganic aerosols, SIA, and organic aerosols, SOA) contribute 70% or more to the $PM_{2.5}$ background concentrations (Putaud et al 2010). Therefore, to reduce PM concentrations it is necessary to lower the precursor emissions that give rise to secondary PM. The precursors of secondary PM are sulphur dioxide (SO_2), nitrogen oxides (NOx), ammonia (NH_3), and anthropogenic and biogenic volatile organic compounds (VOCs). Secondary PM components are sulphate (SO_4^{2-}), nitrate (NO_3^-) and ammonium (NH_4^+), and organic carbonaceous compounds. The main SIA components in PM are ammonium sulphate ($(NH_4)_2SO_4$) and ammonium nitrate (NH_4NO_3). These salts result from the neutralization of sulphuric acid (H_2SO_4) and nitric acid (HNO_3) with ammonia (NH_3). While NH_3 is mainly emitted from agriculture, SO_2 and NOx emissions have mainly an industrial and traffic-related origin. SO_2 emissions were reduced considerably over the last decades, and NH_4NO_3 has become the main contributor to SIA (Harrison et al., 2013; ETC, 2013).

Box 1. The role of NH_3 in PM formation

The formation of $(NH_4)_2SO_4$ from and NH_4NO_3 in the atmosphere compete with each other for (1) the available OH-radicals to form H_2SO_4 and HNO_3, respectively, and (2) for the availability of NH_3. Ammonia tends to be trapped to preferably form $(NH_4)_2SO_4$, thus the formation of NH_4NO_3 will depend on the availability of NH_3. Due to its stability and consequent long atmospheric residence time, $(NH_4)_2SO_4$ is a well-known tracer of long-range transport of PM. NH_4NO_3 is unstable at temperatures higher than 25-30ºC.

1.2 Agricultural sources of SIA

The main source of NH_3 emissions in the agricultural sector is manure from livestock production, mainly through the processes of storage and management. The NH_3 content in manure and therefore its volatilisation is dependent on livestock type. Emissions primarily arise from the decomposition of urea in animal wastes and uric acid in poultry wastes.

Emissions depend on the animal species, age, weight, diet, housing systems, waste management and liquid manure storage techniques. (ETC, 2013; Steinmann et al., 2015).

The application of NH_3 fertilizers to the soil surface results in NH_3 emission to the atmosphere by volatilisation, especially from soils with a high pH values in the soil solution (i.e. pH higher than 7). In European countries, NH_3 volatilisation from field-applied fresh manure, a commonly used organic fertilizer, is a major contributor to the overall NH_3 emissions from the agricultural sector (Sutton et al., 2011; Vonk et al., 2016).

1.3 Overview of measures to mitigate air pollutant emissions from agriculture

Emission reductions of air and greenhouse gas (GHG) pollutants from agriculture as well as adaptation to climate change are major challenges that the European agricultural sector will need to face over the coming years. Measures like covering liquid storage facilities have a relatively low GHG reduction potential (0.1 Tg CO_2-eq/year), but can decrease NH_3 emissions significantly. Modern application techniques of manure on soils (injecting instead of spraying) have also a high potential to reduce NH_3 emissions. Both measures are easy to implement by farmers. Nitrogen balance at farm level (e.g. avoiding urea fertilizer losses) has a high potential in both reduction of GHG and NH_3 emissions.

Box. 2 Cost-effective measures to mitigate air pollutant and greenhouse gas emissions from agriculture

---

The AgriClimateChange European project (European Parliament, 2014) identified the following measures to mitigate emissions of air pollutants and GHGs from agriculture:

- four agronomic measures: 1) nitrogen balance at farm level, 2) introduction of leguminous plants on arable land to improve fertility and increase carbon sequestration, 3) conservation agriculture based on no-tillage to increase carbon sequestration, and 4) implementation of cover crops to restore fertility and reduce the need to use N fertilisers;
- three livestock measures: 1) manure storage covering, 2) manure spreading closer to the ground, and 3) use of manure and farm residues to feed biogas plants;
- four energy measures: 1) use of biomass for heating, 2) photovoltaic installation, 3) fuel reduction, and 4) electricity reduction;
- one agro-environmental measure (AEM) based on maintaining and encouraging farms to develop low carbon farming practices based on the farmers skills and interests.

The implementation of the six inexpensive measures above (nitrogen balance, low carbon AEM, electricity reduction, fuel reduction, leguminous plants and manure spreading) could reduce GHG emissions by 61.7 Tg CO_2-eq/year. The advantage is that all these measures are easy (or average, in the case of leguminous plants) for farmers to implement. Other measures such as manure storage and photovoltaic installations are also easy to implement, but are medium- to high-cost measures.

The implementation of cover crops has a high GHG emissions reduction potential and a low-average cost; however it is difficult to implement by farmers. Similarly, the implementation of biogas plants has also a high reduction GHG emissions potential, but it is difficult for farmers to implement and moreover has a high cost associated.

When focusing specifically ammonia, the most important abatement techniques and strategies to reduce NH_3 emissions from agriculture are summarised in López-Aparicio et al. (2013; see also UNECE, 1999, 2012; Amann et al., 2011; Oenema and Velthof, 2012; Henderson et al. 2015):

- 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.

Finally, regarding primary airborne particles, agricultural field operations causing primary PM emissions in conventional crop production include soil tillage and seed bed preparation, planting, fertilizer and pesticide application, harvesting and post-harvest processes (Arslan and Aybek, 2012). Therefore, these are the main activities to be targeted by mitigation measures.

1.4 Impacts of agricultural emissions on air quality

SIA originating from agricultural but also from urban and industrial sources constitutes a significant fraction of PM, and especially of the fine PM fraction ($PM_{2.5}$). This section focuses on the overall impacts of SIA on air quality, taking into account the major limitation that very few source-specific data are available in the literature for the agricultural sector.

As stated above, NH_3 emissions originate mainly from agricultural sources. However, this is not the case for NOx and SO_2. Erisman 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 NH_3 emissions are decreased. Backes et al. (2016) found that a reduction of NH_3 emissions by 50% led to a 24% reduction of the total $PM_{2.5}$ concentrations in north-western Europe. The observed reduction was mainly driven by reduced formation of ammonium nitrate (NH_4NO_3), and emission reductions during winter had a larger impact than during the rest of the year. However, Renner and Wolker (2010) ran modelling scenarios with changing NH_3 emissions for the SNAP-code[1] "agriculture". They concluded that, if NH_3 levels in the air are very high, NH_3 emission reductions have only a limited effect on SIA concentrations. According to these authors, the formation of SIA in the considered modelling domain (in Germany) was limited by the precursors SO_2 and NOx.

Table 1 shows a non-exhaustive summary of studies which provide quantitative information about SIA and its constituents in $PM_{2.5}$ and/or PM_{10}, at different scales and based on both measurements and modelling. In their review of 2010, Putaud et al. concluded that the sum of SO_4^{2-} and NO_3^- accounts for approximately 17-20% of the PM_{10} mass from rural to kerbside environments in Northern, central and Southern Europe (Table 1). However, higher contributions (40% of SIA in PM_{10}) have been reported in urban background environments in Germany (Quass et al., 2004) and in six European cities (i.e. Duisburg, Prague, Amsterdam, Helsinki, Barcelona and Athens; Sillanpaa et al., 2006). SIA contributions to PM_{10} of about 20-30% are reported at European level (WHO, 2003). In some European areas such as in The

[1] SNAP-codes: Standardized Nomenclature for Air Pollutants

Netherlands the highest SIA levels frequently coincide with exceedances of the PM_{10} daily limit value (Weijers et al., 2012). The source apportionment review by Belis et al. (2013) concluded that gaseous precursors emitted by combustion sources (e.g. traffic in urban areas) and agriculture undergo gas-to-particle conversion, and this atmospheric process is the strongest source for SIA mass concentrations over Europe, and the biggest contributor to the organic PM fraction (SOA). According to these authors, traffic and agriculture are the important source categories to target throughout the year in order to abate exceedances of air quality limit and target values set in the EU's Air Quality Directive (EU, 2008). The source apportionment of the SIA fraction remains almost impossible to determine by modelling or observation based techniques due to non-linearity in the chemistry schemes.

Table 1. Contribution of SIA (sulphate, nitrate, ammonium) to PM_{10} or $PM_{2.5}$ reported in the literature. Studies listed in chronological order.

Location/Scale	Contribution	Type of study	Study
Europe	Sulphate: 25-50% $PM_{2.5}$ Nitrate: 5-35% $PM_{2.5}$	Modelling	Schaap, 2003
24 European sites	Ammonium: 4-7% PM_{10} Ammonium: 7-10% $PM_{2.5}$	Experimental	Putaud et al., 2004
7 European sites	SIA: 10-42% PM_{10} SIA:32-45% $PM_{2.5}$	Experimental	Querol et al., 2004
Germany (urban background)	SIA: 40% PM_{10} (17% nitrate, 16% sulphate, 7% ammonium) SIA: 44% $PM_{2.5}$ (17% nitrate, 18% sulphate, 9% ammonium)	Experimental	Quass et al., 2004
6 European sites (urban background)	SIA: 40% $PM_{2.5}$ Sulphate: 14-31% $PM_{2.5}$ Ammonium: 7-10% $PM_{2.5}$	Experimental	Sillanpaa et al., 2006
Europe	SIA: 20-30% PM_{10} SIA: 30-40% $PM_{2.5}$	Modelling	WHO, 2006
UK (regional)	Ammonium: 14% PM_{10}	Experimental	Whyatt et al., 2007
60 European sites	Sulphate + nitrate: 17-20% PM_{10}	Experimental	Putaud et al., 2010
The Netherlands	SIA: 30-40% PM_{10} SIA: 25-35% PM_{10} (PM_{10}<40 $\mu g/m^3$) SIA: 45-55% PM_{10} (PM_{10}>40 $\mu g/m^3$)	Experimental	Weijers et al., 2010
Germany, 3 European locations	Sulphate: 2.7-3.3 $\mu g/m^3$ Nitrate: 1.2-3.3 $\mu g/m^3$ Ammonium: 1.3-1.6 $\mu g/m^3$	Modelling	Renner & Wolke, 2010
US, different regions	Ammonium: 5-11% of total $PM_{2.5}$	Modelling	Hristov, 2011
Europe	Ammonia: 20% SPM	Modelling	Moldanová et al., 2011
France (Paris)	SIA: 25% $PM_{2.5}$ SIA: 30% PM_{10}	Experimental	AirParif, 2012

The values reported in Table 1 for the contribution of NH_3 to PM mass 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 NH_4^+ have been detected (Querol et al., 2008), with higher ammonium levels along the eastern coast of Spain where the highest NH_3 agricultural emissions are recorded but where also the relative humidity is highest. The latter implies a higher atmospheric stability of NH_4NO_3. 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 PM_{10} and

$PM_{2.5}$ are generally organic matter, sulphate and nitrate, and that there is a clear decreasing gradient in SO_4^{2-} and NO_3^- contribution to PM_{10} when moving from rural to urban to kerbside sites. In contrast, the total carbon/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. A review the air quality situation in European cities (Querol et al., 2004) concluded that the contribution of SIA (from traffic, industrial emissions including power generation and agriculture) ranged from 3 to 9 $\mu g/m^3$ for PM_{10} and from 3 to 8 $\mu g/m^3$ for $PM_{2.5}$ at regional background sites. These concentrations were somewhat lower in Sweden (3-5 $\mu g/m^3$). SIA levels were very similar in urban areas. In intensively industrialised regions or heavily polluted urban areas additional SIA inputs (up to 5-6 $\mu g/m^3$) may be detected for PM_{10} and $PM_{2.5}$. NH_3 is transported from rural to urban areas and thus contributes to air pollution in cities (Putaud et al. 2004). NH_3 originating from the Netherlands can even contribute considerably to the PM mass in German urban areas (e.g. in the city of Münster; Vogt et al., 2005).

Uncertainties remain in the assessment of agricultural emissions and thus formation of SIA. In the past, airborne PM levels in regions with high NH_3 emissions were underestimated by air quality models, leading to discrepancies between modelled and measured PM concentrations (Wu et al., 2008). Measurements can also underestimate PM concentrations, e.g. due to losses caused by volatilisation (Keck and Wittmaack, 2005).

Table 2. Non-exhaustive compilation of studies quantifying the contribution from agricultural emissions to atmospheric pollutants (PM_{10}, $PM_{2.5}$, PAH, POP, NH_3, organic carbon (OC), etc.).

Agricultural activity	Type of site	Location	Contribution	Study
Mixed	Rural	Grönheim (Germany)	25-20% to total transported dust	Goossens et al., 2001
Biomass burning	Urban	Beijing (China)	10-32% to OC	Duan et al., 2004
Biomass burning	-	Arctic	Impact on TSP, not quantified	Stohl et al., 2007
Biomass burning	Rural	Valencia (Spain)	20-40% to PM_{10}	Viana et al., 2008
Mixed	Urban	Los Angeles (US)	2% to $PM_{2.5}$; 0.1% to O_3	Jacobson et al., 2008
Mixed	Urban	Kosetice (Czech Rep.)	Impact on POPs, not quantified	Dvorska et al., 2008
Biomass burning **Biomass burning**	Rural Urban	Jhu-Shan (Taiwan) Sin-Gang (Taiwan)	14% to PAH 19% to PAH	Lai et al., 2009
Fertiliser application & livestock	Urban	Toronto (Canada)	Impact on NH_3, not quantified	Godri et al., 2009
Biomass burning	Urban	Taichung city (Taiwan)	75% to $PM_{2.5}$; 30% to $PM_{2.5-10}$	Cheng et al., 2009
Biomass burning	Urban	Barcelona (Spain)	3-11% to $PM_{2.5}$	Reche et al., 2012a
Mixed	Urban	Paris (France	1% to $PM_{2.5}$	AirParif, 2012
Agriculture, forestry & land use change	Urban	Eastern Po Valley	5% to $PM_{2.5}$ 93% to NH_3	Pecorari et al., 2014

In addition to SIA production, agricultural emissions impact air quality through controlled combustion of agricultural residues. The burning of agricultural residues is a significant source of primary PM, especially $PM_{2.5}$ as it may contribute to up to 23% of primary $PM_{2.5}$ emissions from the agricultural sector (ETC, 2013). It is also a relevant source of secondary organic aerosols (SOA; de Gouw & Jiménez, 2009).

In summary, NH_3 plays a decisive role in PM formation chemistry by determining the amounts of ammonium sulphate ($(NH_4)_2SO_4$) and ammonium nitrate (NH_4NO_3). Both secondary inorganic aerosols are significant PM constituents. The role of NH_4NO_3 is exacerbated during high PM pollution episodes (typically during early spring) such as the March 2014 episode where its contribution to total PM load reached 50% in the fine fraction of PM (e.g. EEA, 2014).

The complexity of atmospheric chemistry leading to the formation of NH_4NO_3 challenges quantifying the benefit that can be expected when reducing NH_3 emissions, as documented in Bessagnet et al. (2014). This benefit can change in space and time, and annual average PM concentrations might respond differently than exceedances during high pollution episodes. Only air quality models allow to quantitatively assess such responses. A study presented by Bessagnet et al. (2014) focussed on annual values with NH_3 emission reductions affecting the whole agricultural sector (cattle breeding and fertilizer spreading), based on the Gothenburg Protocol target values. In the scenario analyses presented in the next chapter, the focus is on emissions during the early spring period and on the fertilizer application sub-sector.

2 Sensitivity of high particulate matter episodes to the reduction of ammonia emissions

The purpose of this section is to better illustrate the benefits that can be expected from various types of NH_3 mitigation measures: (i) the impact of annual NH_3 reductions on PM exceedances (section 5.2.1), (ii) the impact of annual NH_3 reduction in various parts of Europe on annual PM load (section 5.2.2), and (iii) the impact of short term NH_3 reduction in various parts of Europe of PM exceedances (section 5.3). While the first two topics could be addressed on the basis of existing modelling work, the last topic was addressed through a dedicated modelling experiment also presented in the text.

2.1 Sensitivity to long term (annual) NH_3 emissions

2.1.1 Sensitivity to exceedances of the daily PM_{10} limit value

Bessagnet et al. (2014) quantified the impact of reductions in NH_3 agriculture emissions in the Europe Union using different scenarios based on the Gothenburg Project for the year 2020, compared to 2009. Despite quite low contributions of ammonium in the PM concentrations (due to its low molecular weight), it has been demonstrated in several studies that cutting ammonia emissions could improve air quality.

Using the air quality model CHIMERE (in addition to others) they found that full implementation of the Gothenburg protocol would reduce the number of stations exceeding more than 35 times of the PM daily limit values of 50 µg m^{-3} from 612 in 2009 to 481 in 2020. They insisted that the efficiency of NH_3 emission reductions are non-linear: a further 30% reduction beyond the Gothenburg Protocol is approximately 3.2 more efficient than a 10% additional reduction although linearity would predict a factor 3.

2.1.2 Sensitivity of annual mean PM_{10} levels

In order to explore the benefit than can be expected from a given reduction of NH_3 emissions in various European countries to the annual mean PM_{10} load, we can rely on the Source Receptor Matrices (SRM) produced by the EMEP model (EMEP, 2015). By performing a large number of model sensitivity experiments, the response in terms of exposure to various air pollutants in countries of the EMEP region expected from an incremental reduction of precursors in the same or other countries of the region can be quantified. These matrices are in particular used to inform the GAINS model (Amann et al., 2011) and publicly available.
In the present report, we used the SRM produced in 2015 for the year 2013[2]. We are interested in particular to the response in country-wide annual mean $PM_{2.5}$ average concentration resulting from a 15% reduction in annual total NH_3 emission in each source country.

Box 4. Source Receptor Matrices (SRM) produced by the EMEP model

---

[2] http://www.emep.int/mscw/SR_data/Tables/2013_SRmatrices_EMEPStatus_1_2015AppC.tgz, accessed 25/05/2016

The SRM are organized so that each column shows where the pollutants emitted by a country ends up (Annex 1). For a 15% reduction in ammonia for the country indicated at the top of the column of an SRM, the numbers in each row (for each receptor country) is the modelled incremental reduction of $PM_{2.5}$ concentrations (positive for a decrease). Each row shows where pollutants in a given country or region come from. The numbers of a given row show which emitter countries are responsible for the change in pollutants in the country given at the beginning of each row.

Three main diagnostics can be derived from the above-mentioned SRM:

- The total reduction in $PM_{2.5}$ concentrations that can be expected for each country from a 15% reduction in NH_3 emissions throughout the domain, which can be assessed by summing up, for each row, all the contributions in each column,
- The total reduction in $PM_{2.5}$ concentrations that can be expected for each country from a 15% reduction in NH_3 emission in that country, which can be assessed from the diagonal of the matrix;
- The fraction of $PM_{2.5}$ reductions, for each country, that can be expected from domestic mitigation measures, therefore the ratio of the above two numbers.

Figure 1 presents various maps that can be derived from the EMEP SRM for ammonia contributing to $PM_{2.5}$. The top left panel shows, for each country, the amount of $PM_{2.5}$ concentrations ($\mu g/m^3$) that are reduced by a uniform 15% reduction of NH_3 emission over all source countries in Europe. It demonstrates that the largest absolute reductions of $PM_{2.5}$ are achieved over North-Central Europe (Poland, Czech Republic, Benelux, but also Netherlands, Germany, Luxemburg and Slovakia). The total potential for $PM_{2.5}$ reduction due to NH_3 mitigation appears limited in Italy, France, U.K. compared to evidence of ammonium nitrate contribution to PM episodes in those countries. This underestimation could be attributed to the country-wide spatial aggregation inherent to EMEP SRMs, therefore an approach by sub-regions of the corresponding countries is recommended in the dedicated modelling experiment presented in section 5.3.

The top right panel shows the absolute $PM_{2.5}$ reduction expected in each country from a 15% reduction of NH_3 reductions in the corresponding country. It shows that the countries where national mitigation is most efficient are large and/or isolated countries (Germany, Poland, Italy).

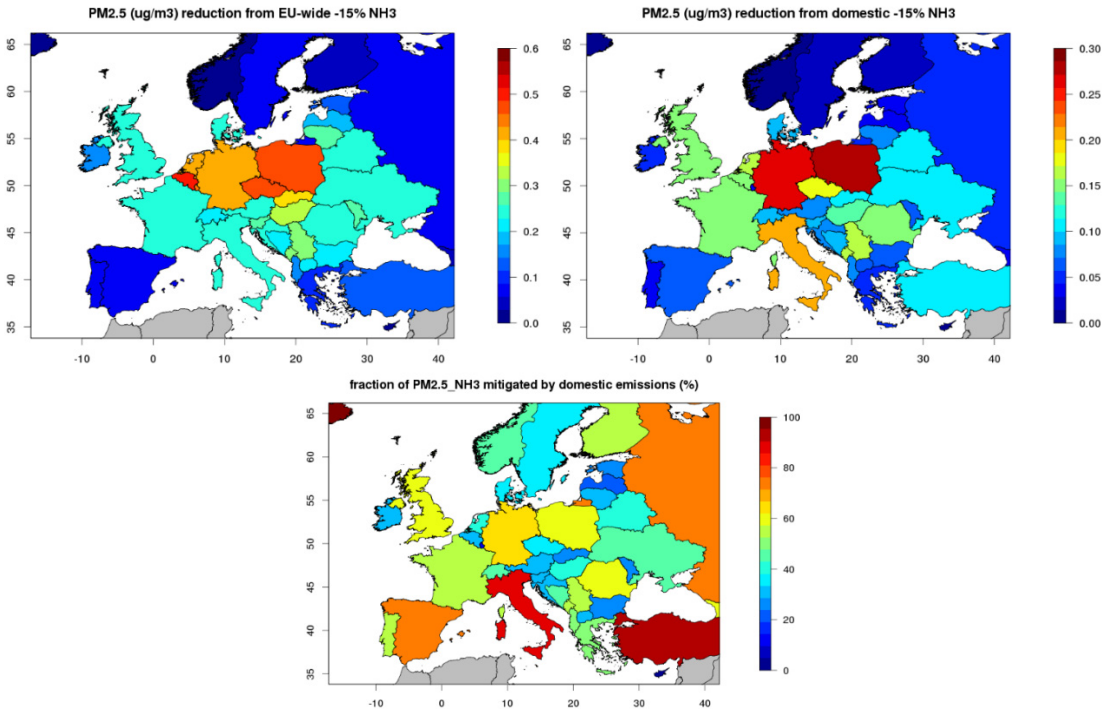


Figure 1 **Sensitivity of annual mean country-wide $PM_{2.5}$ concentrations to Europe-wide (top left) or domestic (top right) reductions of NH_3 by 15%. The lower right panel gives the fraction mitigated by domestic emissions.**

The bottom right panel shows the fraction of emissions (in %) mitigated by domestic emissions, (therefore the ratio of the previous two diagnostics). It must be discussed in comparison with the first panel to point out the countries more or less sensitive to mitigation of NH_3 emission in neighbouring countries:

- Some countries appear very sensitive to domestic emission reductions, but the overall potential to reduce $PM_{2.5}$ concentrations by reducing NH_3 emission is small (Turkey, Iceland, Spain, Finland);
- On the contrary, for some countries, action in neighbouring countries would be beneficial to reduce $PM_{2.5}$: Belgium, the Netherlands, and Luxemburg as well as the Czech Republic and Slovakia;
- There are also countries, where a substantial part of the benefit is expected from domestic mitigation (Germany, Poland, UK, France, Bulgaria, Serbia), although that fraction is at most 60%, and the remainder of the improvement lies in coordinated international NH_3 mitigation.

This analysis allows pointing out which countries in Europe are most sensitive to NH_3 mitigation when it comes to improving annual mean PM pollution. We will also take stock of these findings in designing the model experiment presented in section 2.2 regarding short-term actions where the selected source areas will be: Germany, Poland, Benelux, Central Europe (Czech Republic, Austria, Slovakia, Hungary), and fractions of the following countries: North Italy, North of France and Southern United Kingdom. The rationale for this choice is that (i) Germany and Poland appear here to be sensitive and efficient source and receptor areas, (ii) cluster of countries are legitimate for Benelux and Central Europe, (iii) the total response in Italy, France, and U.K. appears limited presumably because of the country-wide aggregation therefore supporting investigating sub-regions.

2.2 Sensitivity to short term actions during ammonia emission episodes

The purpose of the study is to explore the expected benefit of short term measures targeting specifically fertilizer spreading for a limited period of time, but with a high level of ambition with up to 80% reduction as was demonstrated to be an upper limit of achievable local reductions by (Sanz-Cobena et al., 2014). The results presented here are therefore complementary with the benefit expected from the long term reductions in the Gothenburg agreement in terms of PM exceedances , or the analysis of decrease in annual mean PM load resulting from an incremental reduction of NH_3 emissions according to the EMEP SRM presented .

In addition, we aim to explore the sensitivity of PM levels to NH_3 emission resulting from agricultural spreading of fertilizer exclusively, therefore excluding livestock emissions. The rationale for this choice is to explore the potential of mitigating for a short term time period, or even postponing, fertilizer spreading, whereas such an approach is not relevant for cattle.

2.2.1 Methodology

Selection of the time period

In the present study, simulations are conducted for the period 15/02/2011 – 16/04/2011. The year 2011 is selected because of the larger number of springtime PM_{10}-polluted days (within the interval 2010-2014). Figure 2 shows, for the main target countries, the temporal repartition of the daily exceedances in PM_{10} concentrations, as measured at European background stations. It shows that during the period 15/02 – 15/04, and for the years 2010 to 2014, almost half the exceedances were observed in 2011 in Germany and the Netherlands. More than one third of the exceedances were observed in 2011 in France, Belgium, the UK and Poland. Italy is the only country where 2011 does not present the highest number of exceedances. The March/April period is a high NH_3 emission period because of agricultural spreading of fertilizers and is thus a relevant period for the study of the PM_{10} concentration sensitivity to NH_3 emissions.

Model setup

To conduct the simulations, the chemistry-transport model CHIMERE has been used (Menut et al., 2013). Simulations have been conducted on a continental domain, covering Europe with a resolution of 0.5° (longitude) × 0.25° (latitude).

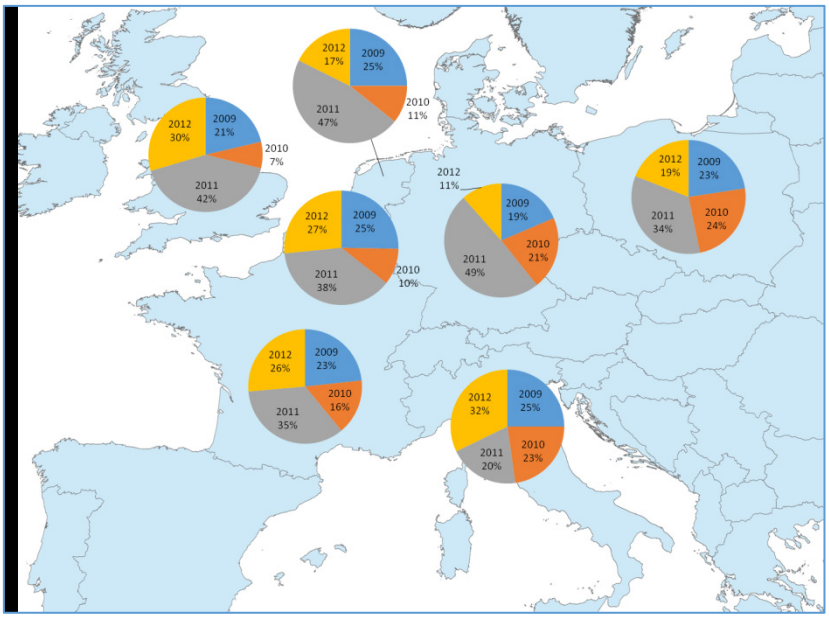


Figure 2 **Fraction (%) of the springtime (15/02-15/04) daily PM_{10} exceedences (> 50µg/m^3) observed for each year between 2009 and 2012. Results are based on daily mean observations provided by the European Air Quality Database (EEA, 2016c).**

Baseline emissions

In the processing of emissions, the agricultural sector (SNAP 10 of EMEP activity sectors) was therefore broken down in two sub-sectors: livestock and fertilizer applications. The split of the SNAP 10 sector into sub-sectors was done according to the officially reported emission data available on the EMEP website to provide the share of ammonia emissions due to fertilizer applications. The 2011 EMEP emission inventory was used. The emission inventory was refined by the use of proxies to redistribute the national emissions.

Different proxies were used to re-grid the emissions from the two agricultural sub-sectors. Livestock emissions were redistributed by using a high-resolution (around 1 km) global livestock database (including cattle, pigs, ducks, chickens, goats and sheep) providing the number of animals by grid cell[3]. The proxy of redistribution is computed as:

$$P_{Livestock} = \sum_{animal} EF_{animal} N_{animal}$$

With $P_{livestock}$ the proxy of redistribution, N_{animal} the number of animals and EF_{animal} a factor of NH_3 emissions by head. The emission factors (taking into account animal housing and storage) were estimated from data of the EEA emission inventory guidebook. The EMEP 0.1x0.1 resolution NFR emissions were not available in a final form when initiating these model simulations, but they will offer a promising alternative in the future. Note that officially gap filled national totals are used, and the methodology proposed here only regards the spatialisation proxy.

[3] Can be downloaded on http://livestock.geo-wiki.org/Application/index.php

Table 3. Emission factors used to compute the livestock redistribution proxy

	Ef_{animal} (kg-NH_3 per animal)
cattle	7.59
pigs	4.24
ducks	0.54
chickens	0.22
goats	0.27
sheep	0.24

The fertilizer proxy is based on a database of N-fertilizer use (resolution around 10 km) based on Mueller et al. (2012).

Figure 3 presents the computed annual NH_3 emissions over the CHIMERE grid for the Livestock and Fertilizer sectors.

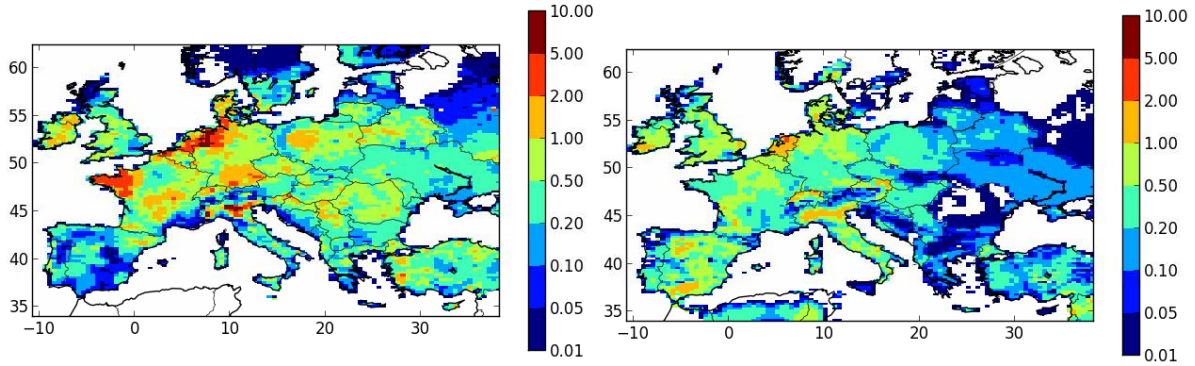


Figure 3 Annual NH_3 emissions (in Mg/km^2) over the CHIMERE domain for Livestock (left panel) and Fertilizers (right panel)

In air quality models, annual emissions are converted into hourly emissions by using temporal factors. Seasonal factors are used to convert annual emissions into monthly emissions and hourly factors are used to give the evolution of emissions during a day.

For fertilizer, most of NH_3 is emitted from February to May with around 57% of annual emissions during March and April (Hamaoui, 2012). A monthly emission factor of 28.5% was used for February to April in this study. May is not simulated in the present exercise.

Figure 4 shows the hourly factors used in this study to provide the hourly evolution of NH_3 emissions, from Chinkin et al. 2003. Whereas NH_3 emissions do not change significantly for the Livestock sector, emissions from the application of fertilizers are high during the day and low during the night due to the diurnal evolution of temperatures.

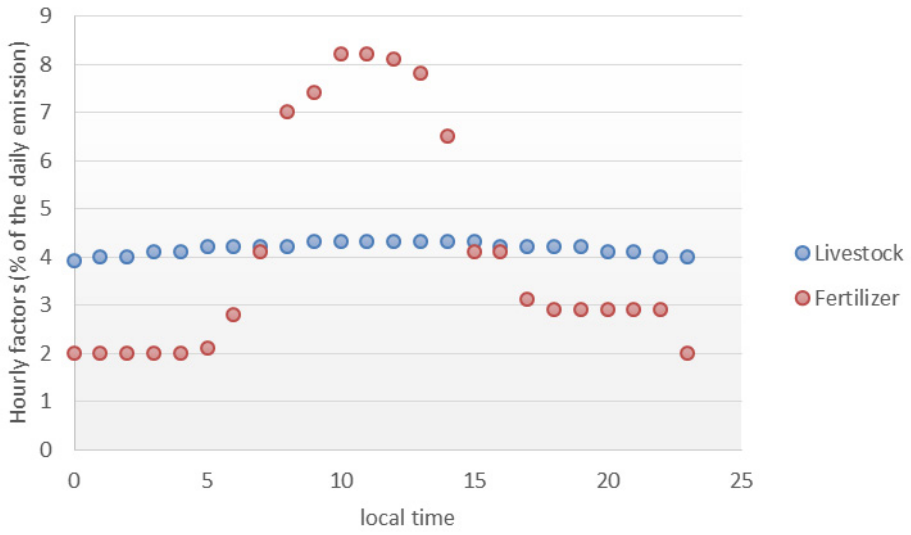


Figure 4 Hourly factors (% of the daily emission value) as a function of local time.

Scenarios

Seven regions over Europe were selected to test significant decreases in NH_3 emissions by fertilizers. Two of them concern whole countries (Poland and Germany), two concern groups of countries (Benelux: Belgium, Netherlands and Luxembourg; Central Europe: Czech Republic, Slovakia, Austria and Hungary) and three are parts of countries (Southern UK, Northern France and Northern Italy). These three "regions" are defined as following:

- Northern France is the part of metropolitan France northern than 47.9°
- England is chosen to be Southern UK
- Northern Italy includes the Emilia-Romagna region and all the northernmost regions.

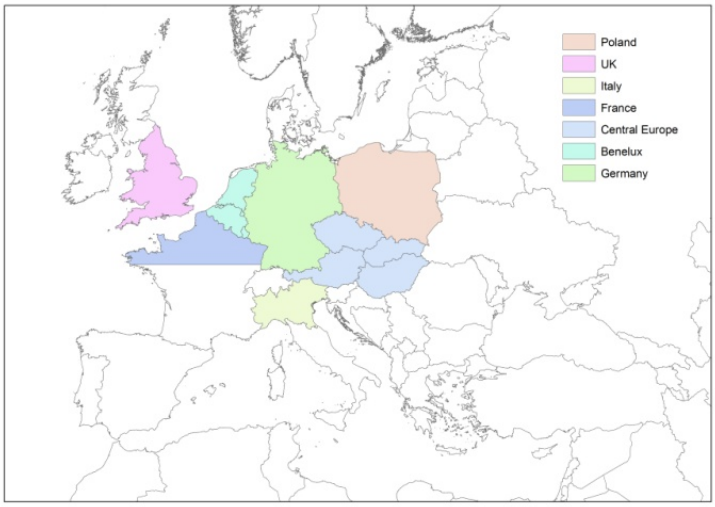


Figure 5 Seven European countries and regions where scenarios will be applied

For each region, we have conducted simulations considering a decrease of 80% and 50% in NH_3 emissions by fertilizers. 16 simulations have been conducted as listed below:

- 1 Reference simulation, without any decrease (referred to as REF).
- 1 Simulation considering an 80% decrease in NH_3 emissions from fertilizer for the seven regions at the same time (referred as TOT80).

- 7 simulations considering an 80% decrease in NH_3 emissions from fertilizer for each of the seven regions: France (referred as FR80), UK (UK80), Germany (GE80), Benelux (BNL80), Central Europe (CE80), Poland (PL80), Italy (IT80).
- 7 simulations considering a 50% decrease in NH_3 emissions from fertilizer for each of the seven regions (referred as FR50, UK50, GE50, BNL50, CE50, PL50, IT50).

Table 4 shows that except Germany, all the regions present very similar amounts of emitted NH_3: about 7 to 8 Gg for a 50% decrease in fertilizer emissions and about 12 Gg for a 80% decrease.

Table 4: Reduction in total NH_3 emissions in April 2011

	Reduction in NH_3 emission (Gg)		**Reduction of NH_3 emissions from the agricultural sector**	
	50% decrease in fertilizer emissions	**80% decrease in fertilizer emissions**	**50% decrease in fertilizer emissions**	**80% decrease in fertilizer emissions**
France	8.0	12.8	8%	13%
UK	7.9	12.7	19%	31%
Germany	14.7	23.5	19%	30%
Benelux	7.1	11.3	24%	38%
Central Europe	7.5	11.9	23%	36%
Poland	7.1	11.4	20%	32%
Italy	7.2	11.5	14%	23%

2.2.2 Results

PM_{10} concentrations

PM_{10} has been selected because Europe still faces important number of exceedance of the daily limit values. In the simulations, PM_{10} concentrations in Europe are between 10 and 30µg/m^3 in most of Europe on average for the period 15/02/2011 – 16/04/2011 (Figure 6). Particularly high concentrations are simulated over the Benelux and especially Northern Italy with mean concentrations reaching more than 60 µg/m^3. The PM concentrations are particularly high over the Chanel and the North Sea due to the shipping emissions and rather low boundary layer heights over the seas.

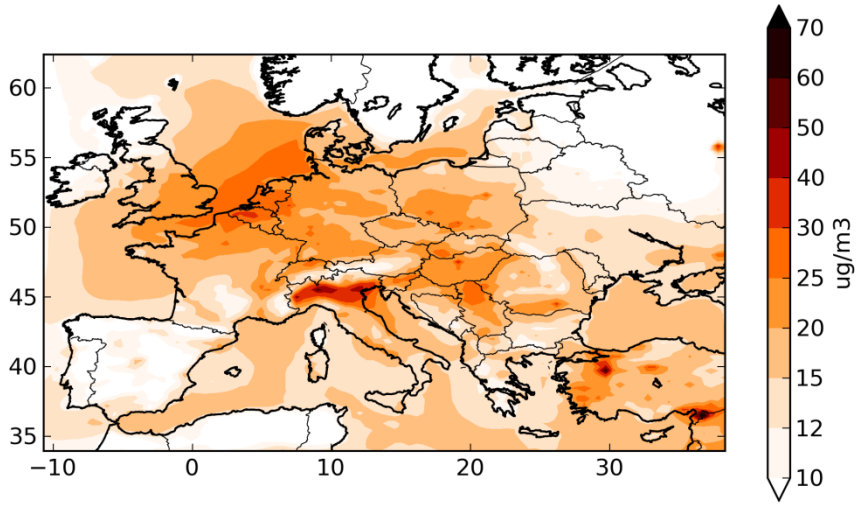


Figure 6 Average PM_{10} concentrations for the 15/02/2011 – 16/04/2011 period

Impact of the reduction scenarios

Figure 7 shows the decrease in mean PM_{10} concentrations over the period 15/02/2011 – 16/04/2011 considering an 80% fertilizer-induced NH_3 decrease in the seven regions at the same time (Scenario TOT80). Figure 7 shows the absolute impact (left) and the relative one (right), computed as follows:

$$\frac{Ref - TOT80}{Ref} \times 100$$

This scenario, which is by far the most restrictive, shows a decrease of up to 7 µg/m^3 above Northern Adriatic Sea and a relative decrease of up to 27% above the English Chanel. The English Chanel region is equidistant from England, Northern France and Benelux. This specific location may explain the high decrease in PM_{10} concentrations.

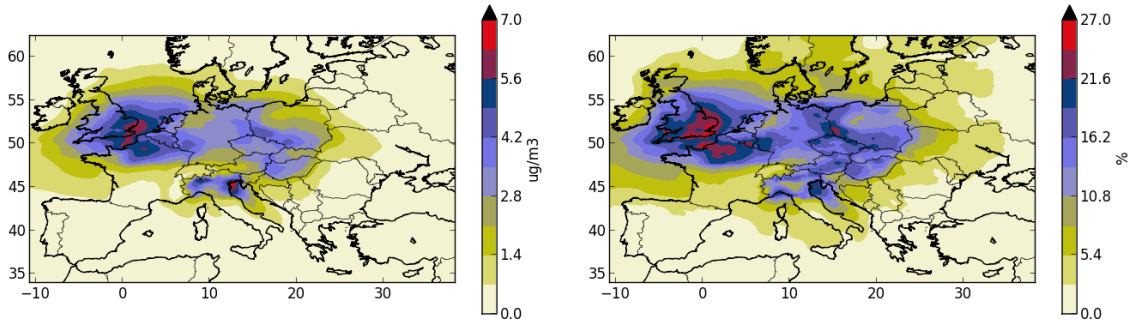


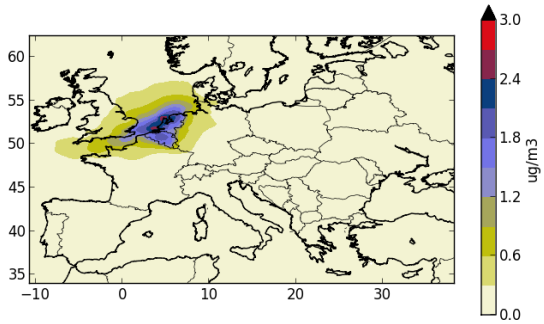
Figure 7 PM_{10} reduction (in µg/m^3 on the left and % on the right) due to the cumulated reduction of 80% in fertilizer-induced NH_3 emissions in the seven regions (TOT80 scenario)

Figure 8 shows the impact of the 14 individual scenarios on mean PM_{10} concentrations over Europe. With a decrease of up to 6 µg/m^3 for an 80% reduction in NH_3 emissions related to

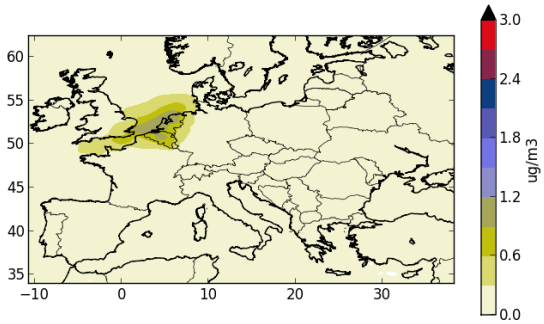
fertilizers, Italy is the country generating the most important impact. However, this impact is geographically limited since decreases in concentrations are only visible over Northern Italy and Northern Adriatic Sea.

As it will be shown later, the impact of an 80% decrease in fertilizer NH_3 emissions is more than twice important than the impact of a 50% decrease. This shows the non-linearity of the relation NH_3 emissions – PM_{10} concentrations. The most we reduce the emissions, the most efficient this decrease will be.

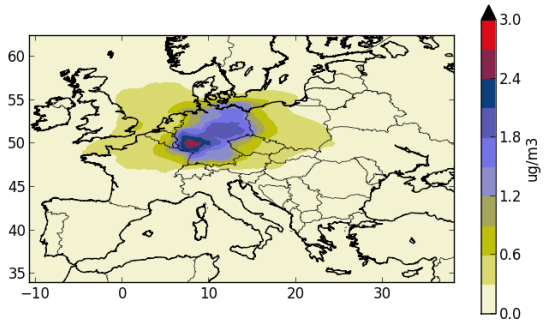
BNL80



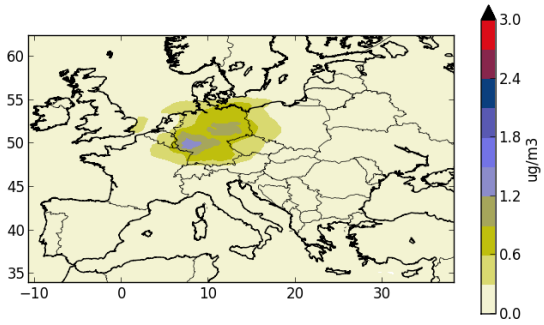
BNL50



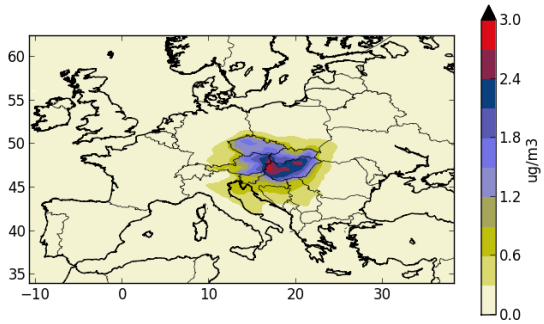
GE80



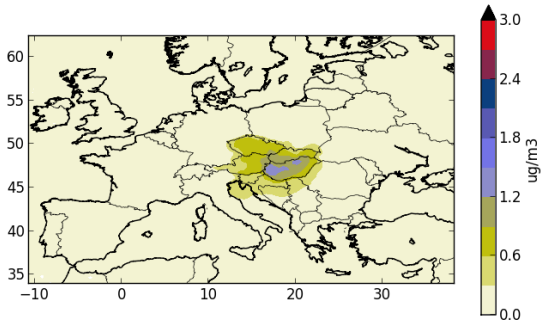
GE50



CE80



CE50



FR80

FR50

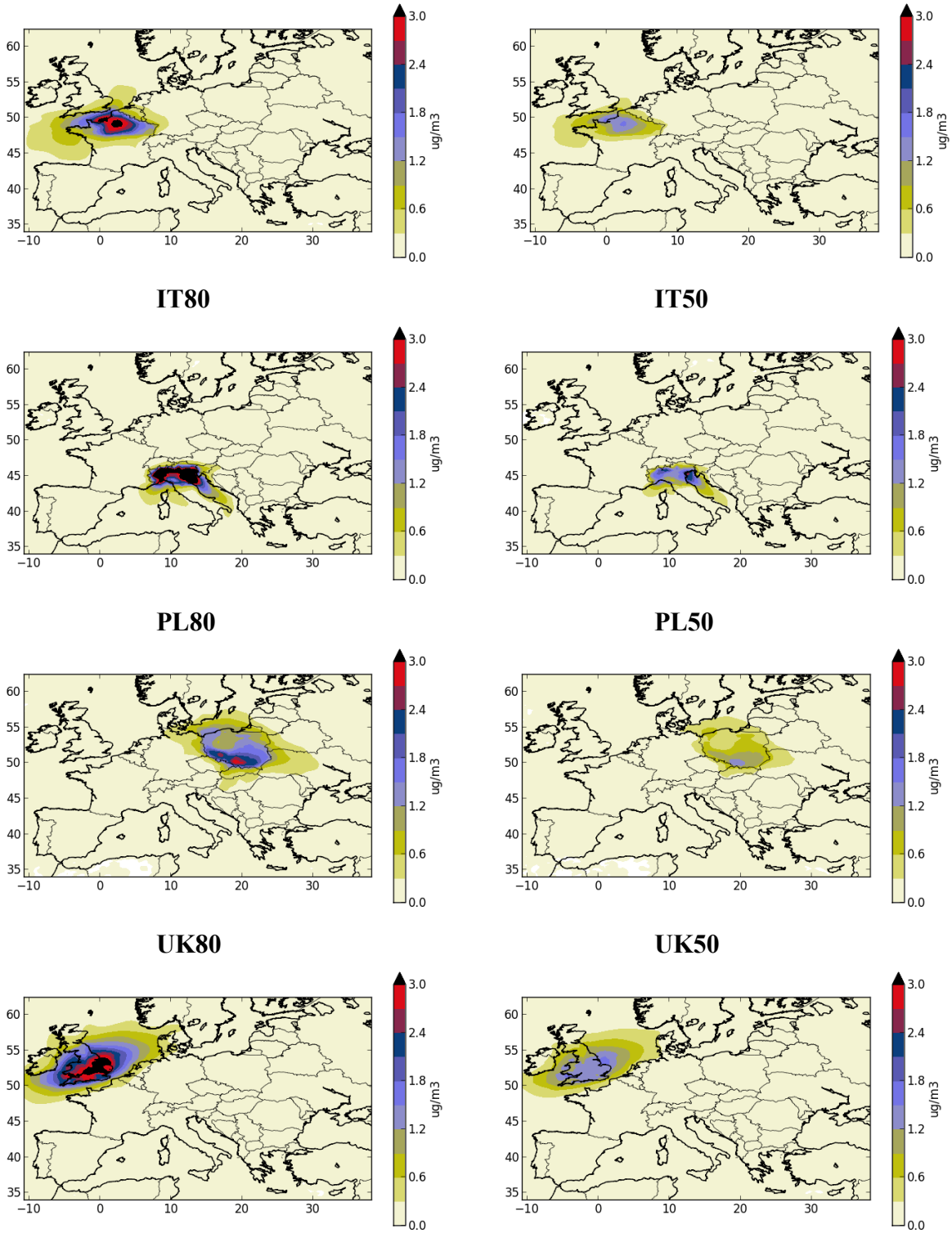


Figure 8 **PM10 reduction due to the reduction of 80% (left) and 50% (right) in NH3 emissions attributed to fertilizer spreading in the seven regions (separately)**

Figure 9 shows the difference, in terms of impact on PM_{10} concentrations, between running a simulation taking into account all the local reduction scenarios at the same time (TOT80) and adding the results of individual regional scenarios. The differences are due to the nonlinearities of the chemistry schemes (e.g. a chemical nonlinearity as shown on Figure 8

plus a "cross-country" interaction because a molecule of NH_3 emitted by a country can react with a molecule of HNO_3 produced in another country) and a European scenario takes advantage of these nonlinearities to enhance the efficiency of national emission reductions. The effects of nonlinearities are particularly important from the north of France to the south of England. In Italy, these nonlinearities are very low since the country is isolated from the other countries.

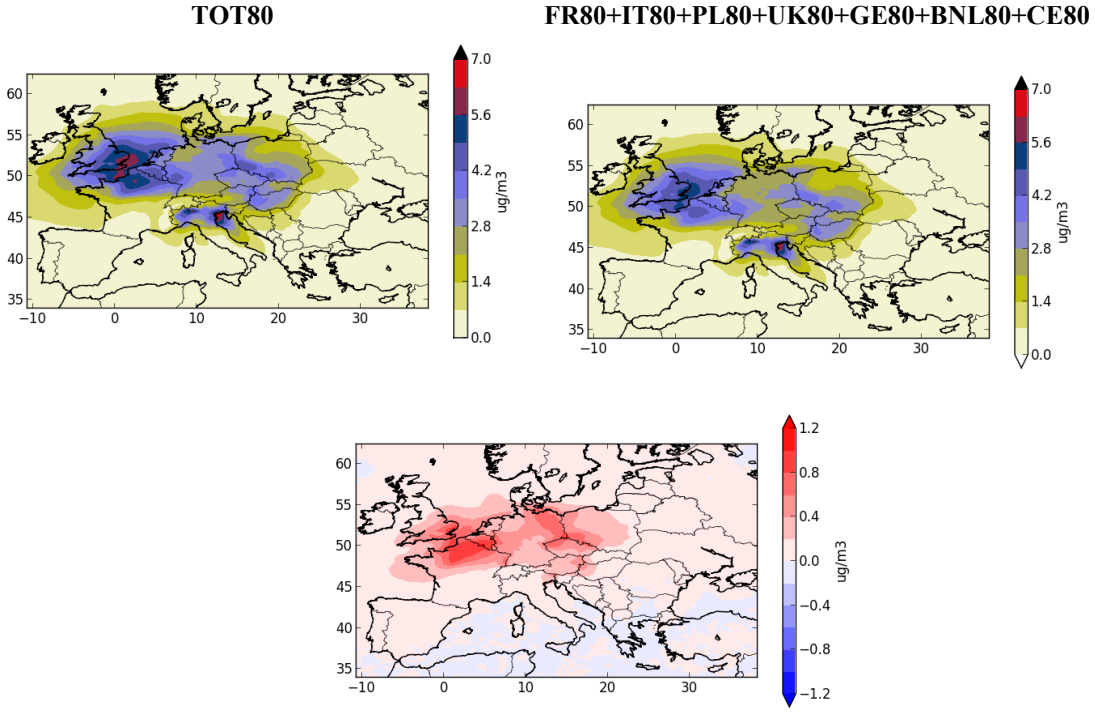


Figure 9 **Difference (bottom) between the simulation where NH_3 emission are reduced throughout the domain (top left) and the cumulated reduction where NH3 emissions are reduced in each country in the experiment (top right)**

Impact of the reduction scenarios on measurement stations

In European Air Quality Directive, PM_{10} daily mean concentrations shall not exceed 50µg m^{-3}. We use measurements provided by the AirBase European (EEA, 2015) measurement network. Between the 15/02/2011 and 16/04/2011, more than 14000 exceedances are observed among 1550 "background" stations. In order to evaluate the influence of NH_3 emission reductions on PM_{10} concentrations and exceedances, descriptive statistics are applied. We use concentrations data stored in AirBase database, and the model outputs from each scenario. All typologies of background stations were used: rural, suburban, and urban. The model outputs are interpolated to extract the modelled concentrations at the AirBase station locations. An increment of concentration at each monitoring site is then calculated, based on the difference of the modelling results from a scenario and the reference run. This increment is then applied to the 2011 observations to get a better estimate for each scenarios. This methodology based on observations and modelling allows assessing the evolution of concentrations for all stations typologies as follows:

- Step 1 : Observed concentrations C_k^{OBS} are collected for each station k
- Step 2 : Modelled concentrations C_k^{XXX} are calculated for each scenario XXX at each station k.
- Step 3 : Estimated concentrations E_k^{XXX} for each scenario XXX is estimated as :

$$\boldsymbol{E_k^{XXX} = C_k^{OBS} + (C_k^{XXX} - C_k^{REF})}$$

Figure 10 shows the impact of the scenarios on the number of exceedances observed in all background European stations. The most stringent scenario (TOT80) would induce a reduction of 3942 exceedances, while each 80% local scenario would induce a reduction of less than 500 exceedances individually. The sum of the seven regional scenarios would therefore induce a reduction of 2794 exceedances, illustrating again the importance of the non-linearity described in Figure 8. The UK scenarios have a low impact compared to scenarios in other countries. This is partly due to the peripheral position of the UK in Europe. Another reason could be the low number of measurement stations located in the UK. The measurement of the local impacts of the NH_3 emission decay in the different regions is influenced by the number of stations located in these regions, therefore the need for more elaborated statistics presented below.

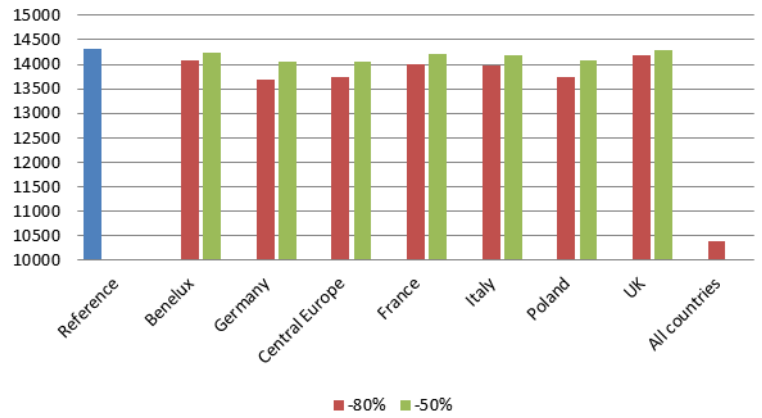


Figure 10 Number of PM_{10} daily threshold overtaking in all European background stations according to region where reduction scenarios are applied

Figure 11 (top) shows the local impact of the different local scenarios, e.g. we consider only the exceedances observed in the regions where the scenario is applied. As mentioned above, the very low number of exceedances observed for the UK is due to the weak number of measurement stations. On the other hand, Poland and countries from central Europe present a large number of stations. Figure 11 (bottom) presents the normalized decrease of the number of exceedances, by comparison with the number of exceedances in the reference scenario. The UK has a strong impact on itself with a decrease of 25% for UK50 scenario and more than 40% for UK80. Considering the 80% scenarios, France and Italy have also important impacts on themselves with a decrease of 20% in the number of the exceedances. On the other hand, Poland only decreases its local exceedances by 11%. Therefore, the high number of exceedances in Poland is not only due to the high number of stations. Those differences may be explained by the exceedance concentrations as we will see in Figure 12.

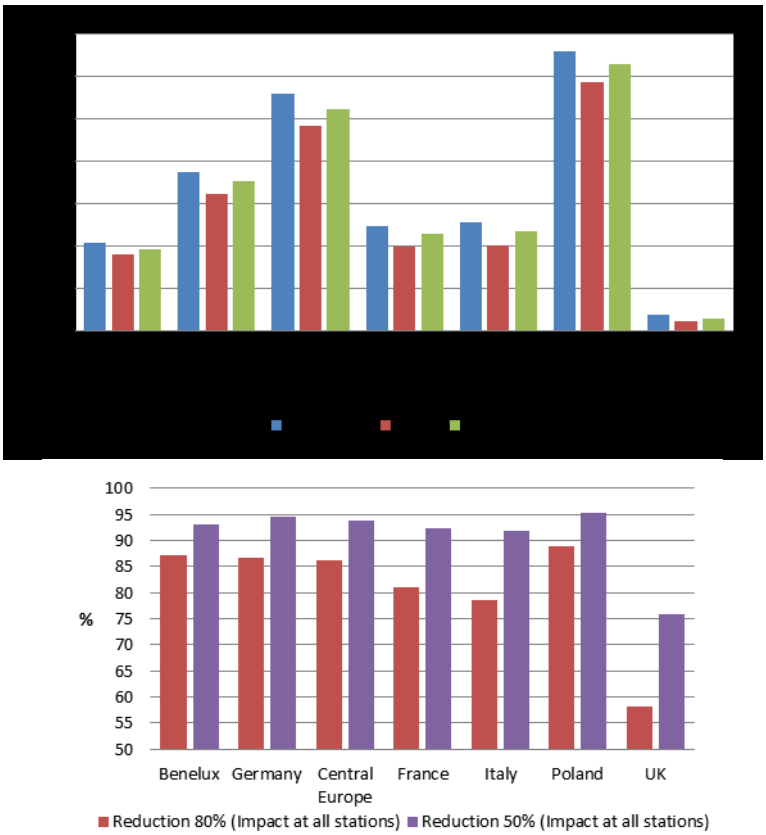


Figure 11 **PM_{10} daily thresholds exceedances at background stations in regions where the scenario is applied. Top: absolute number of stations. Bottom: Percentage relatively to the reference scenario**

Figure 12 shows the average PM_{10} concentrations for days and stations in exceedance (according to the reference scenario) of the daily limit value. Values for the Scenarios 50% and 80% are computed at the same places and times (including values that are not in exceedance anymore once the reduction considered). As seen earlier, Poland presents particularly high concentrations with concentrations almost reaching 90µg/m^3. Decreasing polish NH_3 emissions by fertilizer by 80% would induce a less than 10µg/m3 decrease in PM_{10} concentrations at very high polluted points. The UK, Italy and France have the lowest exceedance mean concentrations, close to the 50µg m^{-3} threshold. This is the reason why we observe in these countries a higher number of exceedance decay when we apply reduction scenarios.

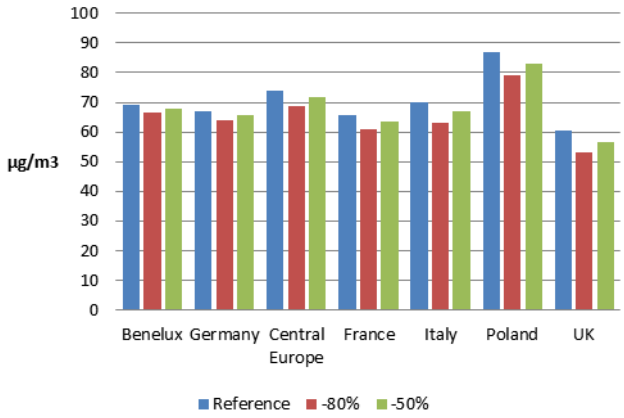


Figure 12 Evolution of PM_{10} concentrations at stations and for the days overtaking daily threshold in the reference scenario for each scenario

Figure 13 shows, for each region, the impact of the region on itself in term of number of exceedances ($\boldsymbol{N_{i80}}$), as well as the impact of the TOT80 scenario on the region ($\boldsymbol{N_{TOT80}}$). It will allow us to compute the ratio between the local impact and the European one on the different regions. This ratio is computed by comparing the reduction of the exceedances obtained with the local scenarios and the reduction of the exceedances obtained with the European one, as follows:

$$\boldsymbol{P_i}(\%) = \frac{\left(\boldsymbol{N_{Ref}} - \boldsymbol{N_{i80}}\right)}{\left(\boldsymbol{N_{Ref}} - \boldsymbol{N_{TOT80}}\right)}$$

Where P_i is the part of the local evolution of the exceedances of the region i. N_s is the number of exceedances simulated with the scenarios. Figure 13 quantifies, for a given country, the advantage of reducing local emissions rather than taking advantage of collective actions. For example, if Northern Italy lowers its NH_3 emissions alone, it will reduce its own exceedances by 22% (Figure 13, top left). A larger action (e.g. an action involving the seven regions) would make northern Italian exceedances decrease by 28% (Figure 13, top right). This means that an isolated Italian action would have 78% the efficiency of a more collective one on its territory (Figure 13, bottom). Its shows that Northern Italy is little affected by NH_3 emissions from other European countries and is able to control most of the PM_{10} decay due to NH_3 emissions. This is also visible on Figure 9c where it is shown that Italy is not affected by the cross-country interaction. On the other hand, France is highly impacted by the rest of Europe. Although a local action would reduce exceedances by almost 20% in France (Figure 13, top left), a European action would eradicate 60% of the exceedances (Figure 13, top right), making the local action only 33% as efficient as a European one for France. Those results are broadly in agreement with those shown in Figure 1.

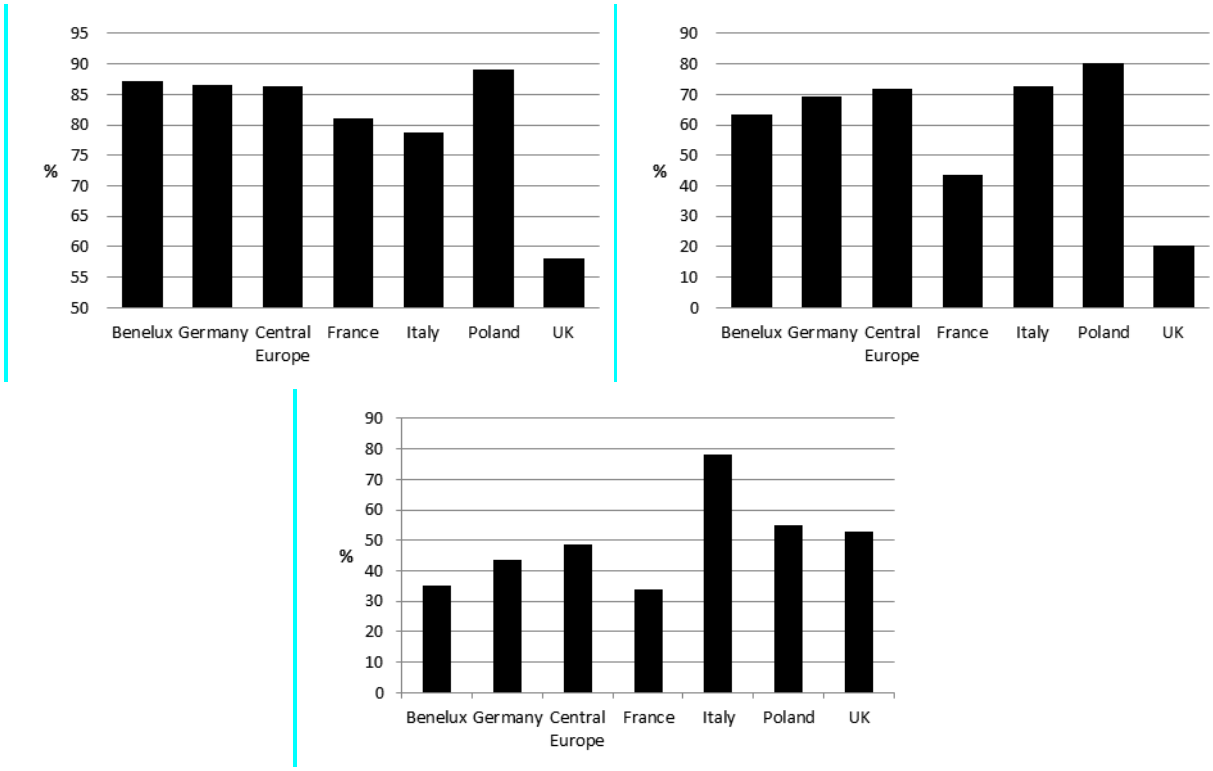


Figure 13 **(top left) Relative number of local exceedances simulated after applying local scenarios, *i.e.* impact of a region on itself (as shown in Figure 11), (top right) Relative number of local exceedances simulated by applying the TOT80 scenario, *i.e.* impact of Europe on each region, and (bottom) percentage of the decrease in exceedances due to the country where the reduction in emission is applied.**

2.3 Discussion and conclusions

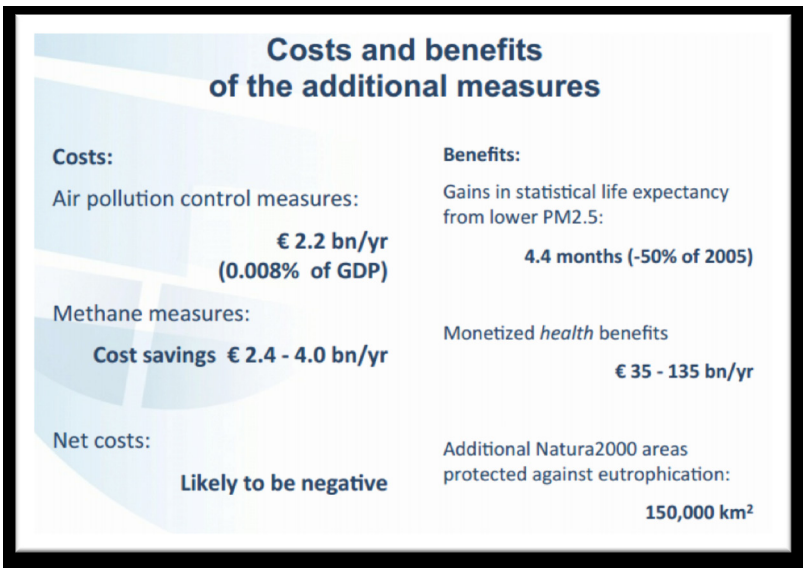
Costs and benefits of ammonia mitigation measures in agriculture

Amann et al. (2015) assessed that currently about 50% of the air pollution in Europe is related to unregulated sources. Further, model analyses suggest that in some cities even up to 50% of PM mass originates from international sources. Ammonium nitrate, resulting from the interaction of NOx emissions from traffic and industry with NH_3 from agriculture constitutes the largest part of the international and national contribution to PM. Whereas there is a theoretical potential of around 20-30% to further reduce the conventional pollutants NOx and SO_2, the potential for NH_3 reduction is much bigger (70-80%). Currently, 80% of ammonia emissions in Europe arise from farms with livestock larger than 500 units, representing only 10% of all farms. Amann et al. conclude that relatively simple and partly cost-neutral and cost-saving measures to reduce emissions are available, and they include:

- improving storage of manure and anaerobic digestion;
- modern application techniques of manure on soils, injecting instead of spraying; and
- avoiding urea fertilizer losses, or substitution by ammonium nitrate.

Monetary analyses show that for relatively small costs large gains in health benefits and protection of nature areas can be achieved (Figure 14).

Figure 14. Costs and benefits of mitigation measures. Source: Amann et al. (2015).



Ammonia abatement as key factor for abating secondary inorganic PM

As discussed by Brunekreef et al. (2015), the proposal for national emission ceilings requires reductions in emissions of SO_2, NOx and NH_3, but at different percentages: for 2020, relative to 2005, emissions of SO_2 across the EU need to be reduced by 59%, emissions of NOx by 42%, but emissions of NH_3 by only 6%. Larger reductions are proposed for 2025 and 2030, but the disparity between sulphur and nitrogen oxides on the one hand and ammonia on the other hand remain. This is hard to defend scientifically, because there is good evidence that all precursor gases need to be reduced in step to achieve the maximum reduction in fine particle concentrations, and that abatement of ammonia is a key factor for abating SIA. Ammonia reductions, which are technically possible, contribute more to reducing particle concentrations than do reductions of sulphur and nitrogen oxides (Megaritis et al., 2013). As described above, a reduction of the ammonia emissions from the agricultural sector related to animal husbandry could be especially efficient due to its seasonal emission pattern (Backes et al, 2016).

Nitrogen pollution was estimated to cost society in Europe between 70-320 billion/year, with about half of these costs related to agricultural emissions of NH_3 and run-off of reactive nitrogen in water (Sutton, 2015). An analysis of the ratio of cost and benefits of emission reductions shows a 3 times larger potential to cost-effectively reduce NH_3 emissions compared to NOx (mainly from transport and industry). Unfortunately, many of the voluntary measures outlined in the UNECE Framework Code for good agricultural practices to reduce ammonia emissions are not yet implemented in the signatory countries. Relatively simple technologies are available to reduce NH_3 emission from manure spreading. Also, increasing the nitrogen use efficiency by better integration of crop-livestock systems is proposed as a possible simple solution. Most EU countries, as well as South and East Asia, are identified as regions where an improvement of the Nitrogen Use efficiency could lead to N-savings between 100-250 kg/ha/yr, amounting globally to reducing 20 Tg N/year, and net benefits of 170 billion US$ per year (Sutton, 2015).

Brunekreef et al. (2015) discuss that, in view of the contribution of agriculture to fine particle concentrations in Europe, the health damage caused by particles from agriculture is estimated to be far greater than the burden placed on this sector by the current proposal for national emission ceilings. These authors consider this to be an inconvenient truth, which should be addressed by policy makers to propose and fund measures that do not threaten the livelihood of the farmer. As the EU starts to promote the circular economy (COM/2014/0398) (IPEX, 2015) there is a strong case to reduce ammonia emissions as part of innovation to increase economy-wide nitrogen use efficiency. European nitrogen pollution losses have a fertiliser value of about €20 billion per year based on the European nitrogen assessment (ENA, 2015) and a fertiliser price of about €0.80/kg nitrogen. This points to a major business opportunity to improve emission reduction and recycling technologies that further strengthen the case for revision of the national emission ceilings.

The largest PM pollution episode occurs in springtime

The complex atmospheric chemistry of ammonia requires using chemistry-transport models to assess the expected benefit of reducing NH_3 emissions on a national or European scale and for short or long term time periods. In this chapter, we reviewed existing work (i) on the benefit of long term reduction of Europe-wide NH_3 emissions in the context of the Gothenburg protocol , and (ii) on the benefit of incremental reduction of NH_3 emission in each country as modelled with the EMEP Source-Receptor Matrices . And (iii) we introduced a new modelling study aimed at quantifying the benefit of short term NH_3 emissions reductions due to fertilizer applications on PM_{10} concentrations during a high PM_{10} concentration period (March-April 2011) .

The analysis of annual NH_3 reduction beyond the target set in the revised Gothenburg Protocol highlighted the need to engage in ambitious reduction measures (Bessagnet et al., 2014). The relationship between reduction and benefit is not proportional, favouring substantial reductions. We are particularly interested in the potency of emission reductions: the improvement in air concentrations that can be expected from a unit reduction of emissions. An important result of the (Bessagnet et al., 2014) study is that the potency increases for ambitious reductions of NH_3 emissions.

The analysis of the main sources (of NH_3 emissions) and receptor (of $PM_{2.5}$ concentrations) according to the EMEP Source Receptor Matrices allowed identifying the countries where mitigation of NH_3 would yield important benefit for $PM_{2.5}$ exposure. Such matrices can also be used to point out the countries most exposed to transboundary effects (Benelux, Czech Republic), or to domestic emissions (Italy). For Germany, Poland, UK, France, Bulgaria and Serbia it was noted that only up to 60% of the response was attributed to domestic emission, the remainder of the improvement lied in coordinated international NH_3 mitigation.

Given that the largest PM pollution episode occurs in springtime, when fertilizer spreading is most common, it is legitimate to target such periods to attempt an optimisation of emission reduction measures. Therefore, we designed additional model experiments targeting fertilizer NH_3 emissions (and ignoring emissions of cattle that exhibit a weak seasonal cycle) for a 2-month period of time (15 February to 16 April 2014). We tested an 80% emission reduction of NH_3 attributable to the spreading of fertilizer during spring. This corresponds at the country level to a reduction from 23 to 38% for the whole agricultural sector. This reduction was tested over seven different European regions, and a reduction of 50% was also investigated.

Conclusions for the mitigation of ammonia emissions from agriculture

Considering the European scenario, for this 2-month period mentioned above, the maximum absolute reduction is reached over the Northern Adriatic Sea (7 $\mu g\ m^{-3}$) and the maximum $\mu g\ m^{-3}$ with a substantial impact on the number of exceedances of the PM_{10} daily limit values. With respect to mitigation, this **demonstrates the relevance of targeting the spreading of fertilizer over short periods, mainly in spring.**

The relationship between the benefit of lowering PM levels in the air and the effort in NH_3 mitigation is not proportional: **the more NH_3 emissions are reduced, the more relevant is the improvement obtained from an incremental reduction**. We found that decreasing NH_3 fertilizer emissions by 80% is more than twice as efficient in reducing PM_{10} concentrations as decreasing emissions by 50%, an assumption of linearity would have given a factor of 8/5.. This highlights a nonlinearity of the impact of NH_3 emission reductions on PM_{10} concentrations. This result, which focused on the most critical period for PM_{10} exceedances is coherent with the conclusion from Bessagnet et al. (2014), which focussed over an entire year.

Considering the impact of the NH_3 emission reductions on the PM_{10} daily limit value exceedances observed at European ground stations, we found that reducing NH_3 emissions by 80% causes a reduction of approximately 3942 exceedances throughout Europe (among a total of more than 14314). This reduction is only 2794 by adding results for each region tested individually. The difference can be attributed to the synergies of pan European measures due to the non linearity of the ammonium nitrate chemistry. **This result shows that European actions is not only the sum of national measures, there is an added synergy due to the specificity of secondary inorganic aerosol chemistry.**

The investigation of regional short-term scenarios confirmed the results of the analysis of EMEP source-receptor matrices: the local response to NH_3 emission reduction can be very different from a country to another. A large part of northern France and the Benelux is highly impacted by other countries. Consistently with the EMEP SRM analysis, we confirm that average PM concentrations in the north of Italy are mostly sensitive to local NH_3 emissions and less influenced by the rest of Europe. However, **by also including PM exceedances in the analysis, we found in Italy a noticeable transboundary effect that could not be pointed out in the analysis of source-receptor matrices**.

References

AirParif, 2012

Amann, M., Bertok, I., Borken-Kleefeld, J., Cofala, J., Heyes, C., Höglund-Isaksson, L., Klimont, Z., Nguyen, B., Posch, M., Rafaj, P., Sandler, R., Schöpp, W., Wagner, F., and Winiwarter, W. (2011) Cost-effective control of air quality and greenhouse gases in Europe: Modeling and policy applications, Environmental Modelling and Software, 26, 1489-1501.

Amann, M. (2015) Cost-effective measures to reduce premature mortality from air pollution in Europe with multiple benefits. In: EXPO2015: Sustainable Food Production and Air Pollution.

Arslan S. and Aybek, A., 2012. Particulate matter exposure in agriculture. In: Air Pollution – A comprehensive perspective. Edited by Intech.

Backes, A.M. , Armin Aulinger, Johannes Bieser, Volker Matthias, Markus Quante (2016) Ammonia emissions in Europe, part II: How ammonia emission abatement strategies affect secondary aerosols. Atmospheric Environment 126, 153-161.

Belis, C.A., Karagulian, F., Larsen, B.R., Hopke, P.K., 2013. Critical review and meta-analysis of ambient particulate matter source apportionment using receptor models in Europe. Atmos. Environ. 69, 94–108. doi:10.1016/j.atmosenv.2012.11.009Bessagnet, B., Beauchamp, M., Guerreiro, C., de Leeuw, F., Tsyro, S., Colette, A., Meleux, F., Rouïl, L., Ruyssenaars, P., Sauter, F., Velders, G. J. M., Foltescue, V. L., van Aardennee, J. (2014) Can further mitigation of ammonia emissions reduce exceedances of particulate matter air quality standards?, Environmental Science & Policy, Volume 44, Pages 149-163.

Brunekreef, B., Harrison, R.M., Künzli, N., Querol, X., Sutton, M.A., Heederik, D.J.J., Sigsgaard, T., 2015. Reducing the health effect of particles from agriculture. Lancet. Respir. Med. 3, 831–2. doi:10.1016/S2213-2600(15)00413-0

Chinkin, L.R., Ryan, P.A., Coe D.L., 2003. Recommended improvements to the CMU ammonia emission inventory model for use by LADCO. Prepared Lake Michigan Air Directors Consortium.

de Gouw, J.A., Welsh-Bon, D., Warneke, C., Kuster, W.C., Alexander, L., Baker, A.K., Beyersdorf, A.J., Blake, D.R., Canagaratna, M., Celada, A.T., Huey, L.G., Junkermann, W., Onasch, T.B., Salcido, A., Sjostedt, S.J., Sullivan, A.P., Tanner, D.J., Vargas, O., Weber, R.J., Worsnop, D.R., Yu, X.Y., Zaveri, R., 2009. Emission and chemistry of organic carbon in the gas and aerosol phase at a sub-urban site near Mexico City in March 2006 during the MILAGRO study. Atmos. Chem. Phys. 9, 3425–3442.

Duan, F., Liu, X., Yu, T., Cachier, H., 2004. Identification and estimate of biomass burning contribution to the urban aerosol organic carbon concentrations in Beijing. Atmos. Environ. 38, 1275–1282.Dvorská, A., Lammel, G., Holoubek, I., 2009. Recent trends of persistent organic pollutants in air in central Europe - Air monitoring in combination with air mass trajectory statistics as a tool to study the effectivity of regional chemical policy. Atmos. Environ. 43, 1280–1287. doi:10.1016/j.atmosenv.2008.11.028EEA (2014) Effects of air pollution on European ecosystems; EEA Technical report 11/2014.

EEA (2016a) European Union emission inventory report 1990–2014 under the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP), EEA Report No 16/2016.

EEA (2016b) Air Quality in Europe – 2016 report, EEA Report No 28/2016.

EEA (2016c) European Air Quality Database

EMEP (2015) Transboundary particulate matter, photo-oxidants, acidifying and eutrophying components, Oslo.

ENA (2015) European Nitrogen Assessment. www.nine-esf.org/ENA-Book (accessed Sept 30, 2015).
Erisman, J.., Schaap, M., 2004. The need for ammonia abatement with respect to secondary PM reductions in Europe. Environ. Pollut. 129, 159–163. doi:10.1016/j.envpol.2003.08.042
Erisman, J.W., Bleeker, A., Hensen, A., Vermeulen, A., 2008. Agricultural air quality in Europe and the future perspectives. Atmos. Environ. 42, 3209–3217. doi:10.1016/j.atmosenv.2007.04.004
European Parliament. 2014. Measures at farm level to reduce greenhouse gas emissions from EU agriculture. Published by: Directorate-General for internal policies, Agriculture and rural development. IP/B/AGRI/IC/2013_154.
Godri, K.J., Evans, G.J., Slowik, J., Knox, A., Abbatt, J., Brook, J., Dann, T., Dabek-Zlotorzynska, E., 2009. Evaluation and application of a semi-continuous chemical characterization system for water soluble inorganic PM2.5 and associated precursor gases. Atmos. Meas. Tech 2, 65–80.
Goossens, D., Gross, J., Spaan, W., 2001. Aeolian dust dynamics in agricultural land areas in Lower Saxony, Germany. Earth Surf. Process. Landforms 26, 701–720.
Harrison, R.M., Jones, A.M., Beddows, D.C.S., Derwent, R.G., 2013. The effect of varying primary emissions on the concentrations of inorganic aerosols predicted by the enhanced UK Photochemical Trajectory Model. Atmos. Environ. 69, 211–218. doi:10.1016/j.atmosenv.2012.12.016
Henderson, B., Falcucci, A., Mottet, A., Early, L., Werner, B., Steinfeld, H. & Gerber, P., 2015. Marginal costs of abating greenhouse gases in the global ruminant livestock sector. Mitigation and Adaption Strategies for Global Change. DOI: 10.1007/s11027-015-9673-9
Hristov, A.N., Hanigan, M., Cole, A., Todd, R., McAllister, T.A., Ndegwa, P.M., Rotz, A., 2011. Review: Ammonia emissions from dairy farms and beef feedlots. Can. J. Anim. Sci. 91, 1–35.
IPEX. Document COM/2014/0398. http://www.ipex.eu/IPEXL-WEB/dossier/document/COM20140398.do (accessed Sept 30, 2015).
Jacobson,M.Z. 2008. Short-term effects of agriculture on air pollution and climate in California. J. Geophys. Res. - Atmos. 113.
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. doi:10.1016/j.atmosenv.2005.03.029
Lai, C.-H., Chen, K.-S., Wang, H.-K., 2009. Influence of rice straw burning on the levels of polycyclic aromatic hydrocarbons in agricultural county of Taiwan. J. Environ. Sci. 21, 1200–1207. doi:10.1016/S1001-0742(08)62404-3
Lopez-Aparicio, S., Guerreiro, C., Viana, M., Reche, C. and Querol, X. 2013. Contribution of agriculture to air quality problems in cities and in rural areas in Europe. ETC/ACM Technical Paper 2013/10.
Moldanová, J., Grennfelt, P., Jonsson, Å., Simpson, D., Spranger, T., Aas, W., Munthe, J., Rabl, A., 2011. Chapter 18. Nitrogen as a threat to European air quality, in: Sutton, et Al. (Eds.), The European Nitrogen Assessment. Sources, Effects and Policy Perspectives. Cambridge University Press, Pp. 405-433.
Oenema O. and Velthof G.L., 2012. Emissions from agriculture and their control potentials. TSAP Report #3. IIASA, June 2012. (http://ec.europa.eu/environment/air/pdf/TSAP-AGRI-20121129_v21.pdf (assessed March 2016)

Pecorari, E., Squizzato, S., Longo, A., Visin, F., Rampazzo, G., 2014. Secondary inorganic aerosol evaluation: Application of a transport chemical model in the eastern part of the Po Valley. Atmos. Environ. 98, 202–213. doi:10.1016/j.atmosenv.2014.08.045

Putaud, J.P., Baltensperger, U., Brüggemann, E., Facchini, M., Fuzzi, S., Gehrig, R., Hansson, H.C., Harrison, R.M., Jones, A., Laj, P., Maenhaut, W., Mihalopoulos, N., Müller, K., Palmgren, F., Querol, X., Rodriguez, S., Spindler, G., Brink, H., Tunved, P., Dingenen, R., Wehner, B., Weingartner, E., Wiedensohler, A., Wåhlin, P., Raes, F., 2004. A European aerosol phenomenology II: physical and chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe. Atmos. Environ. 38, 2579–2595.

Putaud, J.P., Dingenen, R. Van, Alastuey, A., Bauer, H., Birmili, W., Cyrys, J., Flentje, H., Fuzzi, S., Gehrig, R., Hansson, H.C., Harrison, R.M., Herrmann, H., Hitzenberger, R., Hüglin, C., Jones, A.M., Kasper-Giebl, A., Kiss, G., Kousa, A., Kuhlbusch, T.A.J., Löschau, 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., Brink, H. ten, Tursic, J., Viana, M., Wiedensohler, A., Raes, F., 2010. A European aerosol phenomenology III: Physical and chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across Europe. Atmos. Environ. 1–13.

Quass, U., Kuhlbusch, T., Koch, M., 2004. Identification of Source Groups for Fine Dust (English summary) (Report). IUTA-Report LP15/2004.

Querol, X., Alastuey, A., Ruiz, C.R., Artíñano, B., Hansson, H.C., Harrison, R.M., Buringh, E., Brink, H.M. ten, 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., Artiñano, 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., 2012. Biomass burning contributions to urban aerosols in a coastal Mediterranean City. Sci. Total Environ. 175–190.

Renner, E., Wolke, R., 2010. Modelling the formation and atmospheric transport of secondary inorganic aerosols with special attention to regions with high ammonia emissions. Atmos. Environ. 44, 1904–1912. doi:10.1016/j.atmosenv.2010.02.018

Schaap, M., 2003. On the importance of aerosol nitrate over Europe : data analysis and modelling. Utrecht University.

Sillanpää, M., Hillamo, R., Saarikoski, S., Frey, A., Pennanen, A., Makkonen, U., Spolnik, Z., Van Grieken, R., Braniš, M., Brunekreef, B., Chalbot, M.-C., Kuhlbusch, T., Sunyer, J., Kerminen, V.-M., Kulmala, M., Salonen, R.O., 2006. Chemical composition and mass closure of particulate matter at six urban sites in Europe. Atmos. Environ. 40, 212–223. doi:10.1016/j.atmosenv.2006.01.063

Steinmann B., Gattinger A., Krauss M., Berner A., Leiber F., Maurer V., Meier M., Oehen B., Bautze L., Niggli U. (2015), Mitigating the impact of agriculture on air quality and climate change. IFOAM EU Group, Brussels, Belgium, www.ifoam-eu.org.

Stohl, A., Berg, T., Burkhart, J.F., Fjæraa, A.M., Forster, C., Herber, A., Hov, Ø., Lunder, C., McMillan, W.W., Oltmans, S., Shiobara, M., Simpson, D., Solberg, S., Stebel, K.,

Ström, J., Tørseth, K., Treffeisen, R., Virkkunen, K., Yttri, K.E., 2007. Arctic smoke – record high air pollution levels in the European Arctic due to agricultural fires in Eastern Europe in spring 2006. Atmos. Chem. Phys. 7, 511–534. doi:10.5194/acp-7-511-2007

Sutton, M., 2015. Controlling environmental nitrogen, how can it be done, how will it reduce impacts?, in: EXPO2015: Sustainable Food Production and Air Pollution.

Sutton, M.A., Howard, C.M., Erisman, J.W., 2011. The European nitrogen assessment: sources, effects and policy perspectives. Cambridge University Press.

UNECE (2012) 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.

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

Vogt, E., Held, A., Klemm, O., 2005. Sources and concentrations of gaseous and particulate reduced nitrogen in the city of Münster (Germany). Atmos. Environ. 39, 7393–7402. doi:10.1016/j.atmosenv.2005.09.012

Vonk, J., Bannink, A., Bruggen, C. van, Groenestein, C.M., Huijsmans, J.F.M., Kolk, J.W.H. van der, Luesink, H.H., Voshaar, S.V.O., Sluis, S.M. van der, Velthof, G.L., 2016. Methodology for estimating emissions from agriculture in the Netherlands. Wageningen.

Weijers, E.P., Kos, G.P.A., Blom, M.J., Otjes, R.P., Schaap, M., Swaluw, E. van der, 2012. Measurements of secondary inorganic aerosols in The Netherlands. Petten, The Netherlands.

WHO (2003)

WHO, Martuzzi, M., Mitis, F., Iavarone, I., Serinelli, M., 2006. Health impact of PM10 and ozone in 13 Italian cities.

ANNEX 1

Table 5: EMEP Source Receptor Matrix indicating the PM2.5 concentration change (positive for a decrease, in ng/m3) resulting from a 15% reduction in NH_3 emissions

	AL	AM	AT	AZ	BA	BE	BG	BY	CH	CY	CZ	DE	DK	EE	ES	FI	FR	GB	GE	GR	HR	HU	IE	IS	IT	KG	KZT	LT	LU	LV	MD	ME	MK	MT	NL	NO	PL	PT	RO	RS	RUE	SE	SI	SK	TJ	TM	TR	UA	UZ
AL	78.13	0.00	1.05	0.00	1.19	0.03	0.57	0.12	0.19	0.00	1.29	2.26	0.03	0.00	0.31	0.00	0.19	-0.03	0.01	9.98	1.35	2.98	0.00	0.00	3.13	0.00	-0.02	0.14	0.00	0.02	0.00	2.47	6.66	-0.01	0.02	0.01	-0.20	0.00	2.50	43.12	-0.32	0.03	0.15	0.26	0.00	0.00	1.59	0.18	0.00
AM	0.00	14.46	0.03	7.33	0.00	0.00	-0.01	-0.05	0.00	-0.01	0.01	0.05	-0.01	0.00	0.01	-0.01	0.02	-0.01	2.80	-0.01	0.00	-0.01	0.00	0.00	0.06	0.00	-0.33	-0.01	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	-0.07	0.00	-0.04	-0.05	-1.04	-0.01	0.00	0.00	0.00	-0.10	14.23	-0.23	-0.06
AT	0.05	0.00	81.73	0.00	0.62	0.88	0.01	0.91	3.75	0.00	23.50	63.40	0.58	0.01	0.21	0.02	3.64	0.55	0.00	0.01	4.78	9.90	0.13	0.00	29.01	0.00	0.00	0.27	0.10	0.05	0.06	0.01	0.01	0.00	1.21	0.03	12.08	0.00	1.47	2.10	0.10	0.21	9.74	2.90	0.00	0.00	0.13	0.38	0.00
AZ	0.00	1.92	0.01	48.99	-0.01	-0.01	-0.01	-0.07	0.00	0.00	-0.01	-0.05	-0.01	0.00	0.01	-0.01	-0.02	-0.02	4.44	-0.01	0.00	-0.02	0.00	0.00	0.06	0.02	0.05	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	0.00	-0.14	0.00	-0.09	-0.06	1.98	-0.01	0.00	-0.01	0.01	0.60	5.07	-0.13	0.03
BA	0.90	0.00	4.86	0.00	96.36	0.25	0.21	0.69	0.54	0.00	7.90	10.45	0.20	0.01	0.36	0.01	0.81	0.09	0.00	0.20	26.92	16.34	0.02	0.00	8.03	0.00	-0.02	0.38	0.02	0.04	0.12	1.96	0.16	0.00	0.31	0.04	5.55	0.00	4.00	32.07	-0.01	0.17	0.99	2.41	0.00	0.00	0.41	0.54	0.00
BE	0.00	0.00	0.78	0.00	0.00	151.19	-0.01	1.63	1.05	0.00	3.82	140.27	2.43	0.17	1.33	0.15	68.94	34.85	0.00	0.00	0.02	0.45	4.17	0.01	1.30	0.00	-0.01	0.98	4.78	0.24	0.00	0.00	0.00	0.00	76.07	0.15	11.92	0.04	-0.02	-0.05	0.28	0.75	0.05	0.30	0.00	0.00	-0.02	0.17	0.00
BG	0.64	0.02	1.69	0.04	0.89	0.16	58.46	1.21	0.37	0.02	2.31	5.89	0.08	0.01	0.28	0.02	0.74	0.11	0.13	10.80	2.90	6.93	0.03	0.00	1.45	0.00	-0.02	0.28	0.02	0.10	1.41	0.11	1.40	0.00	0.22	0.01	1.67	0.00	37.85	35.89	0.35	0.05	0.42	0.94	0.00	0.01	37.72	8.26	0.00
BY	0.04	0.01	1.01	0.05	0.21	0.61	0.60	106.96	0.30	0.01	3.35	15.25	1.32	0.46	0.16	0.58	1.95	1.06	0.11	0.05	0.78	4.80	0.20	0.00	1.17	0.01	0.33	6.10	0.04	1.71	1.18	0.01	0.02	0.00	1.51	0.09	45.29	0.00	6.45	1.78	16.72	0.78	0.38	1.73	0.00	0.05	6.35	20.39	0.05
CH	-0.01	0.00	2.68	0.00	-0.01	1.17	-0.03	-0.09	90.27	0.00	1.11	43.35	0.18	0.00	0.54	0.00	18.47	0.56	0.00	-0.01	0.04	0.07	0.09	0.00	37.04	0.00	-0.01	-0.02	0.21	0.00	-0.01	0.00	0.00	0.00	1.76	0.01	-0.23	0.01	-0.14	-0.11	-0.09	0.04	0.20	-0.02	0.00	0.00	-0.09	-0.10	0.00
CY	-0.29	0.00	-0.04	-0.01	-0.12	-0.04	-0.55	-0.36	-0.03	4.33	-0.11	-0.41	-0.04	-0.01	-0.72	-0.02	-0.52	-0.08	0.00	-2.43	-0.07	-0.24	-0.01	0.00	-1.78	0.00	-0.41	-0.06	0.00	-0.02	-0.14	-0.02	-0.14	-0.03	-0.04	-0.01	-0.80	-0.04	-1.36	-0.85	-3.01	-0.02	-0.03	-0.09	0.00	0.00	-29.13	-3.45	0.00
CZ	0.10	0.00	20.03	0.00	0.86	2.05	0.18	3.82	1.86	0.00	176.90	112.69	1.92	0.13	0.13	0.11	9.40	1.65	0.00	0.09	4.82	19.59	0.30	0.00	5.50	0.00	0.03	1.50	0.27	0.30	0.14	0.03	0.06	0.00	3.71	0.12	79.25	0.02	3.48	6.59	0.83	0.67	2.81	11.76	0.00	0.00	0.40	1.36	0.00
DE	0.01	0.00	5.88	0.00	0.05	12.18	-0.01	2.28	4.10	0.00	14.71	264.10	4.04	0.21	0.58	0.24	26.84	10.16	0.00	0.00	0.41	1.59	1.36	0.00	2.56	0.00	0.00	1.54	1.09	0.41	0.06	0.00	0.00	0.00	23.21	0.17	27.56	0.02	0.36	0.22	0.79	1.26	0.40	0.63	0.00	0.00	0.02	0.98	0.00
DK	0.00	0.00	0.28	0.00	0.00	3.51	0.01	2.29	0.27	0.00	1.64	64.03	89.94	0.23	0.86	0.62	10.42	18.87	0.00	0.00	0.04	0.29	2.32	0.00	0.17	0.00	0.00	1.39	0.10	0.54	0.08	0.00	0.00	0.00	15.26	1.08	12.79	0.02	0.29	0.01	0.85	10.56	0.03	0.19	0.00	0.00	0.02	1.35	0.00
EE	0.04	0.00	0.26	0.02	0.08	0.69	0.22	7.92	0.12	0.00	1.15	14.40	3.19	28.93	0.14	4.38	1.59	2.46	0.01	0.05	0.20	1.20	0.38	0.00	0.31	0.00	0.13	8.09	0.02	8.67	0.11	0.01	0.04	0.00	2.66	0.36	12.14	0.01	1.85	0.82	9.16	4.46	0.08	0.46	0.00	0.02	1.04	2.19	0.02
ES	0.00	0.00	0.05	0.00	-0.01	0.43	0.00	0.01	0.23	0.00	0.03	1.56	0.04	0.00	56.55	0.00	10.46	0.83	0.00	0.00	0.00	-0.01	0.26	0.00	0.71	0.00	0.00	0.01	0.03	0.00	0.00	0.00	0.00	0.00	0.67	0.00	0.05	1.47	-0.01	-0.02	-0.01	0.01	0.02	-0.01	0.00	0.00	0.00	0.00	0.00
FI	0.01	0.00	0.04	0.00	0.01	0.12	0.01	0.87	0.02	0.00	0.14	1.76	0.31	0.96	0.04	17.05	0.34	0.42	0.00	0.00	0.03	0.14	0.07	0.00	0.08	0.00	-0.03	0.76	0.00	0.47	0.01	0.00	0.00	0.00	0.41	0.31	2.48	0.00	0.11	0.12	1.54	2.54	0.02	0.05	0.00	0.00	0.06	0.23	0.00
FR	0.00	0.00	0.97	0.00	0.00	12.47	-0.01	0.31	4.19	0.00	1.84	44.90	0.61	0.03	4.69	0.02	142.94	13.51	0.00	0.00	0.07	0.21	1.42	0.00	5.44	0.00	0.00	0.16	1.05	0.04	0.00	0.00	0.00	0.00	11.30	0.05	2.35	0.05	0.00	-0.04	0.03	0.14	0.14	0.11	0.00	0.00	0.01	0.01	0.00
GB	0.00	0.00	0.28	0.00	0.00	9.33	0.00	0.85	0.34	0.00	1.25	29.57	3.45	0.10	1.25	0.13	18.72	157.35	0.00	0.00	0.02	0.22	5.81	0.01	0.78	0.00	0.00	0.66	0.29	0.18	0.01	0.00	0.00	0.00	16.33	0.45	4.32	0.10	0.03	-0.01	0.29	1.11	0.04	0.11	0.00	0.00	-0.01	0.28	0.00
GE	0.01	1.13	0.05	6.34	0.00	0.00	0.00	-0.07	0.01	0.00	0.00	0.02	-0.01	-0.01	0.02	-0.01	0.01	-0.02	23.73	0.02	0.00	-0.01	0.00	0.00	0.14	0.00	-0.19	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.10	0.00	-0.05	-0.02	0.98	-0.01	0.01	0.00	0.00	-0.05	8.52	-0.12	-0.04
GL	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
GR	3.55	0.01	0.58	0.02	0.14	-0.01	7.68	0.24	0.13	0.01	0.56	1.24	0.01	0.00	0.01	0.00	-0.11	-0.04	0.07	51.11	0.62	1.65	0.00	0.00	-1.10	0.00	-0.05	0.12	0.00	0.03	0.15	0.09	2.89	-0.05	0.00	0.00	-0.09	-0.03	4.39	11.12	-0.28	0.02	0.08	0.18	0.00	0.00	16.20	2.05	0.00
HR	0.48	0.00	10.46	0.00	21.29	0.28	0.28	0.94	0.86	0.00	12.80	16.35	0.37	0.02	0.34	0.03	1.18	0.09	0.00	0.20	72.35	24.35	0.02	0.00	33.38	0.00	-0.01	0.43	0.02	0.06	0.19	0.29	0.11	0.00	0.45	0.05	10.30	0.00	5.63	20.21	0.05	0.23	9.85	2.96	0.00	0.00	0.54	0.67	0.00
HU	0.34	0.00	16.56	0.01	2.80	0.50	1.39	2.82	0.80	0.01	21.07	25.53	0.64	0.06	0.18	0.09	2.49	0.32	0.01	0.47	15.56	128.76	0.06	0.00	9.82	0.00	0.01	1.05	0.06	0.19	0.37	0.09	0.29	0.00	0.91	0.07	27.48	-0.01	23.89	28.25	0.33	0.44	4.58	17.29	0.00	0.01	2.64	2.41	0.01
IE	0.00	0.00	0.03	0.00	0.00	3.45	0.00	0.81	0.28	0.00	0.20	12.83	1.63	0.09	1.22	0.10	14.66	58.08	0.00	0.00	0.00	-0.01	46.48	0.01	0.19	0.00	0.00	0.46	0.14	0.13	0.00	0.00	0.00	0.00	6.41	0.22	1.48	0.09	-0.01	-0.02	0.23	0.58	0.00	-0.01	0.00	0.00	-0.01	0.24	0.00
IS	0.00	0.00	0.00	0.00	0.00	0.26	0.00	0.00	0.01	0.00	0.00	1.07	-0.06	0.00	0.05	0.00	0.87	1.62	0.00	0.00	0.00	0.00	0.85	-3.43	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.50	-0.03	-0.01	0.00	0.00	0.00	-0.01	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00
IT	0.13	0.00	3.30	0.00	0.42	-0.02	0.02	0.03	2.62	0.00	1.26	4.07	0.05	0.00	1.54	0.00	3.22	-0.06	0.00	0.06	1.78	1.37	0.01	0.00	215.67	0.00	-0.01	0.02	0.00	0.00	0.02	0.03	0.01	0.02	0.03	0.00	0.02	0.02	0.23	0.65	-0.06	0.01	3.14	0.08	0.00	0.00	0.06	0.02	0.00
KG	0.00	0.01	0.00	0.02	0.00	0.00	0.00	-0.01	0.00	0.00	0.00	-0.01	0.00	0.00	0.00	0.00	-0.01	-0.01	0.02	0.00	0.00	0.00	0.00	0.00	0.00	35.74	6.91	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	0.00	-0.01	0.00	-0.38	0.00	0.00	0.00	0.00	5.45	0.12	0.18	-0.04	9.71
KZT	0.00	0.03	0.01	0.23	0.00	0.00	0.01	0.29	0.00	0.00	0.01	0.03	0.00	0.00	0.00	0.02	-0.01	-0.01	0.08	0.00	0.00	0.03	0.00	0.00	0.01	1.46	27.70	0.03	0.00	0.01	0.02	0.00	0.00	0.00	0.00	0.00	0.04	0.00	0.08	0.00	11.14	0.01	0.00	0.01	0.46	1.54	0.37	1.43	3.04
LT	0.07	0.00	0.95	0.01	0.15	1.10	0.36	31.12	0.42	0.00	4.42	28.85	4.39	1.13	0.17	1.01	3.30	3.27	0.01	0.07	0.54	4.69	0.49	0.00	0.62	0.00	0.12	74.88	0.05	7.54	0.14	0.02	0.06	0.00	3.24	0.20	67.12	0.01	4.44	1.85	10.72	2.52	0.25	1.75	0.00	0.02	1.35	4.68	0.02
LU	0.00	0.00	1.32	0.00	0.00	58.17	-0.02	1.38	1.22	0.00	7.05	195.47	1.14	0.09	0.89	0.07	76.27	12.93	0.00	0.00	0.05	0.61	1.81	0.00	1.37	0.00	-0.01	0.64	34.13	0.15	0.01	0.00	0.00	0.00	29.31	0.11	11.18	0.02	0.04	-0.05	0.19	0.30	0.08	0.42	0.00	0.00	0.01	0.08	0.00
LV	0.05	0.00	0.49	0.02	0.11	0.85	0.34	20.24	0.25	0.00	2.57	22.10	4.23	4.07	0.14	2.04	2.56	2.83	0.01	0.06	0.33	2.66	0.37	0.00	0.42	0.00	0.16	28.53	0.05	33.75	0.23	0.01	0.04	0.00	2.75	0.27	30.86	0.01	3.28	1.25	11.74	3.46	0.14	0.98	0.00	0.02	1.77	3.97	0.02
MD	0.11	0.03	1.01	0.13	0.34	0.22	4.70	5.93	0.32	0.01	3.40	7.83	0.69	0.18	0.24	0.17	1.19	0.24	0.27	0.54	1.02	5.77	0.08	0.00	1.50	0.00	0.20	0.62	0.03	0.24	63.60	0.04	0.04	0.00	0.38	0.08	14.47	0.00	75.53	4.76	6.94	0.37	0.26	1.10	0.00	0.03	22.94	46.76	0.02
ME	12.76	0.00	1.96	0.00	12.04	0.10	0.48	0.23	0.29	0.00	2.62	4.42	0.66	0.00	0.58	0.00	0.47	0.03	0.00	0.87	3.93	6.69	0.02	0.00	4.65	0.00	0.00	0.31	0.01	0.05	0.06	37.46	0.54	0.00	0.10	0.01	1.42	0.01	2.91	59.17	-0.05	0.07	0.22	0.84	0.00	0.00	1.05	0.35	0.00
MK	12.79	0.00	1.61	0.00	1.23	0.05	2.13	0.24	0.29	0.00	2.44	4.16	0.04	0.00	0.39	0.01	0.41	0.00	0.01	35.61	2.42	4.37	0.01	0.00	1.71	0.00	-0.04	0.24	0.01	0.06	0.04	0.33	65.32	0.00	0.09	0.01	0.65	0.00	4.20	66.66	-0.33	0.04	0.19	0.75	0.00	0.00	5.21	0.53	0.00
MT	0.02	0.00	0.61	0.00	0.05	0.00	0.04	-0.05	0.18	0.01	0.13	0.70	0.00	0.00	4.18	0.00	2.53	0.03	0.00	0.51	0.31	0.17	0.01	0.00	4.77	0.00	0.00	-0.01	0.01	0.00	0.01	0.02	-0.02	53.65	0.01	0.00	-0.48	0.24	0.11	0.83	-0.06	0.00	0.16	-0.13	0.00	0.00	1.12	0.12	0.00
NL	0.00	0.00	0.45	0.00	0.02	34.63	-0.01	1.44	0.47	0.00	1.76	109.75	5.94	0.23	1.36	0.26	34.25	51.62	0.00	0.00	0.07	0.16	5.29	0.01	0.70	0.00	-0.01	1.43	0.59	0.44	0.00	0.00	0.00	0.00	167.68	0.25	10.69	0.06	-0.05	0.02	0.38	1.57	0.06	0.09	0.00	0.00	-0.04	0.51	0.00
NO	0.00	0.00	0.00	0.00	0.00	0.06	0.00	0.05	0.00	0.00	0.02	1.34	1.44	0.00	0.07	0.21	0.38	1.52	0.00	0.00	0.00	0.00	0.16	-0.01	0.00	0.00	-0.02	0.07	0.00	0.01	0.00	0.00	0.00	0.00	0.26	6.20	0.69	0.01	0.00	0.00	-0.36	1.05	0.00	0.00	0.00	0.00	-0.01	0.00	0.00
PL	0.14	0.00	3.95	0.02	0.60	2.27	0.37	13.22	0.96	0.00	24.25	70.93	4.76	0.34	0.25	0.39	5.55	3.05	0.01	0.13	2.63	14.63	0.52	0.00	3.11	0.00	0.11	5.93	0.13	1.10	0.49	0.04	0.09	0.00	4.75	0.21	284.75	0.00	7.35	5.67	3.76	1.91	1.20	7.03	0.00	0.03	0.79	7.15	0.02
PT	0.00	0.00	0.03	0.00	0.00	0.23	0.00	0.00	0.09	0.00	0.01	0.81	0.05	0.00	21.16	0.00	4.33	0.43	0.00	0.00	0.00	0.00	0.23	0.00	0.34	0.00	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.00	0.36	0.01	-0.01	36.00	-0.01	-0.01	-0.01	0.02	0.01	0.00	0.00	0.00	0.00	0.00	0.00
RO	0.34	0.01	2.16	0.02	0.65	0.17	7.81	2.41	0.39	0.01	3.60	7.32	0.28	0.07	0.27	0.08	0.88	0.16	0.06	1.06	2.39	16.09	0.04	0.00	2.43	0.00	0.01	0.51	0.02	0.16	3.28	0.08	0.20	0.00	0.28	0.04	6.36	0.00	145.08	17.55	0.80	0.21	0.48	1.86	0.00	0.01	14.74	10.63	0.01
RS	3.08	0.00	3.97	0.01	6.43	0.32	3.25	1.02	0.54	0.01	6.40	10.89	0.19	0.01	0.27	0.02	1.04	0.13	0.02	3.28	11.87	21.59	0.04	0.00	2.81	0.00	-0.04	0.46	0.03	0.09	0.49	1.10	3.92	0.00	0.39	0.03	4.85	0.00	28.40	169.25	-0.23	0.15	0.83	2.38	0.00	0.00	5.42	1.38	0.00
RUE	0.01	0.02	0.04	0.16	0.01	0.04	0.05	1.98	0.02	0.00	0.12	0.62	0.07	0.14	0.01	0.26	0.10	0.06	0.14	0.01	0.03	0.15	0.01	0.00	0.07	0.01	4.12	0.32	0.00	0.16	0.08	0.00	0.00	0.00	0.08	0.02	0.93	0.00	0.45	0.08	47.84	0.09	0.01	0.05	0.01	0.13	1.00	3.83	0.10
SE	0.00	0.00	0.06	0.00	0.00	0.51	0.00	0.92	0.08	0.00	0.30	3.85	6.39	0.26	0.16	1.46	1.68	2.31	0.00	0.00	0.01	0.04	0.31	0.00	0.08	0.00	-0.01	0.64	0.02	0.30	0.01	0.00	0.00	0.00	2.04	1.49	3.11	0.01	0.05	0.00	0.28	16.07	0.01	0.02	0.00	0.00	0.01	0.36	0.00
SI	0.08	0.00	27.07	0.00	1.64	0.24	0.01	0.66	1.24	0.00	9.20	18.77	0.38	0.00	0.32	0.01	1.41	0.04	0.00	0.03	22.70	11.28	0.03	0.00	84.23	0.00	-0.01	0.11	0.03	0.02	0.12	0.01	0.01	0.01	0.40	0.03	5.65	-0.01	3.04	2.91	-0.02	0.16	89.96	1.42	0.00	0.00	0.09	0.44	0.00
SK	0.28	0.00	12.25	0.01	1.46	0.58	0.90	3.89	0.89	0.00	34.89	29.03	1.19	0.08	0.27	0.14	3.23	0.56	0.01	0.29	5.74	67.10	0.10	0.00	7.81	0.00	0.03	1.18	0.07	0.26	0.36	0.08	0.23	0.00	1.16	0.12	66.73	0.00	16.31	16.95	0.67	0.64	2.20	100.58	0.00	0.01	1.83	3.59	0.01
TJ	0.00	0.02	0.00	0.10	0.00	0.00	0.00	-0.01	0.00	0.00	0.00	-0.01	0.00	0.00	0.00	0.00	-0.01	0.00	0.03	0.00	0.00	0.00	0.00	0.00	0.00	2.20	1.33	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	0.00	-0.01	0.00	-0.23	0.00	0.00	0.00	42.25	0.97	0.41	-0.03	22.29
TM	0.00	0.12	0.01	1.17	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.02	0.00	0.00	0.00	0.00	-0.01	-0.01	0.28	-0.01	0.00	0.01	0.00	0.00	0.01	0.22	5.77	0.00	0.00	0.00	0.01	0.00	0.00	0.00	-0.01	0.00	-0.05	0.00	0.05	-0.02	0.84	0.00	0.00	0.00	1.04	35.10	1.43	0.17	9.81
TR	0.00	0.13	0.07	0.07	0.00	0.00	0.90	0.08	0.01	0.03	0.05	0.15	-0.01	0.00	0.03	-0.01	-0.01	-0.02	0.25	0.23	0.07	0.21	0.00	0.00	0.09	0.00	-0.12	0.01	0.00	0.00	0.09	0.00	-0.01	0.00	0.01	0.00	-0.13	0.00	1.18	0.43	-0.84	-0.01	0.02	0.01	0.00	-0.01	112.01	0.89	-0.01
UA	0.08	0.05	0.93	0.44	0.33	0.35	2.05	13.06	0.24	0.01	3.16	8.27	0.57	0.13	0.17	0.19	1.18	0.36	0.55	0.22	0.97	6.44	0.08	0.00	1.50	0.01	0.71	0.86	0.03	0.25	5.95	0.02	0.04	0.00	0.58	0.06	23.18	0.00	23.83	3.39	24.35	0.33	0.28	1.69	0.01	0.21	12.47	105.46	0.14
UZ	0.00	0.07	0.00	0.46	0.00	0.00	0.00	0.01	0.00	0.00	0.00	-0.02	0.00	0.00	0.00	0.00	-0.02	-0.01	0.14	-0.01	0.00	0.01	0.00	0.00	-0.01	3.31	4.70	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	-0.04	0.00	0.05	-0.02	-0.34	0.00	0.00	0.01	8.66	3.15	0.80	0.19	63.83