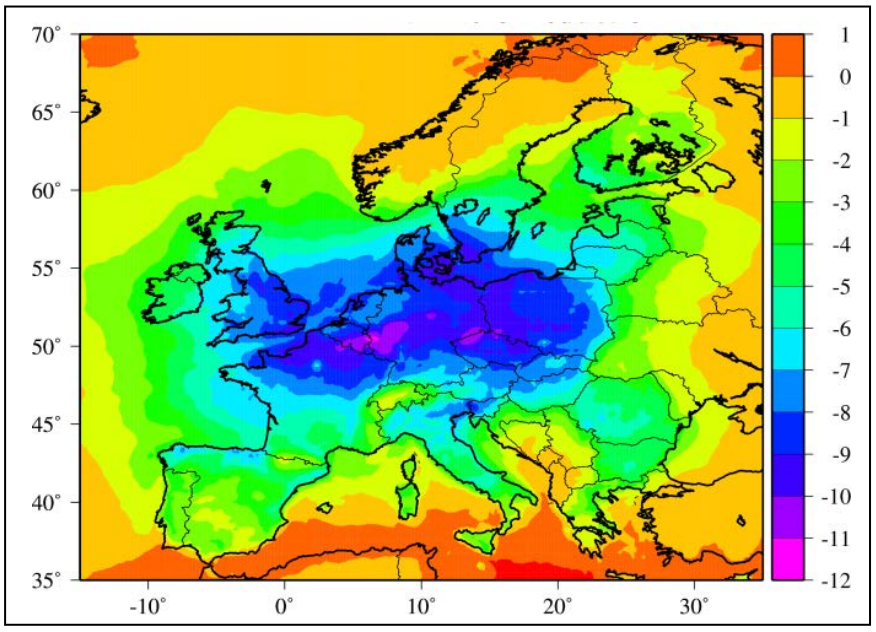
Sensitivity analysis of ammonia emission reductions on exceedances of PM air quality standards



ETC/ACM Technical Paper 2013/12
May 2013

Maxime Beauchamp, Bertrand Bessagnet, Cristina Guerreiro, Frank de Leeuw, Svetlana Tsyro, Paul Ruyssenaars, Ferd Sauter, Guus Velders, Frédérik Meleux, Augustin Colette, Laurence Rouïl

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 UBA-V ÖKO AEAT EMISIA CHMI NILU VITO INERIS 4Sfera PBL CSIC

Front page picture:
Simulation of annual mean $PM_{2.5}$ concentrations reductions (in %) for an ambitious 2020 scenario on ammonia emission reduction based on the emission reductions driven by the Gothenburg protocol.

Author affiliation:
Maxime Beauchamp, Bertrand Bessagnet, Frédérik Meleux, Laurence Rouïl, Augustin Colette: National Institute for industrial Environment and Risks (INERIS), France
Cristina Guerreiro: Norwegian Institute for Air Research (NILU), Norway
Frank de Leeuw, Paul Ruyssenaars, Ferd Sauter, Guus Velders: National Institute for Public Health and the Environment (RIVM), The Netherlands
Svetlana Tsyro: Norwegian Meteorological Institute (Met.no), Norway

DISCLAIMER

ETC/ACM Technical Paper 2013/12
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/

Executive summary

PM (Particulate Matter) concentration levels across Europe continue to cause significant negative impacts on human health. Although ammonium constitutes only a small fraction of the PM mass, it is an important component of secondary inorganic aerosols in a form of ammonium sulphate ($(NH_4)_2SO_4$)) and ammonium nitrate (NH_4NO_3). Several studies point out the importance of agricultural NH_3 emissions to PM concentrations in different European regions, highlighting the need to investigate the potential of NH_3 emission reductions to reduce PM levels over Europe. According to Deutsch et al. (2008) NH_3 emission abatement should be considered in order to reduce $PM_{2.5}$ levels. Erisman et al. (2008) estimate that NH_3 emissions from agriculture in EU15 give a substantial contribution to PM formation in Europe (13%) and states that much larger NH_3 emission reductions than foreseen in current legislation should be aimed at in order to meet the PM concentration targets. The sensitivity of secondary PM formation to NH_3 and other precursor gases emissions has been investigated in previous studies and results show considerable differences in results across Europe due to different climatological conditions and chemical regimes (NH_3 *vs* HNO_3 limited), as well as differences between models. Nevertheless, several studies show an important contribution of NH_3 emission reductions to reducing PM concentration levels in Europe.

The current study has used three different chemical transport models (CHIMERE, EMEP and LOTOS-EUROS) to quantify the reductions of $PM_{2.5}$ and PM_{10} concentrations due to reductions of NH_3 emissions beyond the Gothenburg Protocol (GP), as well as due to the GP alone compared to 2009. Simulations of $PM_{2.5}$ and PM_{10} concentrations using 2009 meteorology were undertaken for five emissions datasets: 2009 emissions (as the reference simulation), GP emissions in 2020, and further 10%, 20% and 30% NH_3 emission reductions in EU27 beyond the GP. Results show that the GP alone will contribute to a 12 to 21% reduction in exceedances of the PM_{10} daily limit value (LV) and 26 to 35% reduction in exceedances of the $PM_{2.5}$ annual LV in 2020, compared with 2009. Hence further measures are needed to achieve compliance.

The modeling results for the scenarios with further 10%, 20% and 30% NH_3 agriculture emission reductions in EU27 beyond the GP show that the reduction achieved in PM levels is not linear with the emission reductions. In fact, the results from the three models show that the higher the NH_3 emission reductions are, the more efficient is the reduction in $PM_{2.5}$ and PM_{10} concentrations. Moreover, the modelling study shows that the expected impact of ammonia emissions on the formation of particulate ammonium was underestimated by the three models. This would imply that the role of ammonia on PM concentration and exceedances of limit values is likely to be even larger than quantified in this study. Currently available mitigation measures could cut ammonia emissions in the EU27 by about 30% on top of current legislation in 2020 (Amman, 2012). A further reduction of 30% of NH_3 agriculture emissions will contribute to reduce the exceedances of the PM_{10} daily LV and $PM_{2.5}$ LV further by respectively 4-8% and 4-7% in 2020, compared to the GP. Annual mean concentrations of $PM_{2.5}$ across Europe may also be considerably reduced, especially in Central and Central-Eastern Europe, reaching a 8-10% reduction in Belgium, Germany, the Czech Republic and Poland. The percentage reduction of the PM_{10} annual mean concentrations over Europe are lower than for $PM_{2.5}$, as expected, ranging from 3 to 8% over most of Europe.

This study shows that the implementation of the emissions reductions imposed by the revised GP for 2020 will not suffice to achieve compliance with PM standards in Europe; hence further European measures should be considered. NH_3 emissions from agriculture can be further reduced with the implementation of proven and feasible measures, in order to reduce PM levels and their impacts on human health across Europe.

Table of content

1 Introduction

One of the most noxious problems in air quality is the persistence of high concentrations of particulate matter (PM) (EEA, 2012). In Europe, about one third of PM_{10} and half of $PM_{2.5}$ have an inorganic chemical speciation consisting in: ammonium (NH_4^+), nitrate (NO_3^-) and sulphate (SO_4^{2-}). These species are the product of oxidation of PM precursor gases: ammonia (NH_3), nitrogen oxides (NO_x) and sulphur oxides (SO_x). Although NH_3 by itself makes a small fraction of the PM mass it 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) as PM constituents.

Historically, the sources of SO_x and NO_x were not too difficult to control. Emissions of the precursors gases SO_x and NO_x declined by 54 % and 26 % in the period 2001–2010. Ammonia emissions have fallen less: only about 10 % between 2000 and 2010. The agricultural sector was responsible for 94 % of the total NH_3 emissions in the EU in 2010. Ammonia emissions are largely from animal excreta and fertilizers. Continued reductions in SO_x and NO_x emissions are likely to decrease in efficiency (and increase in costs) with respect to PM abatements, if strategies for reducing NH_3 emissions are not balanced in properly.

According to Erisman and Schaap (2004) inorganic PM concentrations can only be reduced effectively if all three precursor gases NO_x, SO_x and NH_x are reduced to the same extent. Since that study, the "chemical landscape" of Europe has changed; it is time to re-evaluate the potential for NH_3 emission reductions in the control strategy for PM concentrations. So far, the Gothenburg Protocol[1] under the LRTAP (Long Range Transboundary Air Pollution) convention (see Annex 1) and the National Emission Ceilings Directive[2] set emission reduction targets for NH_3 primarily with the aim of reducing the acidification and eutrophication. Abatement of NH_3 emissions is also required by the Directive 2010/75/EU[3] on industrial emissions, the Nitrates and Water Framework Directives (91/676/EEC[4] and 2000/60/EC[5]), as well as EURO VI emission limits heavy duty vehicles, becoming mandatory for all new registrations from 2014 (Regulation 595/2009/EC[6]). In view of the future negotiations of a revised NEC directive and to conceive strategies to deal with the recurrent PM exceedances in Europe, the policy makers need to be informed about the reduction options in the emissions of NH_3 and the other inorganic precursors which can lead to the desired reductions of inorganic PM concentrations. However, for the future, the projected changes in ammonia emissions indicate either low ammonia emission reductions or possibly increasing ammonia emissions, depending on the considered scenarios (Figure 1).

[1] PROTOCOL to the 1979 Convention on long range transboundary air pollution to abate acidification, eutrophication and ground level ozone.
[2] DIRECTIVE 2001/81/EC OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 23 October 2001 on national emission ceilings for certain atmospheric pollutants
[3] DIRECTIVE 2010/75/EU OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 24 November 2010 on industrial emissions (integrated pollution prevention and control).
[4] Council Directive 91/676/EEC of 12 December 1991 concerning the protection of waters against pollution caused by nitrates from agricultural sources
[5] DIRECTIVE 2000/60/EC OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 23 October 2000 establishing a framework for Community action in the field of water policy
[6] REGULATION (EC) No 595/2009 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 18 June 2009 on type-approval of motor vehicles and engines with respect to emissions from heavy duty vehicles (Euro VI) and on access to vehicle repair and maintenance information and amending Regulation (EC) No 715/2007 and Directive 2007/46/EC and repealing Directives 80/1269/EEC, 2005/55/EC and 2005/78/EC

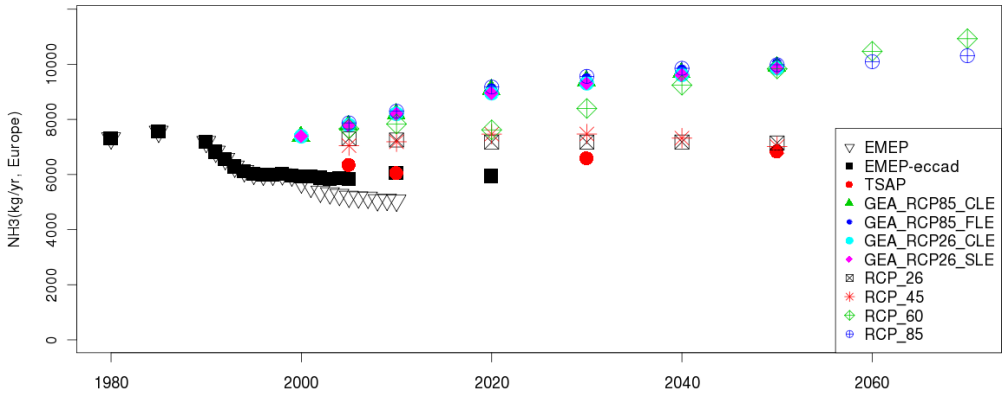


Figure 1: Ammonia emission projections in Europe for different scenarios, TSAP[7] (Amman et al., 2012) is the EU official scenario developed by IIASA, RCPs[8] are IPCC[9] scenarios, GEA[10] are climate and air quality scenarios developed by IIASA (Riahi et al., 2012). EMEP is the official UNECE emission inventory. (Pers. Comm. Augustin Colette –INERIS– with ECCAD[11] numbers).

As shown below in Figure 2, the reduction expected by the Gothenburg Protocol by 2020 (compared to 2005) is usually lower than 10% in most of EU countries. The average reduction of ammonia emissions for the EU27 is **6%.**

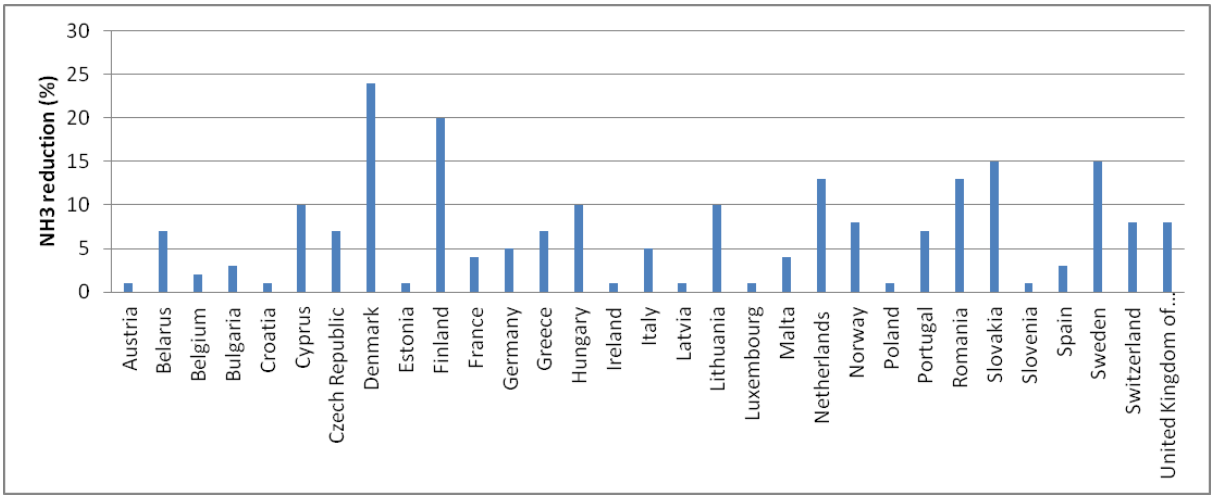


Figure 2: Ammonia emission reductions in Europe expected by the Gothenburg Protocol (reduction in 2020 calculated versus 2005 emissions)

Although there is less ambition in reducing NH_3 emissions, than other PM precursors, there are proven and feasible methods to control and mitigate ammonia emissions from agriculture, including 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. López-Aparicio et al. (2013) presents a review of available NH_3 emission control and mitigation measures for agriculture. The available

[7] Thematic Strategy on Air Pollution
[8] Representative Concentration Pathways
[9] Intergovernemental Panel on Climate Change
[10] Global Energy Assessment
[11] Emissions of atmospheric Compounds & Compilation of Ancillary Data: (http://eccad.pole-ether.fr/eccad_extract_interface/JSF/page_login.jsf)

measures could cut ammonia emissions in the EU27 by about 30% on top of current legislation in 2020 (Amman, 2012).

2 Previous studies

Several studies point out the importance of agricultural emissions to PM concentrations, and particularly of NH_3 emissions to $PM_{2,5}$. For example, Deutsh et al. (2008) estimates that the Flemish agricultural emissions contribute to 12% of the $PM_{2,5}$ levels in Flanders, and to 22% of the PM_{10} levels in Flanders. The high contribution of agricultural emissions to PM_{10} is predominantly due to high emissions of primary particles in the coarse[12] fraction, while the high contribution of the agricultural emissions to the $PM_{2.5}$ is due to the formation of ammonium nitrate and ammonium sulphate in the atmosphere. According to Deutsch et al. (2008) NH_3 emission abatement should be considered in order to reduce $PM_{2,5}$ levels. Erisman et al. (2008) estimate that NH_3 emissions from agriculture in EU15 give a substantial contribution to PM formation in Europe (13%), as well as acidifying emissions (31%) and especially eutrophying emissions (45%) of ecosystems. It states further that much larger NH_3 emissions reductions than foreseen in current legislation should be aimed at, in order to meet targets on acidification, eutrophication and PM concentrations.

The sensitivity of secondary PM formation to NH_3 and other precursor gases emissions has been investigated in different studies. Erisman and Schaap (2004) has investigated the role of ammonia in particle formation and found that secondary PM can only be efficiently reduced if ammonia emissions are reduced in much the same way as SO_2 and NO_x emissions. They state that after the neutralisation of sulphate, nitrate may (partially) compensate for the decline in sulphate (due to SO_2 emission reductions), especially at low temperatures. The results of their modelling exercise using the LOTOS-EUROS model showed that, except for south-western France and Spain, where temperatures are generally high and relative humidity low, ammonia emission reductions are more effective for decreasing Secondary Inorganic Aerosol (SIA as the sum of sulphate, nitrate and ammonium) than SO_2 and NO_x emission reductions.

The EURODELTA II study (Thunis et al, 2008) showed that the relative effectiveness of different emission reductions (NO_x, SO_2, NMVOC[13]s and NH_3) for $PM_{2.5}$ concentration reduction may vary considerably from model to model, as shown in Figure 3. The effectiveness of NH_3 emission reductions in the UK is large compared with NH_3 reductions in other countries and much greater than the effectiveness of reduction of other precursor emissions. In the other countries the ammonia effectiveness is less than or similarly to NO_x or SO_2 emission reductions. According to Thunis et al. (2008) the most effective way of reducing $PM_{2.5}$ concentrations is to reduce primary $PM_{2.5}$ emissions.

[12] Coarse particles refer to particles with diamater above 2.5 µm
[13] Non Methanic Volatil Organic Compounds

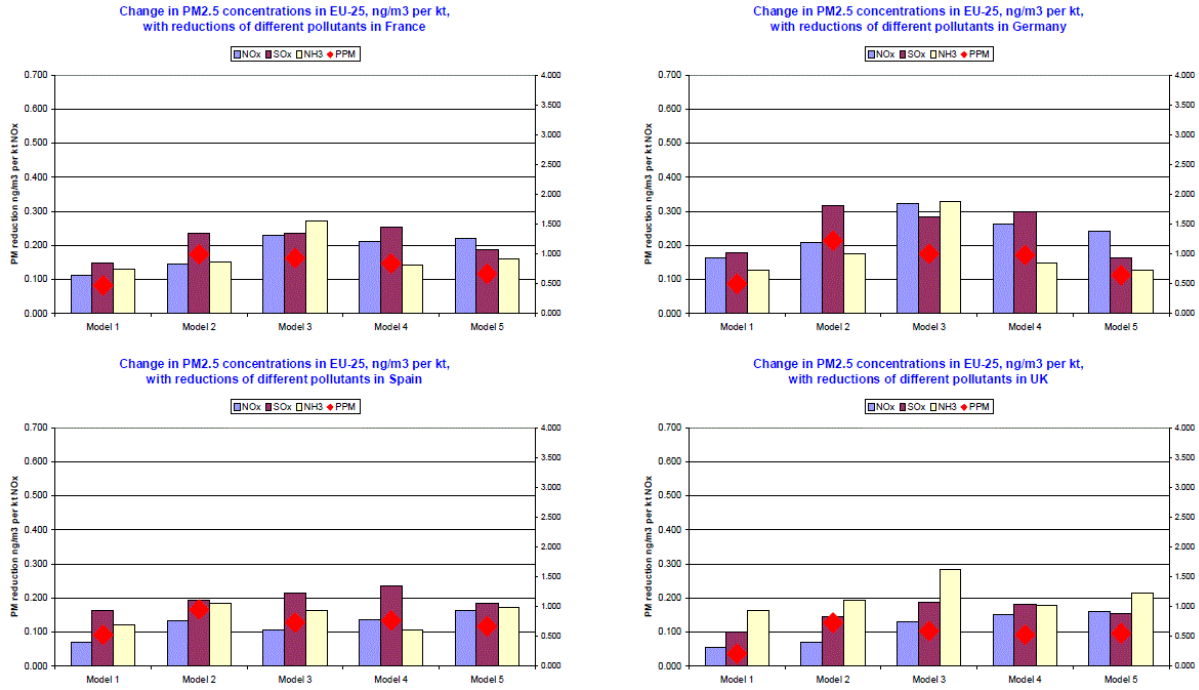


Figure 3: Comparison of effect of reducing NO_x, SO_x, NH_3 and PPM (as Primary PM) emissions on $PM_{2.5}$ concentrations. Effects shown are for the whole EU-25. Primary $PM_{2.5}$ is on the right hand scale, all other pollutants are on the left hand scale. (Figure 16 in Thunis et al., 2008).

For a rural location in southern England, Derwent et al. (2009) examined the linearity of the formation of the secondary PM components by sensitivity studies to 30% reductions in SO_2, NO_x, NH_3, VOC and CO emissions. The chemical environment revealed by these sensitivity studies appeared to be "ammonia-limited" and the PM mass concentrations appeared to be markedly non-linear with PM precursor emissions. The largest reduction in $PM_{2.5}$ mass is modelled for a 30% reduction in NH_3 emissions; however, all precursor reductions except NH_3 result in a reduction in nitrate in coarse PM. The increase in coarse nitrate by reduced NH_3 emissions is described to the interaction between nitric acid, ammonia and sea salt. They have therefore concluded that policy strategies for $PM_{2.5}$ need to take into account emission reductions for a wide range of primary PM components and secondary PM precursors and to focus primarily on the abatement of NH_3. They say further that better understanding of this complex interlinking between emissions and PM formation may help to explain why PM levels have remained constant despite falling primary PM emissions.

Similarly Harrison et al. (2013) modelled concentrations of SIA in PM_{10} at a rural site in Harwell (UK) for a relatively high pollution period (19 March – 19 May 2007). The response of concentrations at Harwell to reductions of precursor emissions (SO_2, NO_x and NH_3) for 1) across the UK only, 2) mainland Europe only, and 3) the whole of Europe has been modelled. As in earlier studies, they showed that the total reductions in SIA concentrations are less than linear with the emission reductions for all precursors. They also showed that the abatement of SO_2 emissions leads to an increase in nitrate concentrations whereas reductions of NO_x lead to increases in sulphate. Further they predict a low response of nitrate aerosol concentrations to NO_x emission reductions. Table 1 shows the results of this study and compares them with the previous study by Derwent et al. (2009) in terms of concentrations (at the same rural site in Harwell)of sulphate, ammonium and $PM_{2,5}$ due to 30% emission reductions of SO_2, NO_x or NH_3, and compared to no emission reductions. Both studies give very similar results and the 30% emission reduction of NH_3 over the whole Europe leads to the highest reduction in SIA concentrations (9%) at the modelled site. Comparatively, a 30% reduction in NO_x and SO_2 emissions lead to a 5% and 6% reduction in SIA concentrations, respectively.

Table 1: Comparison of Derwent et al. (2009) and Harrison et al. (2013) concentration results of nitrate, sulphate, ammonium and $PM_{2.5}$ (compared to reference) due to emission reductions of SO_2, NO_x and NH_3 (Table 1 in Harrison et al., 2013)

Derwent et al., 2009 mean 15.00z values, 2006					Current work 19 March–19 May 2007				
With respect to base case values	Sulphate	Nitrate	Ammonium	$PM_{2.5}$	With respect to no emission reduction	Sulphate	Nitrate	Ammonium	SIA
Across the board cases					**Reductions in all of Europe**				
30% SO_2 case	70%	105%	94%	93%	30% reduction in SO_2	78%	108%	87%	94%
30% NO_x case	105%	80%	92%	96%	30% reduction in NO_x	101%	87%	94%	95%
30% NH_3 case	100%	83%	79%	92%	30% reduction in NH_3	97%	86%	83%	91%
UK-only cases					**Reductions in UK only**				
30% SO_2 case	85%	101%	99%	97%	30% reduction in SO_2	89%	104%	94%	97%
30% NO_x case	103%	92%	97%	99%	30% reduction in NO_x	100%	94%	98%	97%
30% NH_3 case	100%	92%	90%	96%	30% reduction in NH_3	97%	92%	90%	94%
Rest of Europe-only cases					**Reductions in rest of Europe only**				
30% SO_2 case	85%	115%	96%	97%	30% reduction in SO_2	89%	104%	93%	97%
30% NO_x case	101%	88%	94%	97%	30% reduction in NO_x	101%	94%	97%	98%
30% NH_3 case	100%	91%	89%	96%	30% reduction in NH_3	100%	94%	94%	97%

Renner and Wolke (2010) found that a reduction of 50% in the NH_3 regional emissions from agriculture in Germany lead to a maximum reduction of 30% in ammonium nitrate concentrations, while ammonium sulphate remained unchanged.

Another study by Megaritis et al. (2012) found that reducing NH_3 emissions seems to be the most effective control strategy for reducing $PM_{2.5}$, when compared to reductions of other precursor gases, mainly due to a significant decrease of ammonium nitrate. Their modelling results indicated an average reduction of $PM_{2.5}$ concentrations over Europe of 5.5% during summer and 4% during Winter, due to a 50% reduction in NH_3 emissions. While for a 50% reduction in NO_x and SO_2 emissions, the simulated $PM_{2,5}$ reductions were respectively 5% and 5.1% in summer and 0,4% and 2,6% in winter. Megaritis et al. (2012) findings for winter are consistent with other studies (Tsimpidi et al., 2007; Odman et al., 2009; Aksoyoglu et al., 2011). During summer, the 50% NH_3 emission reduction resulted in a decrease of ammonium by 22% over the entire model domain. Nitrate was reduced by 35% in Western Europe and by 27% in southwest Europe, while the corresponding decrease of total $PM_{2.5}$ in these areas was 15% and 10% respectively. Similarly in winter, ammonium was decreased by 24% over the domain, with an average 20% decrease in nitrate concentration. Overall, ammonium nitrate reduction accounts for almost 80% of total $PM_{2.5}$ reduction in both periods. The reduction of NH_3 produces also a slight decrease of sulphate levels due to the effect of NH_3 on cloud pH and on the rate of in-cloud sulphate production.

Pay et al. (2012) found on the other hand that the continental regions in Europe tend to be HNO_3 limited for nitrate formation, rather than NH_4-limited. They concluded that the formation of SIA in Europe tends to be limited by SO_2 and HNO_3 gaseous precursors due to the relatively high NH_3 emissions, mainly from agriculture, especially in northwestern Europe. So they recommend regulatory strategies in this part of Europe to be focused on the reduction of NO_x and SO_2 rather than NH_3 emissions. The comparison of their modeling results with EMEP measurements has nevertheless shown that the model overestimates the ratio Free ammonia[14]/Total-NO_3 over the Iberian Peninsula and at some coastal stations in north and north-east Europe, indicating that these areas are more NH_4-limited than the model results suggest.

A study in the Netherlands (Weijers et al., 2010) showed that the SIA contribution to PM10 and $PM_{2.5}$ in the Netherlands is higher than previously thought, pointing to a need to focus more on the mitigation of PM precursors emissions in the Netherlands and in Europe in order to attain PM limit values. They found that SIA dominates the PM composition, especially when increased PM levels

[14] Free ammonia indicator quantifies the amount of ammonia available, after neutralizing SO_4^{2-}, for NH_4NO_3 formation. This indicator is based on the fact that $(NH_4)_2SO_4$ aerosol is the favored form for sulfate. Free ammonia is defined as the total ammonia minus twice the sulfate concentration on a molar basis.

occur. The average contribution of SIA to PM_{10} in the Netherlands was estimated to be 30 to 40%, increasing to between 45 and 55% on days when PM_{10} was above 40 µg m^{-3} and becoming 25 to 35% when PM_{10} was less than 40 µg m^{-3}. Long-range transport and meteorology are the main factors influencing these higher levels.

3 Aim of the present study

The objective of this study is to analyse the impact of the Gothenburg Protocol emission reductions and additional ammonia emission reductions on European PM concentrations levels and on the exceedances of the air quality standards for PM. To tackle this issue, a set of model runs are performed by the ETC/ACM for the reference year 2009 and several emissions reduction scenarios, using three different models to consider the variability of model responses (Table 2). They are based on the CHIMERE model (Menut et al., 2013) which has already been implemented and evaluated in France for scenario analysis and sensitivity studies (Bessagnet et al. 2005), as well as LOTOS-EUROS (Schaap et al., 2008; Sauter et al., 2012) used for the Dutch authorities and the EMEP model (Simpson et al, 2012), a CTM model widely used for policy support in Europe. The use of several models results can provide valuable information on the uncertainty of the analysis. The calculations of PM concentrations including NH_4^+, NO_3^- and SO_4^{2-} are performed using the 2009 meteorology for the whole year. Emission inputs to dispersion modelling consist in a European emission inventory with a spatial resolution of 0.125°×0.0625°. The models need to demonstrate their reliability as a basis for policy and decision making. Moreover, the model predictions of the speciated inorganic PM concentrations calculated in the baseline run (using current emissions) would need to be compared and found consistent with observations.

Table 2: Models involved in the study

Team	Model	Model acronym in this report
INERIS	CHIMERE	CHIM
NILU/Met.NO	EMEP (MSC-W)	EMEP
RIVM	LOTOS-EUROS	LOTO

The analyses will be based on the impact of NH_3 emission reductions on PM concentrations. An extract of EU air quality standards for PM is presented in Table 3.

Table 3: EU air quality standards for PM

Pollutant	Concentration	Averaging period	Legal nature	Permitted exceedances each year
Particles (PM_{10})	50 µg m^{-3}	24 hours	LV* entered into force 1.1.2005 (a)	35
	40 µg m^{-3}	year	LV entered into force 1.1.2005 (a)	n/a
Fine particles ($PM_{2.5}$)	25 µg m^{-3} 20 µg m^{-3}	year	entered into force as TV by 1.1.2010 as LV by 1.1.2015 LV**(b) enters into force 1.1.2020	n/a

(a) Under the new Directive the Member State was able to apply for an extension until three years after the date of entry into force of the new Directive (i.e. May 2011) in a specific zone. Request was subject to assessment by the Commission. In such cases within the time extension period the limit value applies at the level of the limit value + maximum margin of tolerance (35 days at 75µg/m3 for daily PM10 limit value, 48 µg/m^3 for annual PM10 limit value).

(b) Indicative limit values to be reviewed by the European Commission in 2013

* Target value, ** Limit value

Under EU law a limit value is legally binding from the date it enters into force subject to any exceedances permitted by the legislation. A target value is to be attained as far as possible by the attainment date and so is less strict than a limit value.

4 Emissions

4.1 Scenarios

One reference scenario was based on official EMEP emissions for 2009 (update in 2012). Four scenarios were built, one representing the Gothenburg 2020 protocol emissions and three scenarios are expected to show the effect of an additional effort on ammonia emission reductions (Table 4). The Gothenburg protocol emissions ceilings are built with the national emission ceiling numbers reported in Annex II issued from UNECE (2012) (see Annex 1 of this report). Coarse PM primary emissions have not been given in the Gothenburg protocol; estimates for the national emissions of primary coarse PM are obtained from the EMEP webdab emission web site (see Annex 1). To allocate the national emission by activity sector, the same sectoral distribution by pollutant of the reference year 2009 is applied for these national ceilings.

Table 4: Description of scenarios

Scenario name in this report	Description
2009REF (or REF)	Official 2009 (from EMEP numbers downloaded on www.emep.int and updated in 2012)
2020GOT (or GOT)	Gothenburg 2020 emissions
2020G10 (or G10)	Gothenburg 2020 + additional 10% reduction for NH3, EU27 only
2020G20 (or G20)	Gothenburg 2020 + additional 20% reduction for NH3, EU27 only
2020G30 (or G30)	Gothenburg 2020 + additional 30% reduction for NH3, EU27 only

4.2 Spatial pattern of emissions

The first step consists in calculating the spatial patterns of emissions for the year 2007. The year 2007 was selected to take advantage of the high resolution MACC inventory generated by TNO (Kuenen et al. 2011).

The emission dataset was delivered by INERIS for all model resolutions separately. Prescribed time profiles and height distributions were used following the EURODELTA protocol (*cf* Thunis et al., 2008).

The gridded distribution of anthropogenic emissions used for this exercise provided by INERIS are based on a merging of databases from:

- *TNO 0.125°×0.0625° emissions for 2007 from MACC (cf. Kuenen et al., 2011)*
- *EMEP 0.5°×0.5° for 2009 (cf. Vestreng et al., 2007)*
- *Emission data from the GAINS database (see http://gains.iiasa.ac.at/gains)*
- *INERIS expertise on re-gridding with various proxies (population, landuse, Large Point Source data)*

First the large point sources (LPS) from the fine scale (0.125°×0.0625°) TNO-MACC emissions data for 2007 were added to surface emissions to get only one type of emissions. For the various activity sectors the processing steps were the following:

- SNAP 2: The country emissions were re-gridded with coefficients based on population density and French bottom-up data, the methodology (Bessagnet et al., 2012) was extrapolated to the whole of Europe. For $PM_{2.5}$ emissions, the annual EMEP totals were kept except for the countries CZ, BA,BE, BY, ES, FR, HR, IE, LT, LU, MD, MK, NL, CS, TR. For these countries, $PM_{2.5}$ emissions from GAINS were used. Additional factors were applied on two Polish regions (×4 or ×8) for $PM_{2.5}$ and PM_{10} emissions (Personal communication from IIASA) with formerly coal mining activity. These coal mine regions still show high emissions of PM due to continued domestic uses of coal.

- SNAP 3,7,8,9,10: TNO-MACC emissions spatial distribution was used as proxy to regrid EMEP 0.5°x0.5° annual totals into the finer modelling grid.
- SNAP 1,4,5,6: EMEP 0.5°x0.5° emissions were regridded by adequate proxies ("artificial landuse", EPER data for industries[15]).

For countries where TNO-MACC emissions are not available EMEP 0.5°×0.5° emissions are used (Iceland, Liechtenstein, Malta and Asian countries) and reggrided with adequate proxies ("artificial landuse", EPER data for industries).

4.3 Calculation of emissions

The spatial distribution calculated in the previous section for the year 2007 is scaled by sectors and countries for each emitted pollutant to calculate the emissions for a given scenario listed in Table 4 and the reference year 2009. For countries where emission ceiling are not mentioned in the protocol, 2020 CLE emissions (IIASA, 2004) are used (constituting other European countries and maritime emissions). CO and PPMcoarse emissions are also from 2020 Current Legislation (CLE) emissions for the 2020 scenarios.

The list of the following emitted species has been used in the models:

- $PPM_{2.5}$: Primary PM with particle diameter below 2.5 µm
- PPM_{coarse} : Primary PM with particle diameter between 2.5 µm and 10 µm
- NH_3 : Ammonia
- NO_x : Nitrogen oxides
- NMVOC : Non methanic volatile organic compounds
- SO_2 : Sulphur dioxide
- CO : Carbon monoxide

PPM includes elemental carbon, organic material and other anthropogenic dust.

5 Simulation with Chemistry transport models

5.1 Methodology

The simulations were performed with the three models over an area encompassing the EU27 (except Azores and Canary Islands). The 2009 meteorology was chosen as a reference year. The four scenarios described below were simulated using the 2009 meteorology.

The simulations were performed over different domains with different resolutions by the modelling teams (Table 5). **In this study we assume that a low resolution simulation (0.25° to 0.5°) is sufficient to study the effect of additional ammonia emission reduction measures on PM concentrations.** This assumption is supported by Cuvelier et al. (2013) who studied the impact of the horizontal resolution in air quality models.

[15] The EPER Decision is based on Article 15(3) of Council Directive 96/61/EC concerning integrated pollution prevention and control. EPER is a web-based register, which enables the public to view data on emissions to water and air of 50 key pollutants from large and medium-sized industrial point sources in the European Union. The register is hosted by the European Environment Agency. http://www.eea.europa.eu/data-and-maps/data/eper-the-european-pollutant-emission-register-4

Table 5: Model domains and resolution

Model	Resolution	Computation domain
CHIMERE	0.25° x 0.25°	-15°E/35.25°E – 35°N/70.25°N
EMEP	0.25° x 0.25°	-25°E/45°E – 30°N/70°N
LOTOS-EUROS	0.5° x 0.25°	-14.75°E/34.75°E – 35.125°N/69.875°N

Only the emissions were prescribed, all other input parameters were not prescribed. This means that the models use different meteorological input data as well as land use data. The output required for the exercise was prescribed on an hourly basis. The output species contain the Secondary Inorganic Aerosols (SIA is the sum of ammonium, sulphate and nitrate) and the total PM in both 2.5 and 10 µm fractions (PM_{10} and $PM_{2.5}$). In the report we use the following acronyms for SIA and their precursors:

- NO3-10 : Nitrate for particles with diameter below 10 µm
- NH4-10 : Ammonium for particles with diameter below 10 µm
- SO4-10 : Sulphate for particles with diameter below 10 µm
- HNO3 : nitric acid (gas)
- NH3 : ammonia (gas)
- TNO3 : Total nitrate (TNO3 = NO3-10 + $HNO3_{eq.\ Nitrate}$)
- TNH4 : Total ammonium (TNH4 = NH4-10 + $NH3_{eq.\ Ammonium}$)

The participating models differ in the availability of PM components and formation routes. For instance, EMEP and LOTOS-EUROS contain coarse mode nitrate formation, whereas CHIMERE does not. Also, CHIMERE and EMEP compute Secondary Organic Aerosol (SOA), where the LOTOS-EUROS modelling team considers their SOA model formulations too uncertain for use in policy support. In each model, the PM_{10} concentration is calculated as follows in each model:

EMEP $PM_{10} = PPM_{coarse} + PPM_{fine} + SO_4^{2-} + NO_3^- + NH_4^+ + \text{Sea Salt} + \text{SOA} + \text{Dust}$

CHIMERE $PM_{10} = PPM_{coarse} + PPM_{fine} + SO_4^{2-} + NO_3^- + NH_4^+ + \text{Sea Salt} + \text{SOA} + \text{Dust}$

LOTOS-EUROS $PM_{10} = PPM_{coarse} + PPM_{fine} + SO_4^{2-} + NO_3^- + NH_4^+ + \text{Sea Salt}$

where PPM stands for Primary Particulate Matter. As mentioned later in Table 6, the various contribution of resuspended dust is treated differently in the models. In CHIMERE, dust comes only from the boundary conditions, no resuspension schemes are used, SOA have biogenic and anthropogenic origins (Bessagnet et al., 2009). The EMEP model includes on-line calculated windblown and road dust and also African dust from boundary conditions; SOA originates from both anthropogenic and biogenic VOCs. LOTOS-EUROS includes a dust model with dust emissions from windblown dust, resuspension by traffic and agricultural land management; dust boundary conditions are taken from the MACC/G-AER data (Denier van der Gon et al., 2009; Schaap et al., 2009).

5.2 Synthetic model description

The models are synthetically described in Table 6 and Table 7 with the main references for the various chemical and physical processes.

Table 6: Model description part 1

MODEL	EMEP MSC-W	CHIMERE	LOTOS-EUROS
version	V4.1.3	CHIMERE2013a	V1.8
operator	met.no	INERIS/IPSL-CNRS	TNO/KNMI/RIVM
contact	Svetlana Tsyro	Maxime Beauchamp Bertrand Bessagnet	Martijn Schaap Ferd Sauter
email	s.tsyro@met.no	maxime.beauchamp@ineris.fr bertrand.bessagnet@ineris.fr	Martijn.Schaap@tno.nl Ferd.Sauter@rivm.nl
RESOLUTIONS			
Vertical layers	20 sigma	9 sigma	4 (3 dynamic layers and a surface layer)
Vertical extent	100 hPa	500 hPa	3500 m
Depth first layer	90 m	20 m	25 m
DYNAMIC AND BIOGENIC EMISSIONS			
BVOC	Based upon maps of 115 species from Koeble and Seufert (2001) and hourly temperature and light. See Simpson et al. (2012)	MEGAN model (Guenther et al., 2006)	Based upon maps of 115 species from Koeble and Seufert (2001) and hourly temperature and light. See Denier van der Gon et al. (2009)
Forest fires	Not used here	Not used here	MACC GFAS
Soil-NO	Simpson et al. (2012)	MEGAN model (Guenther et al., 2006)	Not used here
Lightning	Climatological fields from Köhler et al. (1995)	None	None
Sea salt	Monahan et al. (1986) and Martensson (2003), see Tsyro et al. (2011).	Monahan et al. (1986)	Martensson et al. (2003) and Monahan et al. (1986)
Windblown Dust	Parameterisation described in Simpson et al. (2012)	Vautard et al. (2005), not used here	Denier van der Gon et al. (2009).
Agricultural land management	None	None	Denier van der Gon et al. (2009),
Dust traffic suspension	Denier van der Gon et al. (2009).	None	Denier van der Gon et al. (2009),
Saharan dust inflow	Yes	Yes	Not used here
LANDUSE			
Landuse database	CCE/SEI for Europe (de Smet, and Hettelingh, 2001), elsewhere Corine Land Cover 2000.	GLOBCOVER (24 classes), Bicheron et al. (2008)	Corine Land Cover 2000 (13 classes)
Resolution	Flexible, CCE/SEI ~ 5 km	~300 m	1/60 x 1/60 degrees
BOUNDARY CONDITIONS			
Boundary conditions	MACC	Global model INCA (Guibert et al., 2005)	MACC
Frequency	Daily for all species	Monthly climatologies for all species	Boundary conditions are provided hourly or 3-hourly depending on the species

Table 7: Model description part 2

MODEL	EMEP MSC-W	CHIMERE	LOTOS-EUROS
METEOROLOGY			
Description	ECMWF	ECMWF	ECMWF
Resolution	0.25 deg x 0.25 deg	0.25 deg x 0.25 deg	0.25 deg x 0.25 deg
Advection	Bott (1989a,b) scheme	Van Leer (1984) scheme	Walcek (1998) scheme
Vertical diffusion	Kz approach	Kz approach following Troen and Mart (1986)	Kz approach
PROCESSES			
Dry deposition	Resistance approach for gases, Venkatram and Pleim (1999) for aerosols (Simpson et al., 2012)	Resistance approach Emberson (2000a,b)	DEPAC3.1, Van Zanten et al. (2010), Wichink Kruit et al. (2010)
Landuse class	16 classes	9 classes	9 classes
Compensation points	No, but zero NH_3 deposition over growing crops	No	Only for NH_3 (for stomatal, external leaf surface and soil (= 0))
Stomatal resistance	DO3SE-EMEP: Emberson et al, 2000, Tuovinen et al., 2004. Simpson et al., 2012	Emberson (2000a,b)	Emberson (2000a,b)
Wet deposition gases	In-cloud and sub-cloud scavenging coefficients	In-cloud and sub-cloud scavenging coefficients	In-cloud and sub-cloud scavenging coefficient
Wet deposition particles	In-cloud and sub-cloud scavenging	In-cloud and sub-cloud scavenging	In-cloud and sub-cloud scavenging coefficient
Gas phase chemistry	EmChem09soa in Simpson et al. (2012)	MELCHIOR	TNO-CBM-IV
Cloud chemistry	Aqueous SO_2 chemistry, pH dependent	Aqueous SO_2 chemistry and ph dependent SO_2 chemistry. The pH is calculated.	Banzhaf et al. (2011)
Coarse nitrate	Yes	No	Yes
Ammonium nitrate equilibrium	MARS (Binkowski and Shankar, 1995)	ISORROPIA (Nenes et al., 1999)	ISORROPIA2 (Fountoukis and Nenes, 2007)
SOA formation	VBS-NPAS –Simpson et al. (2012)	After Bessagnet et al., 2009	Not used
VBS	Yes, Bergström et al. (2012), Simpson et al. (2012)	No	Not used
Aerosol model	Bulk- approach (2 modes)	8 bins (40 nm - 10 µm)	Bulk- approach (2 modes)
Aerosol physics	Not used	Coagulation / condensation / nucleation	Not used

6 Models evaluation

The evaluation is carried out on the available EMEP observations for 2009. Only daily observations were used. All available data were used (Table 8). The measurements were downloaded from the EBAS database[16]. All the observations are background measurements coherent with the resolutions of the models in this exercise.

Table 8: Number and name of sites used for the evaluation for each pollutant – Information on station locations, altitude and other metadata can be found at http://ebas.nilu.no/

Pollutant	Number of EMEP sites	EMEP Sites
PM_{10}	72	AT02, AT05, AT48, CH01, CH02, CH03, CH04, CH05, CY02, CZ01, CZ03, DE01, DE02, DE03, DE07, DE08, DE09, DE44, DK05, ES01, ES06, ES07, ES08, ES09, ES10, ES11, ES12, ES13, ES14, ES16, ES17, ES78, FR09, FR13, FR15, FR18, GB06, GB36, GB43, GB48, GR02, HU02, IT01, LV10, LV16, MD13, NL07, NL09, NL10, NL91, PL05, SE11, SE12, SE14, SI08, FR09, FR13, FR15, FR18, GB06, GB36, GB43, GB48, GR02, HU02, NL07, NL09, NL10, NL91, SE11, SE12
$PM_{2.5}$	48	AT02, CH02, CH05, CY02, CZ03, DE02, DE03, DE44, ES01, ES07, ES08, ES09, ES10, ES11, ES12, ES13, ES14, ES16, ES78, FR09, FR13, FR15, FR18, GB36, GB48, GLOB, IE31, IT04, LV10, LV16, NL09, NL10, NL11, NL91, PL05, SE11, SE12, SE14, SI08, FR09, FR13, FR15, FR18, GB36, GB48, IE31, SE11, SE12
SO4-10 [(a)]	38	AM01, AT02, CH01, CH02, CH05, CZ01, CZ03, DE44, DK03, DK05, DK08, DK31, ES01, ES07, ES08, ES09, ES10, ES11, ES12, ES13, ES14, ES16, ES17, ES78, FI09, FI17, FI36, FR09, FR13, FR15, GB02, GB06, GB07, GB13, GB14, GB36, GB48, HU02
NO3-10	47	AM01, AT02, DE44, ES01, ES07, ES08, ES09, ES10, ES11, ES12, ES13, ES14, ES16, ES17, ES78, GB36, GB48, HU02, IE05, IE06, IE08, IT01, KZ01, LV10, LV16, MD13, NL08, NL10, NL11, NL91, NO01, NO15, NO39, NO42, NO55, NO56, PL02, PL03, PL04, PL05, RU18, SK02, SK06, GB36, GB48, NL11
NH4-10	42	AM01, AT02, DE44, DK03, DK05, DK08, DK31, ES09, ES78, FI09, FI17, FI36, GB36, GB48, HU02, IE05, IE06, IE08, IT01, LV10, LV16, MD13, NL08, NL10, NL11, NL91, NO01, NO15, NO39, NO42, NO55, NO56, PL02, PL03, PL04, PL05, RU18, SK06, GB36, GB48, NL11
TNO3	47	AT02, CH02, CH05, CZ01, CZ03, DK03, DK05, DK08, DK31, ES01, ES07, ES08, ES09, ES10, ES11, ES12, ES13, ES14, ES16, ES17, FI09, FI17, FI36, FR09, FR13, FR15, HU02, IE01, IT01, LT15, LV10, LV16, MD13, NO01, NO15, NO39, NO55, NO56, PL02, PL03, PL04, PL05, SE05, SE11, SE12, SE14, SI08
TNH4	42	AT02, CH02, CH05, CZ01, CZ03, DK03, DK05, DK08, DK31, ES01, ES07, ES08, ES09, ES10, ES11, ES12, ES13, ES14, ES16, ES17, FI09, FI17, FI36, FR09, FR13, FR15, HU02, IE01, IT01, LT15, LV10, LV16, MD13, PL02, PL03, PL04, PL05, SE05, SE11, SE12, SE14, SI08
HNO3	19	AM01, AT02, GB48, HU02, IT01, MD13, NL11, NO01, NO15, NO39, NO42, NO55, NO56, PL05, SK02, SK06, GB48, NL11
NH3	20	AM01, AT02, DK03, DK05, DK08, DK31, GB48, HU02, IT01, MD13, NL07, NL11, NL91, PL05, SK06, GB48, NL07, NL11, NL91

(a) Sulphate and corrected sulphate (to account for the sea salt contributions) concentrations were used for the comparisons

Figure 4 presents the annual averaged PM_{10} and $PM_{2.5}$ concentrations simulated by the models for 2009. The corresponding observations are reported in the figure with coloured circles with the same

[16] EBAS is a database hosting observation data of atmospheric chemical composition and physical properties. EBAS hosts data submitted by data originators in support of a number of national and international programs ranging from monitoring activities to research projects. EBAS is developed and operated by the Norwegian Institute for Air Research (NILU). For a complete list of programmes and projects for which EBAS serves as a database, please consult the information box in the Framework filter of the web interface (http://ebas.nilu.no/)

colour scale. The spatial pattern between CHIMERE and EMEP are quite similar both for the PM_{10} and the $PM_{2.5}$. Only the south of the domain differs because of different "dust" boundary conditions. LOTOS-EUROS has a negative bias everywhere in Europe attributed partly to missing "dust" boundary conditions.

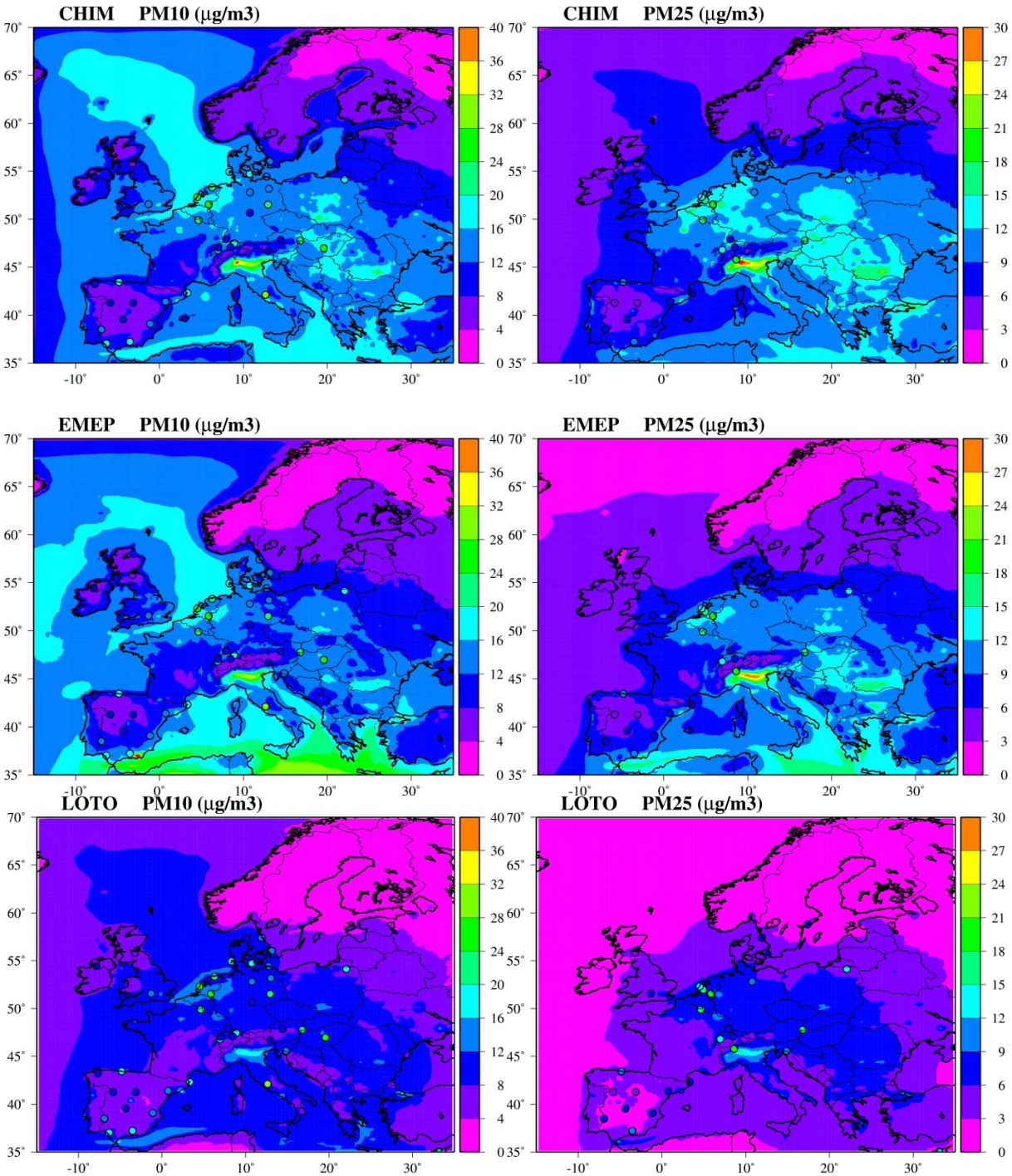


Figure 4: Average modelled PM_{10} and $PM_{2.5}$ concentrations in 2009 for EMEP, CHIMERE and LOTOS-EUROS models (coloured circles are the observations, only stations with at least 75 % of data coverage were plotted on the figures).

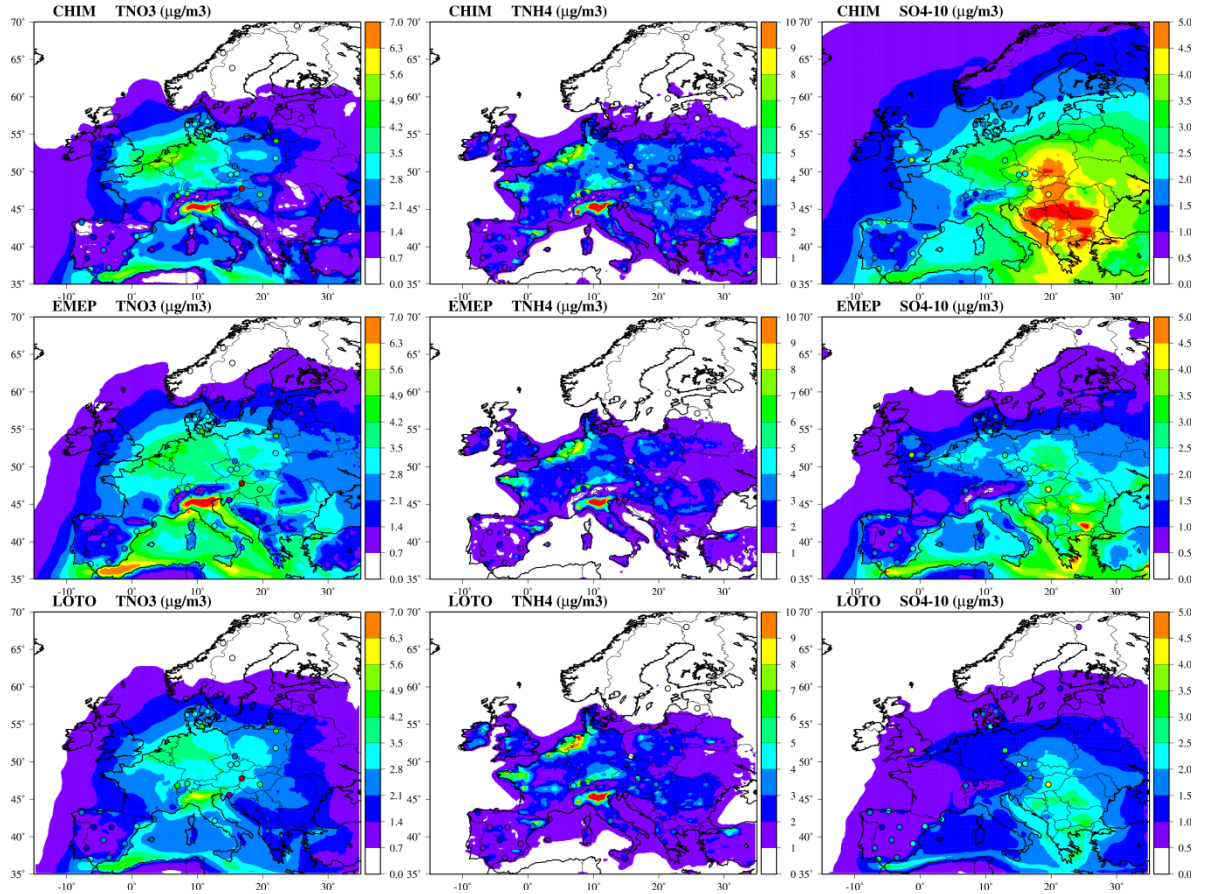


Figure 5: Average modelled Total nitrate (TNO3), Total Ammonium (TNH4) and Sulphate (SO4-10) concentrations in 2009 for EMEP, CHIMERE and LOTOS-EUROS models (coloured circles are the observations, only stations with at least 75 % of data coverage were plotted on the figures).

Figure 5 presents the annual averaged Total nitrate (TNO3), Total Ammonium (TNH4) and Sulphate (SO4-10) concentrations simulated by the models for 2009. For TNO3 and TNH4 concentrations, the models perform similarly over Europe. CHIMERE produces lower TNO3 concentrations in the south of Europe due to the missing coarse nitrate formation. LOTOS-EUROS exhibits lower TNO3 concentrations in the Pô Valley. For the sulphate, CHIMERE gives the highest concentrations on average in Europe, while LOTOS-EUROS and EMEP underestimate them. Clearly the maximum of concentrations is located in Central and Eastern Europe and over the Mediterranean Sea.

Table 9 summarizes the performances of the model in terms of error statistics as the mean bias, correlation (spatio-temporal) and the root mean square error for the PM, SIA and associated gases (NH_3 and HNO_3). Performances are rather similar between CHIMERE and EMEP models, the negative bias for PM_{10} and $PM_{2.5}$ are very close and explained by a lack of sources and/or missing processes (*e.g.* secondary organic aerosols and local dust). As previously shown LOTOS-EUROS has a strong negative bias on PM_{10} and $PM_{2.5}$. CHIMERE displays a slight positive bias on sulphate (+0.27 µg m^{-3}) whereas LOTOS-EUROS has a large negative bias (-0.95 µg m^{-3}). Statistics on nitrate and ammonium are also very close for all three models: between CHIMERE and EMEP, CHIMERE overestimate by 20% the ammonium concentrations whereas EMEP and LOTOS-EUROS underestimate by 20%, but correlation and RMSE are similar for all models. The correlation and RMSE for nitrate are similar for all three models; a negative bias, ranging from -0.80 (CHIMERE) to -0.2 (EMEP) µg m^{-3} is found. The large underestimate for CHIMERE is due to the missing formation of coarse nitrate. LOTOS-EUROS has a very low absolute bias on total ammonium (-0.02 µg m^{-3}). CHIMERE, EMEP, LOTOS-EUROS underestimate the nitrates, the largest underestimate is for CHIMERE due to the missing formation of coarse nitrate in the models.

Even if regional differences exist, the models show similar performances on SIA chemistry, the main differences from model to model are observed for sulphate concentrations.
If we consider the highest concentrations for each PM component, the sulphate concentrations are rather well reproduced by CHIMERE; LOTOS-EUROS and EMEP have a larger negative bias. For the three models the nitrate and total nitrate are largely underestimated for LOTOS-EUROS and CHIMERE, a smaller negative bias is observed for EMEP. Ammonium concentrations are also underestimated by the models. Clearly, SIA concentrations are underestimated by the models during PM pollution episodes and this contributes to the global underestimate of $PM_{2.5}$ and PM_{10} concentrations during these events. For the $PM_{2.5}$, the correlation coefficients have the same order of magnitude considering "all observations" and "the highest concentrations" for LOTOS-EUROS and CHIMERE; the EMEP model exhibits a drop of correlation coefficients for the two set of data.

Table 9: Performances of models for PM, the PM chemical composition, NH_3 and HNO_3 at EMEP sites based on daily data considering all observations (left panel) and the 10% highest observations for each site (right panel). Obs.: observations values, Cor.: Correlation coefficient, RMSE: Root mean square error, Nb: number of available daily observations. The unit are µg m^{-3} for Obs., Model values, RMSE and Biases.

All observations

Pollutant	Obs.	Model name	Model val.	Bias	Cor.	RMSE	Nb.
PM10	15.65	CHIM	12.02	-3.64	0.57	**10.15**	16405
		EMEP	11.76	-3.89	0.55	10.63	
		LOTO	8.47	-7.18	0.49	12.30	
PM25	10.71	CHIM	9.36	-1.35	0.68	**6.85**	11043
		EMEP	8.17	-2.55	0.63	7.56	
		LOTO	5.23	-5.49	0.64	9.07	
SO4-10	1.93	CHIM	2.21	0.27	0.56	1.74	9975
		EMEP	1.41	-0.53	0.59	**1.49**	
		LOTO	0.98	-0.95	0.61	1.63	
NH4-10	1.05	CHIM	1.26	0.20	0.51	1.61	9930
		EMEP	0.81	-0.25	0.50	**1.59**	
		LOTO	0.77	-0.28	0.48	1.61	
NO3-10	2.02	CHIM	1.22	-0.80	0.36	4.61	11432
		EMEP	1.84	-0.18	0.32	4.60	
		LOTO	1.50	-0.52	0.33	**4.57**	
NH3	2.09	CHIM	1.53	-0.57	0.70	2.56	3877
		EMEP	1.52	-0.58	0.71	2.52	
		LOTO	1.91	-0.18	0.71	**2.39**	
HNO3	1.05	CHIM	0.50	-0.55	0.18	1.40	3125
		EMEP	0.72	-0.32	0.21	**1.37**	
		LOTO	0.64	-0.40	0.11	1.41	
TNH4	1.92	CHIM	2.20	0.28	0.50	2.05	13354
		EMEP	1.70	-0.22	0.52	**1.88**	
		LOTO	1.90	-0.02	0.47	2.00	
TNO3	2.39	CHIM	1.52	-0.87	0.34	4.29	15261
		EMEP	2.32	-0.07	0.34	4.28	
		LOTO	1.90	-0.49	0.34	**4.22**	

10% highest concentrations for each site

Pollutant	Obs.	Model name	Model val.	Bias	Cor.	RMSE	Nb.
PM10	35.41	CHIM	19.85	-15.57	0.46	**22.39**	1724
		EMEP	21.92	-13.49	0.22	24.24	
		LOTO	13.05	-22.36	0.32	28.08	
PM25	26.47	CHIM	17.00	-9.47	0.64	**14.99**	1172
		EMEP	16.33	-10.14	0.38	17.60	
		LOTO	10.23	-16.24	0.53	20.75	
SO4-10	4.85	CHIM	4.41	-0.45	0.42	**3.09**	1025
		EMEP	2.93	-1.92	0.37	3.53	
		LOTO	2.08	-2.77	0.40	3.95	
NH4-10	3.48	CHIM	2.76	-0.72	0.32	**4.36**	1021
		EMEP	1.80	-1.68	0.31	4.60	
		LOTO	1.70	-1.78	0.30	4.66	
NO3-10	6.86	CHIM	3.46	-3.40	0.20	13.55	1188
		EMEP	4.12	-2.74	0.17	**13.40**	
		LOTO	3.34	-3.52	0.19	13.50	
NH3	5.25	CHIM	2.31	-2.93	0.71	5.64	397
		EMEP	2.26	-2.99	0.73	5.57	
		LOTO	2.75	-2.50	0.76	**4.90**	
HNO3	2.58	CHIM	0.70	-1.88	-0.04	3.13	331
		EMEP	1.02	-1.56	-0.02	**3.03**	
		LOTO	0.87	-1.71	-0.13	3.11	
TNH4	4.69	CHIM	3.40	-1.29	0.36	**4.38**	1367
		EMEP	2.72	-1.97	0.38	4.52	
		LOTO	2.89	-1.81	0.34	4.55	
TNO3	7.05	CHIM	3.53	-3.52	0.15	12.63	1571
		EMEP	4.79	-2.27	0.13	**12.38**	
		LOTO	3.73	-3.32	0.14	12.50	

7 Impact of ammonia emission reductions on exceedances of PM standards

7.1 Methodology

In order to evaluate the influence of NH_3 emission reductions on PM_{10} concentrations and exceedances descriptive statistics is applied. We use concentrations data stored in AirBase[17] and the model outputs from each scenario (2009REF, 2020GOT, 2020G10, 2020G20, 2020G30). The model outputs are interpolated to extract the modelled concentrations at the Airbase station locations. A delta of concentration at each monitoring site is then calculated, based on the difference of the modelling results from a 2020 scenario and the 2009 REF reference run. This delta is then applied to the 2009 observations to get a better estimate of 2020 concentrations for the various emission scenarios (Figure 6). This methodology based on observations and modelling allows determining the evolution of concentrations for all stations typologies.

[17] AirBase is the public air quality database system of the EEA. It contains air quality monitoring data and information submitted by the participating countries throughout Europe. http://acm.eionet.europa.eu/databases/airbase

Step 1: Observational data	Step 2 : model simulation	Step 3		Step 4
AirBase observations 2009	Output model 2009REF	Interpolation on AirBase stations	C_k^{REF}	
	Output model 2020GOT		C_k^{GOT}	$C_k + (C_k^{GOT} - C_k^{REF})$
C_k concentration at site k	Output model 2020G10		C_k^{G10}	$C_k + (C_k^{G10} - C_k^{REF})$
	Output model 2020G20		C_k^{G20}	$C_k + (C_k^{G20} - C_k^{REF})$
	Output model 2020G30		C_k^{G30}	$C_k + (C_k^{G30} - C_k^{REF})$

Figure 6: Steps of the calculation of future PM concentrations at the AirBase stations location for the scenario analyses

We focus on the impact of NH_3 reduction scenario on PM_{10} and $PM_{2.5}$. Directive 2008/50/EC specifies that the annual mean must not exceed 20 µg.m^{-3} in case of $PM_{2.5}$ (stage 2 indicative limit value in 2020), and 40 µg m^{-3} in case of PM_{10}. A second limit value is defined for PM_{10}: the number of days that exceed the daily limit value, fixed at 50 µg m^{-3}, must not exceed 35. These limit values will be used as indicators to quantify the impact of scenarios. It is noticeable that, according to Directive 2008/50/EC, only stations with a data capture larger than 90% are used. Since this percentage must not include losses of data due to the regular calibration or the normal maintenance of the instrumentation, an adjustment is necessary: it is decreased at 85% according to recent discussions from IPR guidance[18]. Indeed, according to the "Guidance on the Annexes to Decision 97/101/EC[19] on Exchange of Information as revised by Decision 2001/752/EC[20]", 5% is a good general approximation of the proportion of measurement time in a calendar year dedicated to planned equipment maintenance and calibration.

7.2 Impact of scenarios on the number of stations in exceedance

Annex 3, 4 and 6 summarize the impact of the Gothenburg Protocol and further reductions in NH_3 emissions respectively on the number of stations exceeding the daily limit value for PM_{10} more than 35 days, the yearly mean limit value on PM_{10} and the yearly mean limit value on $PM_{2.5}$. Annex 5 and Annex 7 report respectively the evolution of annual average PM_{10} and $PM_{2.5}$ concentrations at stations in relation to their limit values. Table 10, Table 11, Table 12 and Table 13 summarize the effect of each scenario on PM_{10} and $PM_{2.5}$ exceedances. Table 11, Table 12 and Table 13 break down the numbers reported in Table 10 by EU countries.

The response obtained with EMEP and CHIMERE are quite similar, whereas LOTOS-EUROS results show the same kind of responses as with these models, but with a lower magnitude. The results show a clear response of Gothenburg 2020 protocol emission reductions on the number of PM_{10} stations in exceedance. On average in EU27 the number of stations exceeding the number of exceedance days of PM_{10} limit value is reduced by 21.4%, 20.6% and 12.4% respectively for CHIMERE, EMEP and LOTOS-EUROS with the Gothenburg Protocol scenario. This reduction in EU27 is mainly driven by the decrease of exceeding stations in Italy, Poland and Germany (Table 11).

[18] COMMISSION IMPLEMENTING DECISION of 12 December 2011 laying down rules for Directives 2004/107/EC and 2008/50/EC of the European Parliament and of the Council as regards the reciprocal exchange of information and reporting on ambient air quality (notified under document (2011/850/EU).

[19] COUNCIL DECISION of 27 January 1997 establishing a reciprocal exchange of information and data from networks and individual stations measuring ambient air pollution within the Member States (97/101/EC)

[20] COMMISSION DECISION of 17 October 2001 amending the Annexes to Council Decision 97/101/EC establishing a reciprocal exchange of information and data from networks and individual stations measuring ambient air pollution within the Member States (2001/752/EC).

It is noteworthy that an additional 10% or 20% agriculture emissions reduction for NH_3 does have a smaller impact in most regions on the number of stations in the exceedance of the PM_{10} daily limit value (compared to the number of exceedances under the Gothenburg protocol scenario) than the 30% emission reduction. 10%, 20%, 30% emission reductions respectively give a decrease of 10, 21 and 40 exceeding stations for CHIMERE; 8, 24 and 40 exceeding stations for EMEP; 5, 14 and 23 for LOTOS-EUROS, compared to the Gothenburg protocol (Table 10). For the 30% NH_3 emission reduction scenario (2020G30), most of those stations no longer exceeding the daily limit values are urban, suburban or traffic stations: 30, 31 and 21 stations respectively for CHIMERE, EMEP and LOTOS-EUROS models. As summarized in Table 10, the Gothenburg protocol scenario reduced by 30%, 28% and 19% the number of stations exceeding the PM_{10} annual limit value respectively for CHIMERE, EMEP and LOTOS-EUROS. For the $PM_{2.5}$ annual limit value, the Gothenburg protocol scenario reduced by 35%, 30% and 26% the number of exceeding stations respectively for CHIMERE, EMEP and LOTOS-EUROS. The decrease is slightly higher for the $PM_{2.5}$ limit value because most of the impact of ammonia emission reductions is concentred in the fine fraction of PM.

The additional G10 scenario does not bring any significant effect on the number of exceeding stations; additional emission reduction (G20 and G30) seems to be more efficient. Clearly, we observe a non-linear effect in the reduction of concentrations or the number of stations exceeding the thresholds values; the reduction of ammonia emissions looks more and more efficient from G10 to G30 scenarios for CHIMERE. For LOTOS-EUROS and EMEP, the additional decrease of exceedances of the daily limit value is the same for G20 and G30.

Table 10: Influence of the Gothenburg Protocol and further NH_3 emission reductions in the number of stations in exceedance of the PM limit values in the EU27, aggregated by station typology. Impact of scenarios for the different models.

Number of stations in exceedances		OBS [(a)]	Pvalid[(b)]	CHIMERE				EMEP				LOTOS-EUROS			
				GOT	*G10*	*G20*	*G30*	*GOT*	*G10*	*G20*	*G30*	*GOT*	*G10*	*G20*	*G30*
PM_{10} limit value : 35 days > 50µg.m^{-3}	*rural*	16	5.9%	10	9	9	8	9	9	7	7	14	14	13	13
	suburban	65	19.7%	53	51	49	49	51	50	48	48	59	58	56	54
	urban	267	27.3%	203	198	192	183	210	204	197	189	228	225	221	220
	traffic	175	20.3%	142	140	138	136	143	142	141	136	157	157	155	149
	industrial	89	19.0%	73	73	72	65	73	73	69	66	78	77	77	77
	Total	**612**	**21.0%**	**481**	**471**	**460**	**441**	**486**	**478**	**462**	**446**	**536**	**531**	**522**	**513**
PM_{10} limit value : annual mean > 40 µg.m^{-3}	*rural*	3	1.4%	2	2	2	2	2	2	2	2	3	3	3	3
	suburban	13	4.9%	11	11	11	11	11	11	11	11	11	11	11	11
	urban	69	8.9%	50	50	49	48	51	50	50	49	59	57	56	56
	traffic	43	6.4%	27	27	27	27	29	28	28	26	31	30	29	29
	industrial	13	4.0%	8	8	8	8	8	8	8	8	10	9	9	9
	Total	**141**	**6.26%**	**98**	**98**	**97**	**96**	**101**	**99**	**99**	**96**	**114**	**110**	**108**	**108**
$PM_{2.5}$ limit value : annual mean > 20 µg.m^{-3}	*rural*	7	10.4%	3	3	3	3	3	2	2	2	5	5	3	3
	suburban	12	22.6%	8	8	8	7	8	8	8	8	8	8	8	8
	urban	48	22.6%	30	29	28	27	34	33	31	28	36	36	36	35
	traffic	26	21.3%	18	18	18	18	19	19	19	19	20	20	20	20
	industrial	12	25.0%	9	9	8	8	9	9	9	8	9	9	9	9
	Total	**105**	**20.9%**	**68**	**67**	**65**	**63**	**73**	**71**	**69**	**65**	**78**	**78**	**76**	**75**

(a) OBS are the observed exceedances from AirBase

(b) P_{EV} is the percentage of exceeding stations among the valid stations (only stations with a data capture larger than 85% are used in calculations).

Table 11 : Influence of the Gothenburg Protocol and further NH_3 emission reductions in the number of stations (all typology) with more than 35 days the PM_{10} daily limit values in exceedance for EU27 countries. Impact of scenarios for the different models.

Number of stations in exceedances		OBS	CHIMERE				EMEP				LOTOS-EUROS			
			GOT	G10	G20	G30	GOT	G10	G20	G30	GOT	G10	G20	G30
PM_{10} limit value : 35 days > 50µg.m^{-3}	*AT*	7	3	3	3	3	5	4	3	3	4	4	3	3
	BE	19	17	13	10	9	14	13	9	9	18	17	16	14
	BG	37	37	37	37	37	37	37	37	37	37	37	37	37
	CY	1	1	1	1	1	1	1	1	1	1	1	1	1
	CZ	37	32	32	31	31	32	32	31	29	33	32	32	32
	DE	35	17	17	16	13	17	17	16	15	23	23	23	21
	DK	0	0	0	0	0	0	0	0	0	0	0	0	0
	EE	0	0	0	0	0	0	0	0	0	0	0	0	0
	ES	47	46	46	44	42	43	42	39	38	46	45	45	45
	FI	0	0	0	0	0	0	0	0	0	0	0	0	0
	FR	39	22	22	22	19	23	23	22	20	28	27	26	24
	GB	2	0	0	0	0	0	0	0	0	0	0	0	0
	GR	7	7	7	7	7	7	7	7	7	7	7	7	7
	HU	9	7	7	7	7	7	7	6	6	7	7	7	7
	IE	0	0	0	0	0	0	0	0	0	0	0	0	0
	IT	195	151	149	147	144	156	155	152	148	172	172	169	168
	LT	1	1	1	1	0	0	0	0	0	1	1	1	1
	LU	0	0	0	0	0	0	0	0	0	0	0	0	0
	LV	3	2	2	2	2	3	3	3	3	3	3	3	3
	MT	1	1	1	1	1	1	1	1	1	1	1	1	1
	NL	1	0	0	0	0	0	0	0	0	0	0	0	0
	PL	124	103	99	97	94	107	105	104	99	115	114	111	110
	PT	12	6	6	6	6	7	6	6	6	10	10	10	10
	RO	16	14	14	14	13	12	12	12	12	15	15	15	15
	SE	4	4	4	4	4	4	4	4	4	4	4	4	4
	SI	3	1	1	1	1	1	1	1	1	1	1	1	1
	SK	12	9	9	9	7	9	8	8	7	10	10	10	9
	EU27	**612**	**481**	**471**	**460**	**441**	**486**	**478**	**462**	**446**	**536**	**531**	**522**	**513**

Table 12 : Influence of the Gothenburg Protocol and further NH_3 emission reductions in the number of stations per country (all typology) in exceedance of the PM_{10} yearly mean limit values for EU27 countries. Impact of scenarios for the different models.

Number of stations in exceedances		OBS	CHIMERE				EMEP				LOTOS-EUROS			
			GOT	G10	G20	G30	GOT	G10	G20	G30	GOT	G10	G20	G30
PM_{10} limit value : annual mean > 40 µg.m^{-3}	*AT*	0	0	0	0	0	0	0	0	0	0	0	0	0
	BE	0	0	0	0	0	0	0	0	0	0	0	0	0
	BG	24	23	23	23	23	23	23	23	22	23	23	23	23
	CY	0	0	0	0	0	0	0	0	0	0	0	0	0
	CZ	10	8	8	8	8	8	8	8	8	8	8	8	8
	DE	1	1	1	1	1	1	1	1	1	1	1	1	1
	DK	0	0	0	0	0	0	0	0	0	0	0	0	0
	EE	0	0	0	0	0	0	0	0	0	0	0	0	0
	ES	7	7	7	7	7	7	7	7	6	7	7	7	7
	FI	0	0	0	0	0	0	0	0	0	0	0	0	0
	FR	7	2	2	2	2	4	4	4	4	4	4	4	4
	GB	0	0	0	0	0	0	0	0	0	0	0	0	0
	GR	3	1	1	1	1	1	1	1	1	2	2	2	2
	HU	0	0	0	0	0	0	0	0	0	0	0	0	0
	IE	0	0	0	0	0	0	0	0	0	0	0	0	0
	IT	50	27	27	27	27	27	27	27	26	36	34	32	32
	LT	0	0	0	0	0	0	0	0	0	0	0	0	0
	LU	0	0	0	0	0	0	0	0	0	0	0	0	0
	LV	0	0	0	0	0	0	0	0	0	0	0	0	0
	MT	1	1	1	1	1	1	0	0	0	1	1	1	1
	NL	0	0	0	0	0	0	0	0	0	0	0	0	0
	PL	33	24	24	23	22	25	24	24	24	27	26	26	26
	PT	0	0	0	0	0	0	0	0	0	0	0	0	0
	RO	2	2	2	2	2	2	2	2	2	2	2	2	2
	SE	0	0	0	0	0	0	0	0	0	0	0	0	0
	SI	0	0	0	0	0	0	0	0	0	0	0	0	0
	SK	3	2	2	2	2	2	2	2	2	3	2	2	2
	EU27	**141**	**98**	**98**	**97**	**96**	**101**	**99**	**99**	**96**	**114**	**110**	**108**	**108**

Table 13 : Influence of the Gothenburg Protocol and further NH_3 emission reductions in the number of stations per country (all typology) in exceedance of the $PM_{2.5}$ yearly mean limit values for EU27 countries. Impact of scenarios for the different models.

Number of stations in exceedances		OBS	CHIMERE				EMEP				LOTOS-EUROS			
			GOT	G10	G20	G30	GOT	G10	G20	G30	GOT	G10	G20	G30
$PM_{2.5}$ limit value : annual mean > 20 µg.m^{-3}	*AT*	4	2	2	2	2	2	2	2	2	2	2	2	2
	BE	10	7	7	6	4	7	7	7	5	7	7	7	7
	BG	4	4	4	4	4	4	4	4	4	4	4	4	4
	CY	0	0	0	0	0	0	0	0	0	0	0	0	0
	CZ	11	8	8	8	8	8	8	8	8	8	8	8	8
	DE	4	3	2	2	2	3	3	3	2	3	3	3	3
	DK	0	0	0	0	0	0	0	0	0	0	0	0	0
	EE	0	0	0	0	0	0	0	0	0	0	0	0	0
	ES	6	5	5	5	5	5	5	5	5	5	5	5	5
	FI	0	0	0	0	0	0	0	0	0	0	0	0	0
	FR	15	7	7	7	7	10	10	9	9	11	11	11	10
	GB	0	0	0	0	0	0	0	0	0	0	0	0	0
	GR	2	0	0	0	0	0	0	0	0	1	1	1	1
	HU	2	1	1	1	1	1	1	1	1	1	1	1	1
	IE	0	0	0	0	0	0	0	0	0	0	0	0	0
	IT	31	18	18	18	18	19	18	17	17	22	22	20	20
	LT	0	0	0	0	0	0	0	0	0	0	0	0	0
	LU	0	0	0	0	0	0	0	0	0	0	0	0	0
	LV	0	0	0	0	0	0	0	0	0	0	0	0	0
	MT	0	0	0	0	0	0	0	0	0	0	0	0	0
	NL	0	0	0	0	0	0	0	0	0	0	0	0	0
	PL	9	9	9	9	9	9	9	9	9	9	9	9	9
	PT	0	0	0	0	0	0	0	0	0	0	0	0	0
	RO	2	0	0	0	0	1	0	0	0	1	1	1	1
	SE	0	0	0	0	0	0	0	0	0	0	0	0	0
	SI	2	1	1	0	0	1	1	1	0	1	1	1	1
	SK	3	3	3	3	3	3	3	3	3	3	3	3	3
	EU27	**105**	**68**	**67**	**65**	**63**	**73**	**71**	**69**	**65**	**78**	**78**	**76**	**75**

7.3 Linearity of scenarios

In order to test the linearity of additional ammonia emission reductions on $PM_{2.5}$ and PM_{10} concentrations, we compare the impact on $PM_{2.5}$ and PM_{10} concentrations of additional 10% reduction of NH_3 emissions for the three scenarios 2020G10, 2020G20 and 2020G30 at urban AirBase stations (Figure 7). Actually, the -20% and -30% scenarios are compared to the -10 % case. In case of linearity the -20% and -30% scenarios would give respectively 2 and 3 times the impact of the -10% scenario (solid black line in Figure 7). However, the linearity of scenarios is controlled by the chemical regime of aerosols.

We observe that CHIMERE, EMEP and LOTOS-EUROS are over the "ideal linearity" ratio in rural sites and this over linearity is enhanced in the 2020G30 scenario. This means that the ammonia is certainly in excess in the atmosphere but this excess of ammonia is less important in 2020G20 and 2020G30, implying a more efficiency of more ambitious NH_3 emission scenarios. The linearity ratios are quite close among the models, with CHIMERE being in between EMEP and LOTOS-EUROS.

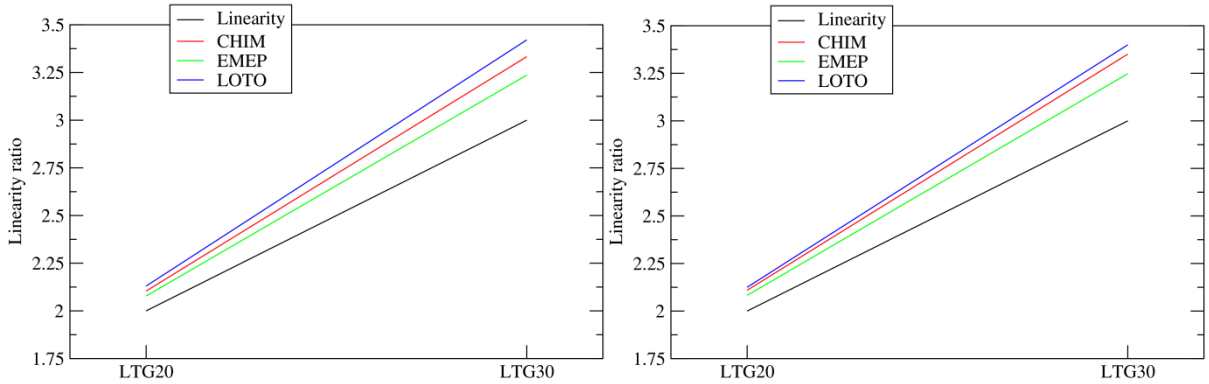


Figure 7: Test of linearity (LT) for 2020G20 and 2020G30 scenarios to test the linearity of additional emission reductions on $PM_{2.5}$ (right) and PM_{10} (left) concentrations at urban AIRBASE stations. For LTG20, $Linerity\ ratio_{G20} = \frac{(G20-GOT)}{(G10-GOT)}$ and for LTG30, $Linearity\ ratio_{G30} = \frac{(G30-GOT)}{(G10-GOT)}$. The solid black line corresponds to the ideal case of linearity.

Figure 8 illustrates the spatial variations of the linearity ratio on PM_{10} for the three models. For 2020G20 the ratios are slightly above 2 in most of continental regions in the three models. LOTOS-EUROS ratios are slightly higher from France to the Eastern Europe. CHIMERE displays a noisier pattern in the South East of the domain as well as EMEP displaying sharp gradients of linearity in the north and the south of the domain. The linear ratio reaches 3.2 to 3.6 in countries located in the centre of the domain for the scenario 2020G30.

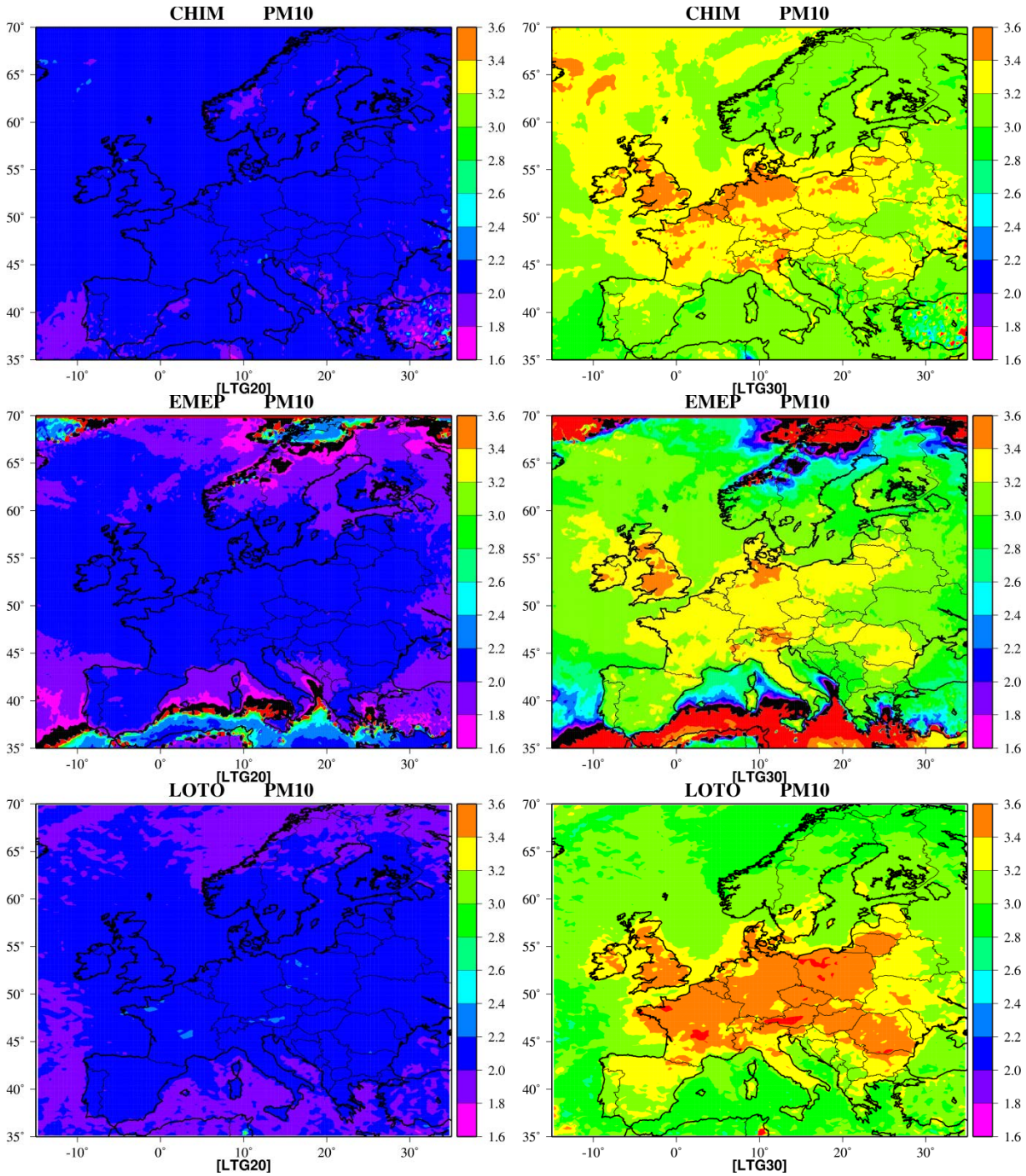


Figure 8: Maps of averaged linearity ratio on PM_{10} for 2020G20 and 2020G30 scenarios for the three models (black colour for values < 1.6 and red colour for values > 3.6). An ideal linear behaviour would lead to a ratio of 2 in 2020G20 and 3 in 2020G30.

On average for all stations in Europe, there is a clear link between the number of days in exceeding the daily limit value of 50 µg m^{-3} and the yearly mean concentrations for the PM10 (Figure 9, left panel). The slope of the linear regression (in red on the graph) is 3.96 (R^2=0.88 for the regression) meaning that on average, **for a given station a decrease of 1 µg m^{-3} of the yearly mean PM_{10} concentration leads to about 4 exceedances less of the daily limit value for PM_{10}.**

In Figure 9 (right panel), the ratio between the delta of number PM_{10} daily exceedances and the delta of the PM_{10} yearly mean averaged for all stations in Europe is displayed. This ratio is calculated as: $\frac{[Nb.of\ exceedances]_{scenario}-[Nb.of\ exceedances]_{GOT}}{[Yearly\ mean]_{scenario}-[Yearly\ mean]_{GOT}}$ with scenario=G10, G20 or G30, the reference is 2020GOT scenario in the calculation.

This ratio slightly increases from G10 to G30 for all models, for CHIMERE and EMEP the ratio is close to 2 and close to 1.6 for LOTOS-EUROS. If the models had this ability to capture the amplitude of episodes (and then exceedances), this ratio would have been close to 4 assuming a similar chemical regime in 2009 and 2020. However, as it has been shown in section 6 devoted to the model evaluation, all models underestimate the PM peaks with an underestimate of SIA concentrations while the average concentration of SIA is correctly simulated. **This statement explains why this ratio is smaller than 4 and means that the impact of ammonia emission reduction scenarios on the number of daily exceedances is certainly underestimated by the models.** However, we calculated this ratio with the scenario 2020GOT and then under a different chemical regime to that of the "real world" 2009, then the comparison cannot be quantitative and uncontroversial.

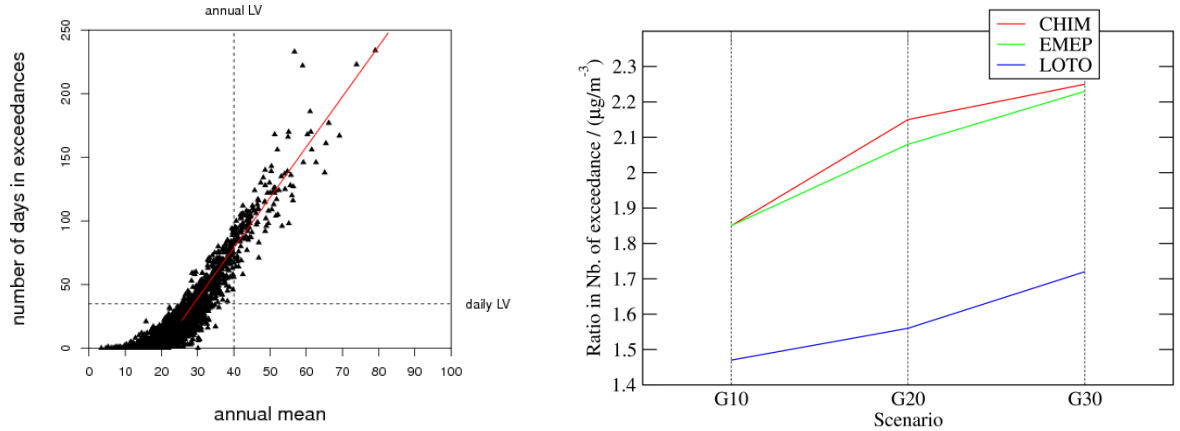


Figure 9: *Left panel*: number of daily exceedances of PM_{10} *versus* PM_{10} yearly mean concentrations for each station in Europe for 2009 (one triangle symbol by station). *Right panel*: evolution of the ratio between the delta of number PM_{10} daily exceedances and the delta of the PM_{10} yearly mean averaged for all stations in Europe for the three scenarios G10, G20 and G30, the delta is calculated between the given scenario and 2020GOT as $\frac{[Nb.of\ exceedances]_{scenario}-[Nb.of\ exceedances]_{GOT}}{[Yearly\ mean]_{scenario}-[Yearly\ mean]_{GOT}}$.

7.4 Spatial patterns of annual PM concentrations reductions

Figure 10 displays the concentration decline throughout Europe between the 2009 situation and the expected situation with Gothenburg protocol emissions. The response is clearly the same between CHIMERE and EMEP, CHIMERE gives a slightly higher decrease particularly over the Paris basin, Lyon, Sofia, Budapest, Zurich, Rome, and Milan. In Spain and the Netherlands, the decrease is very low. In these countries, the small decrease of concentrations is mainly attributed to low evolution of emission between 2009 and 2020. Higher decreases are observed for EMEP in Greece. LOTOS-EUROS displays a lower decrease of PM_{10} concentrations everywhere in Europe, particularly over the north of Italy and eastern Europe.

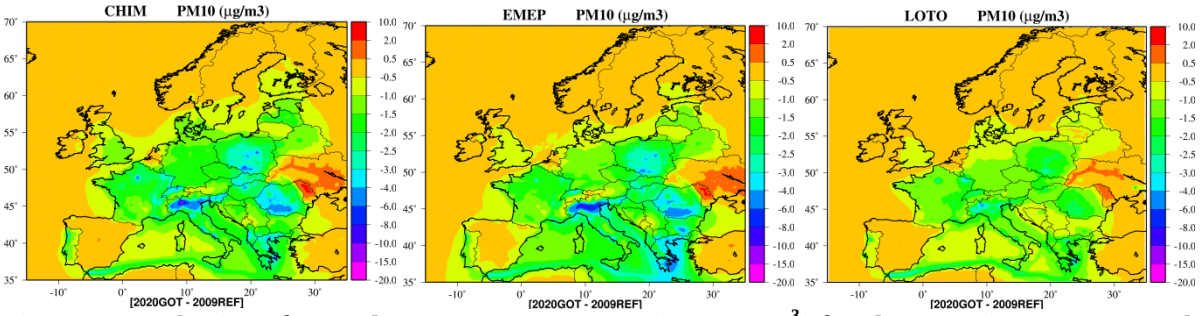


Figure 10: Reduction of annual mean PM_{10} concentrations (µg m^{-3}) for the scenario 2020GOT: Delta between scenario and reference (2009REF) for the three models

An additional emission reduction of NH_3 has an impact in a large region from the north of France and south East of England to Poland and Romania (Figure 11). The Pô Valley area is also influenced by ammonia emission decreases. The highest decreases are simulated in Benelux, western part of Germany, northern part of France and north Italy. CHIMERE and EMEP behave similarly whereas LOTOS-EUROS gives a lower impact of ammonia emission reductions. The expected ammonia emission reduction in Brittany (West part of France) has a weak local impact. This region only affects over lands concentrations under westerly winds that are usually linked to low pollution conditions. As previously mentioned, LOTOS-EUROS underestimates sulphate concentrations in the 2009REF simulation. This could lead to a higher availability of NH_3 in LOTOS-EUROS implying a lower efficiency of ammonia emission reductions.

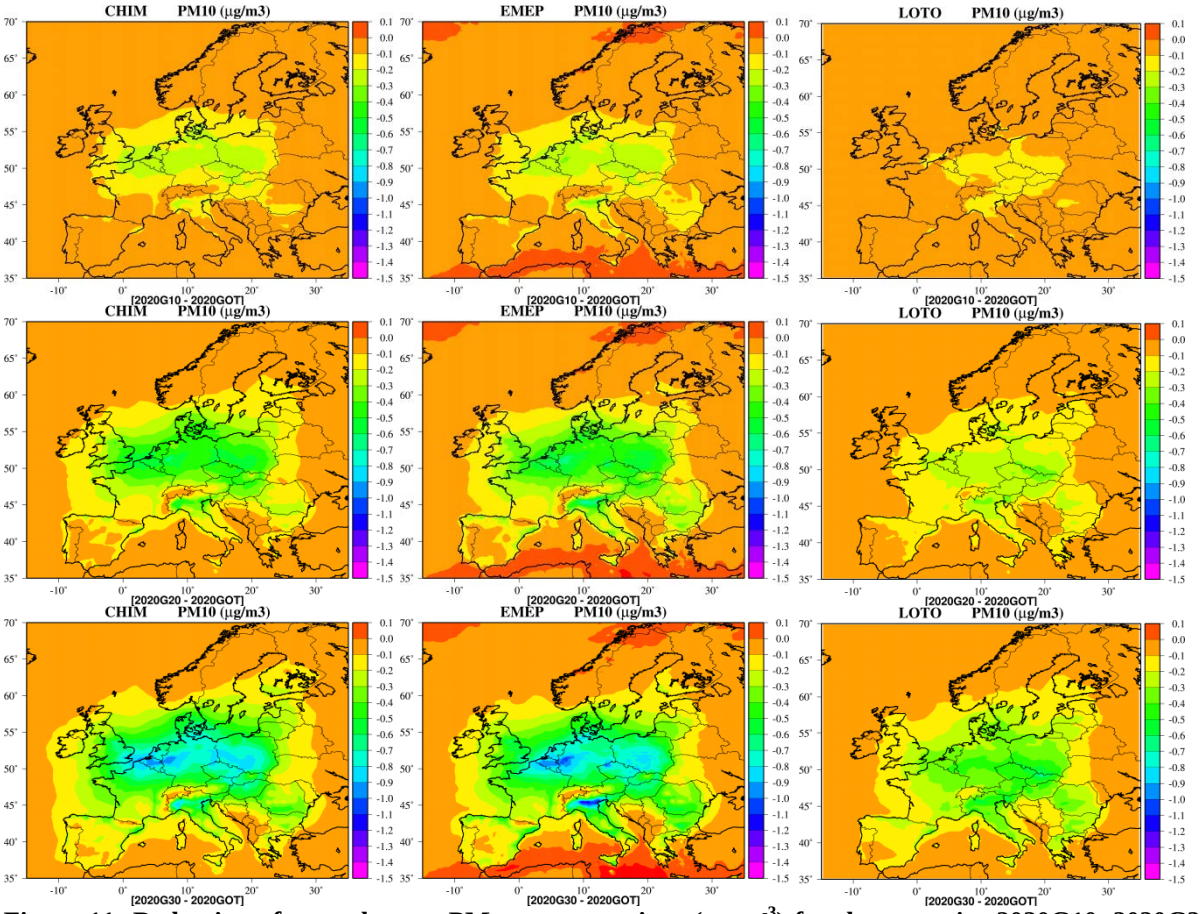


Figure 11: Reduction of annual mean PM_{10} concentrations (µg m^{-3}) for the scenarios 2020G10, 2020G20 and 2020G30: Delta between scenario and 2020GOT for the three models.

Figure 12 displays the annual mean concentration reduction expected with the scenario 2020G30 in percent points for PM_{10} and $PM_{2.5}$ compared to the 2020GOT scenario. The spatial patterns are

similar between CHIMERE and EMEP with a stronger reduction for EMEP. The most important reductions in % are observed over the south of England, north of France, Benelux, Germany, Czech Republic and Poland for CHIMERE and EMEP; they reach 10% for EMEP, 8% for CHIMERE in these regions for the $PM_{2.5}$ concentrations. For LOTOS-EUROS the spatial pattern of reductions is more spread over Europe with highest reductions over Poland and Sweden for the $PM_{2.5}$ concentrations. The differences of spatial patterns between LOTOS-EUROS and CHIMERE / EMEP are also observed for the SIA fraction. In the south of England the range of reduction lies from 6 to 12% from LOTOS-EUROS to CHIMERE close to the values reported by Harrison et al. (2013) with an expected reduction of 9% in a rural site in the south of England with a 30% cut of ammonia emissions in Europe.

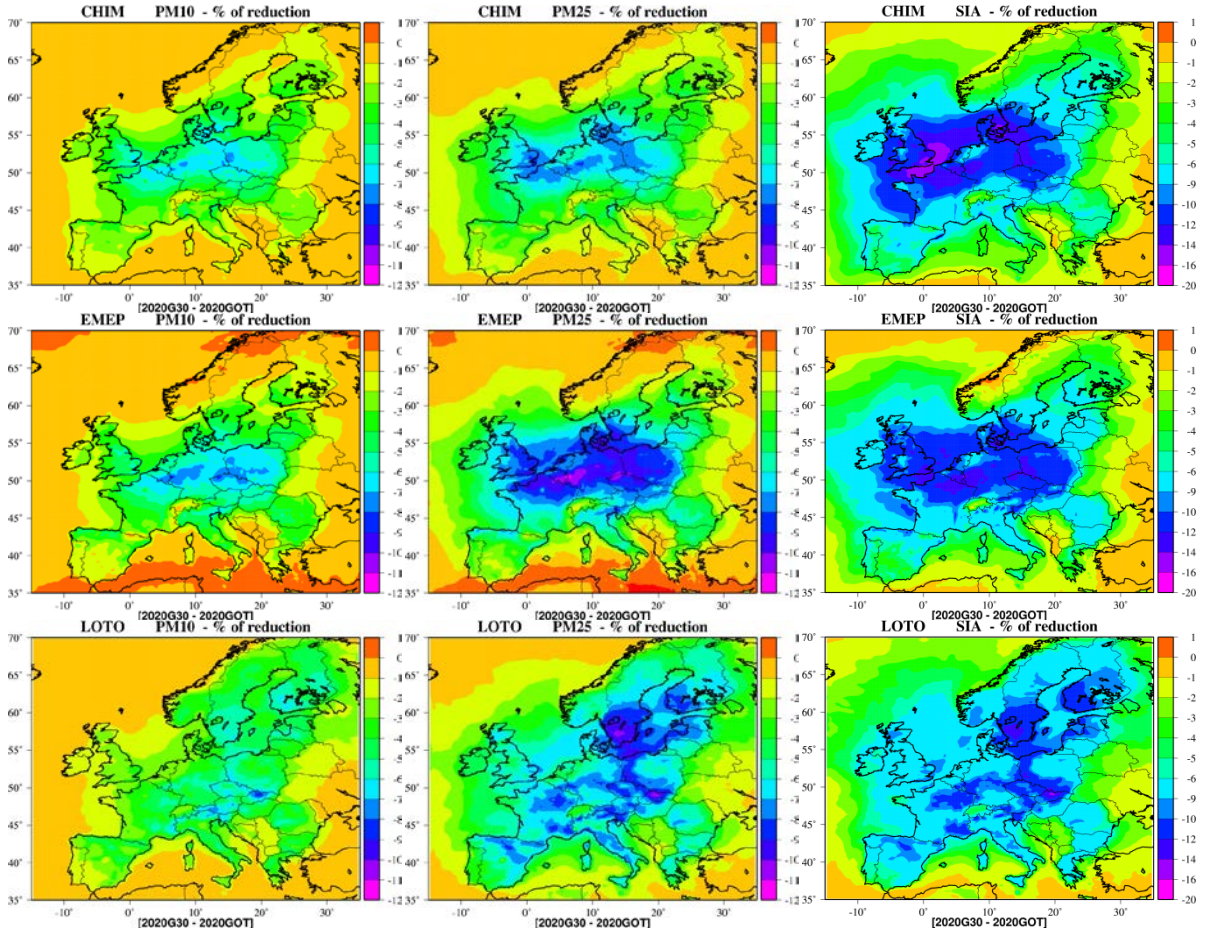


Figure 12: Reduction of annual mean PM_{10}, $PM_{2.5}$ and SIA concentrations (in %) for the scenario 2020G30 compared to 2020GOT emission scenario for the three models

Figure 13 represents the average concentrations of free ammonia (*F-NH_X*) for the scenario 2020GOT. The free ammonia is defined as the difference in molar concentrations of the total ammonium and of sulphate concentrations as follows: $F\text{-}NH_x = TNH4 - 2 \times SO_4^{2-}$. The free ammonia is the amount of ammonia available, after neutralizing sulphate, for ammonium nitrate formation mainly. There is clearly an excess of ammonia in Europe in the scenario 2020GOT, with the highest concentrations of free ammonia over the emitting areas. To complete the analysis the G_{ratio} is used (Ansari and Pandis, 1998 in Pay et al., 2012). This ratio indicates whether fine-particle nitrate formation is limited by the availability of HNO_3 or NH_3. All the terms in the following equation are expressed on a molar basis : $G_{ratio} = \frac{F\text{-}NH_x}{TNO3}$:

- G_{ratio} > 1 indicates that nitric acid is limiting,
- G_{ratio} < 0 indicates the ammonia is severely limiting,

- G_{ratio} between 0 and 1 indicates ammonia is available for reaction with nitric acid, but ammonia is the limiting species.

The maps of G_{ratio} in Figure 13 show that the three models simulate a global limitation by the nitric acid with severe limitations over ammonia hot spot emission areas. The models predict lower G_{ratio} in some countries in Central Europe. These maps indicate a general behaviour, even in region with G_{ratio} > 1, ammonia emission reductions have a significant impact because this ratio is strongly time dependent and can often decrease below 1. A close look at the spatial patterns of G_{ratio} in Figure 13 shows that often for models, the maximum effect of ammonia emission reductions shown in Figure 12 is located outside the highest G_{ratio}.

It is noticeable that LOTOS-EUROS displays slightly higher free ammonia concentrations and higher G_{ratio} on average in Europe, explaining less sensitivity of LOTOS-EUROS to ammonia emission reductions. LOTOS-EUROS is the only model that takes a compensation point into account in the deposition process (see Table 7). Depending on the ambient NH_3 concentration and the NH_x concentration in soil/vegetation there might be an upward flux (secondary emission) instead of deposition. Such a situation occurs in the high density emission regions. This process is not included in CHIMERE and EMEP which might partly explain the negative bias in NH_3 (caused by over estimating the deposition flux). Due to this additional feedback, LOTOS-EUROS results might be less sensitive for NH_3 reductions.

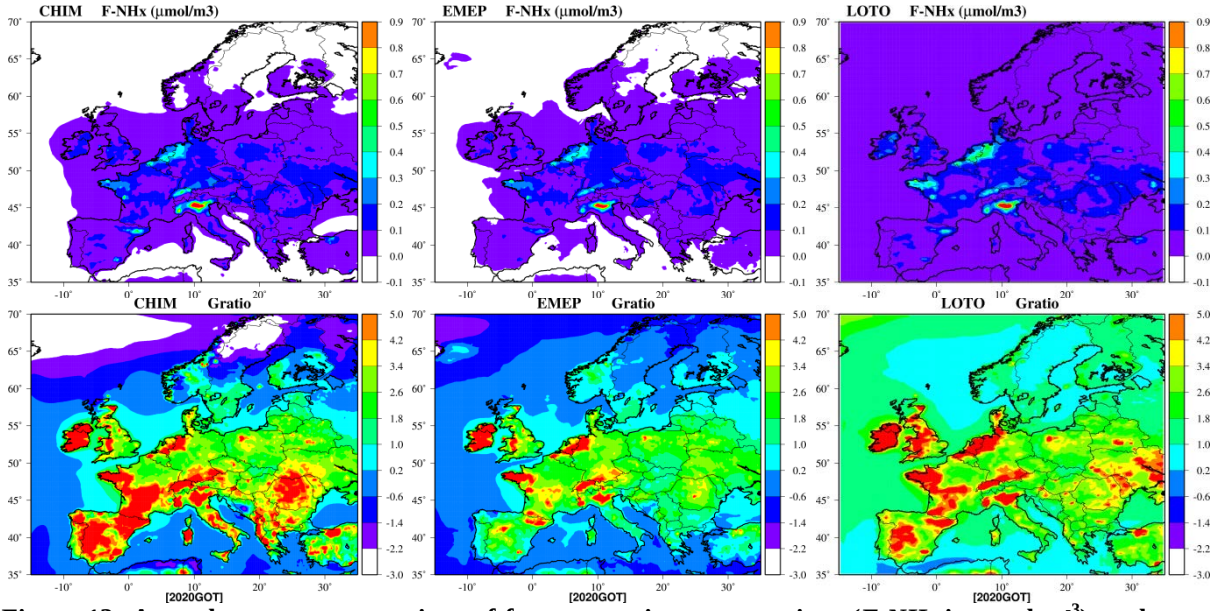


Figure 13: Annual mean concentrations of free ammonia concentrations ($F\text{-}NH_x$ in µmol m^{-3}) at the top and the annual mean G_{ratio} at the bottom for the three models in the scenario 2020GOT (red color for values above the scale maximum).

8 Conclusions

Three air quality models were used to assess the ammonia emission reduction scenarios on PM_{10} and $PM_{2.5}$ air quality standards. In term of chemical regime, even if nitric acid is known to be limiting on average in Europe, most of scenario modelling studies have shown a large impact of ammonia emission reductions on PM concentrations.

The simulations were performed on the meteorological year 2009. Five simulations were performed: one reference with 2009 emissions, a Gothenburg protocol scenario (horizon 2020), and three more stringent scenarios on ammonia emissions with 10%, 20% and 30% ammonia emission reduction (applied to the Gothenburg emission scenario). First, the models were evaluated against observational data using the reference 2009 simulation. On SIA components, the models area able to reproduce annual means concentrations with a small bias. CHIMERE has a negative bias due to the missing formation of coarse nitrate, LOTOS-EUROS and EMEP underestimate the sulphate concentrations. Regarding the highest concentrations, a large bias is observed for all models for nitrate and ammonium, these species are important contributors to PM concentrations during episodes.

Usually LOTOS-EUROS gives lower decrease of PM concentrations when applying all type of scenario. The reasons are difficult to explain because all models ran with different boundary conditions and also the model design is different. The model evaluation points out a large underestimate of LOTOS-EUROS on the sulphate concentrations. In addition, LOTOS-EUROS uses a compensation points for the calculation of ammonia emission fluxes. This implies a different chemical regime that could explain a lower sensitivity to ammonia emission reductions for LOTOS-EUROS. The most important reduction of the number of stations in exceedance is expected in Italy and Poland because the initial number of exceeding stations is important, which is high at these countries. The Benelux, West of Germany and North of Italy exhibit the highest concentration decreases expected due to ammonia emission reductions.

In 2009, based on our calculation with AirBase data, 612 monitoring stations exceeded the PM_{10} daily limit value of 50 µg m^{-3} more than 35 days. According to the models, the number of remaining stations exceeding the PM_{10} daily standards after the implementation of the Gothenburg protocol in 2020 is 481, 486 and 536 respectively for CHIMERE, EMEP and LOTOS-EUROS. This indicates that the Gothenburg protocol alone may contribute to a 12 to 21% reduction the exceedances of the PM_{10} daily LV and further measures must be considered to reduce PM levels across Europe. A 30% reduction of ammonia emissions in addition to the Gothenburg protocol scenario enhances the reduction of the number of exceeding station. From the Gothenburg Protocol scenario, this reduction reaches 40, 40 and 23 stations respectively for CHIMERE, EMEP and LOTOS-EUROS models. Most of those stations no longer exceeding the daily limit values are urban, suburban or traffic stations: 30, 31 and 21 stations respectively for CHIMERE, EMEP and LOTOS-EUROS models. An analysis carried out on scenario impacts shows that the model response on daily exceedances might be underestimated because models tend to underestimate the high concentrations of PM components and particularly the nitrate and ammonium concentrations.

The simulated reduction of annual mean $PM_{2.5}$ concentration, due to a further reduction of 30% NH_3 emissions from agriculture compared to the Gothenburg scenario, indicate that the reduction may reach 8% (CHIMERE) to 10% (EMEP) in some areas. The most important reductions in % are observed over the south of England, north of France, Benelux, Germany, Czech Republic and Poland for CHIMERE and EMEP; for LOTOS-EUROS the spatial pattern of reductions is more spread over Europe with highest reductions over Poland and Sweden for the $PM_{2.5}$ concentrations. The percentage

reduction of the PM_{10} annual mean concentrations over Europe are lower than for $PM_{2.5}$, as expected, ranging from 3 to 8% over most of Europe.

This study shows that the implementation of the Gothenburg protocol (decrease in $PM_{2.5}$, NO_x, SO_x, NMVOC and NH_3) will be an important step towards compliance with PM limit values over Europe, but is far from assuring compliance in 2020. Further measures to reduce PM levels across Europe must therefore be considered. Further reductions of NH_3 emissions from agriculture are feasible and are likely to give an important contribution to reduce PM levels are exceedances in Europe. The linearity study presented in this report shows that an increase of ammonia emission reductions higher than 10% enhances the efficiency of the emission reductions on annual concentrations and exceedances of PM limit values.

9 References

Aksoyoglu, S., Keller, J., Barmpadimos, I., Oderbolz, D., Lanz, V. A., Prévôt, A. S. H., and Baltensperger, U. (2011) Aerosol modelling in Europe with a focus on Switzerland during summer and winter episodes, *Atmos. Chem. Phys*., 11, 7355–7373, doi:10.5194/acp-11-7355-2011.

Amman, M. (edt.) (2012) Future emissions of air pollutants in Europe – Current legislation baseline and the scope for further reductions. *TSAP Report #1*, International Institute for Applied Systems Analysis (IIASA).

Ansari, A.S., Pandis, S. (1998) Response of inorganic PM to precursor concentrations. *Environ. Sci. Technol,* 32, 2706-2714.

Banzhaf, S., Schaap, M., Kerschbaumer, A., Reimer, E., Stern, R., van der Swaluw, E., and Builtjes, P. (2011) Implementation and evaluation of pH-dependent cloud chemistry and wet deposition in the chemical transport model REM-Calgrid, *Atmos. Environ*., 49, 378–390,doi:10.1016/j.atmosenv.2011.10.069.

Bergström, R., Denier van der Gon, H. A. C., Prévôt, A. S. H., Yttri, K. E., and Simpson, D. (2012) Modelling of organic aerosols over Europe (2002–2007) using a volatility basis set (VBS) framework: application of different assumptions regarding the formation of secondary organic aerosol, *Atmos. Chem. Phys*., 12, 8499-8527, doi:10.5194/acp-12-8499-2012.

Bessagnet et al. (2012) The CHIMERE atmospheric model, *EC4MACS publications*. http://www.ec4macs.eu/content/report/EC4MACS_Publications/MR_Final%20in%20pdf/CHIMERE_Methodologies_Final.pdf

Bessagnet B., L. Menut, G. Curci, A. Hodzic, B. Guillaume, C. Liousse, S. Moukhtar, B. Pun, C. Seigneur, M. Schulz (2009) Regional modelling of carbonaceous aerosols over Europe - Focus on Secondary Organic Aerosols, *Journal of Atmospheric Chemistry*, 61, 175-202.

Bessagnet, B., A. Hodzic, O. Blanchard, M. Lattuati, O. Le Bihan, H. Marfaing, L. Rouïl (2005) Origin of particulate matter pollution episodes in wintertime over the Paris Basin, *Atmos. Environ*., Volume 39, Issue 33, October 2005, Pages 6159-6174, ISSN 1352-2310, http://dx.doi.org/10.1016/j.atmosenv.2005.06.053.

Bicheron P., Defourny P., Brockmann C., Schouten L., Vancutsem C., Huc M., Bontemps S., Leroy M., Achard F., Herold M., Ranera F. and Arino O. (2008) "GlobCover 2005 – *Products description and validation report*", Version 2.1, Available on the ESA IONIA website (http://ionia1.esrin.esa.int/).

Binkowski, F. and Shankar, U. (1995) The Regional Particulate Matter Model .1. Model description and preliminary results, *J. Geophys. Res*., 100, 26191–26209.

Bott, A. (1989a) A positive definite advection scheme obtained by nonlinear re-normalization of the advection fluxes, *Mon. Weather Rev*., 117, 1006–1015.

Bott, A. (1989b) Reply, *Mon. Weather Rev*., 117, 2633–2636.

Cuvelier, C., P. Thunis, D. Karam, M. Schaap, C. Hendriks, R. Kranenburg, H. Fagerli, A. Nyiri, D. Simpson, P. Wind, M. Schulz, B. Bessagnet, A. Colette, E. Terrenoire, L. Rouïl, R. Stern, A. Graff, J.M.

Baldasano and M.T. Pay (2013) ScaleDep: Performance of European chemistry-transport models as function of horizontal spatial resolution, *MSC-W Technical Report 1/2013*.

Denier van der Gon, H., Jozwicka, M., Hendriks, E., Gondwe, M., and Schaap, M. (2010) Mineral dust as a component of particulate matter, Tno, bop - wp2 - report, report 500099003, TNO Delft, The Netherlands, www.pbl.nl, iSSN:1875–2322 (print) ISSN: 1875- 2314 (on line).

Derwent, R., Witham, C., Redington, A., Jenkin, M., Stedman, J., Yardley, R., and Hayman, G. (2009) Particulate matter at a rural location in southern England during 2006: Model sensitivities to precursor emissions. *Atmos. Environ.,* 43 (2009) 689–696.

de Smet, P.A.M. and Hettelingh, J.-P. (2001) Intercomparison of Current Landuse/Land Cover Databases, In Posch, M. , de Smet, P.A.M. , Hettelingh, J.-P. , and Downing, R.J. , editors, Modelling and Mapping of Critical Thresholds in Europe. Status report 2001. Coordination Centre for Effects, RIVM, Bilthoven, The Netherlands.

Deutsch, F., Vankerkom, J., Janssen, L., Janssen, S., Bencs, L., van Grieken, R., Fierens, F., Dumont, G., and Mensink, C. (2008) Modelling concentrations of airborne primary and secondary PM_{10} and $PM_{2.5}$ with the BelEUROS-model in Belgium. *Ecological Modelling,* 217, 230–239.

EEA (2012) Air quality in Europe — 2012 report, EEA Report No 4/2012, Office for Official *Publications of the European Union*, ISBN 978-92-9213-328-3. http://www.eea.europa.eu/publications/air-quality-in-europe-2012

Emberson, L.D., Ashmore, M.R., Simpson, D., Tuovinen, J.-P., Cambridge, H.M. (2000a) Towards a model of ozone deposition and stomatal uptake over Europe. *EMEP/MSC-W 6/2000*, Norwegian Meteorological Institute, Oslo, Norway, 57 pp.

Emberson, L.D., Ashmore, M.R., Simpson, D., Tuovinen, J.-P., Cambridge, H.M. (2000b) Modelling stomatal ozone flux across Europe. *Water, Air and Soil Pollution*, 109, 403-413.

Erisman, J.W., Bleeker, A., Hensen, A., Vermeulen, A. (2008) Agricultural air quality in Europe and the future perspectives. *Atmospheric Environment,* 42, 3209–3217.

Erisman, J.W. and Schaap, M. (2004) The need for ammonia abatement with respect to secondary PM reductions in Europe. *Env. Pollut.,* 129, pp. 159-163.

Fountoukis, C. and Nenes, A. (2007) ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K^+–Ca^{2+}–Mg^{2+}–$NH4^+$–Na^+–SO_4^{2-}–NO_{3-}–Cl^-–H2O aerosols, *Atmos. Chem. Phys*., 7, 4639-4659, doi:10.5194/acp-7-4639-2007.

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C. (2006) Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), Atmos. Chem. Phys., 6, 3181-3210, doi:10.5194/acp-6-3181-2006.

Guibert, S., V. Matthias, M. Schulz, J. Bösenberg, R. Eixmann, I. Mattis, G. Pappalardo, M. R. Perrone, N. Spinelli, and G. Vaughan (2005) The vertical distribution of aerosol over Europe - Synthesis of one year of EARLINET aerosol lidar measurements and aerosol transport modeling with LMDzT-INCA, Atmos. Environ., 39 (16), 2933-2943.

Harrison, R.M., Jones, A.M., Beddows, D.C.S., and 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.

IIASA (2004) CAFE Scenario Analysis Report Nr. 2, "The "Current Legislation" and the "Maximum Technically Feasible Reduction" cases for the CAFE baseline emission projections" (IIASA, November 2004) http://ec.europa.eu/environment/archives/cafe/activities/pdf/cafe_scenario_report_2.pdf

Koeble, R. and Seufert, G. (2001) Novel Maps for Forest Tree Species in Europe, in: A Changing Atmosphere, 8th European Symposium on the Physico-Chemical Behaviour of Atmospheric Pollutants, Torino, Italy, 17–20 September, 2001, http://ies.jrc.ec.europa.eu/Units/cc/events/torino2001/torinocd/ Documents/Terrestrial/TP35.htm.

Köhler, I., Sausen, R., and Klenner, G. (1995) NOx production from lightning, The impact of NOx emissions from aircraft upon the atmosphere at flight altitudes 8–15 km (AERONOX), edited by: Schumann, U., *final report to the Commission of the European Communities*, Deutch Luft und Raumfart, Oberpfaffenhofen, Germany.

Kuenen, J., H. Denier van der Gon, A. Visschedijk, H. van der Burgh, R. van Gijlswijk (2011) MACC European emission inventory 2003-2007*, TNO report*, TNO-060-UT-2011-00588.

López-Aparicio, S., Guerreiro, C., Viana, M., Reche, C., Querol, X. (2013) Contribution of agriculture to Air Quality problems in cities and in rural areas in Europe. ETC/ACM Technical Paper 2013/10.

Martensson, E., Nilsson, E., de Leeuw, G., Cohen, L., and Hansson, H.-C. (2003) Laboratory simulations and parameterisation of the primary marine aerosol production, *J. Geophys. Res*., 108, 4297, doi:10.1029/2002JD002263.

Megariti, A.G., Fountoukis, C., Charalampidis, P. E., Pilinis, C., and Pandis, S. N. (2012) Response of fine particulate matter concentrations to changes of emissions and temperature in Europe. *Atmos. Chem. Phys. Discuss*., 12, 8771–8822.

Menut L, B. Bessagnet, D. Khvorostyanov, M. Beekmann, N. Blond, A. Colette, I. Coll, G. Curci, G. Foret, A. Hodzic, S. Mailler, F. Meleux, J.L. Monge, I. Pison, G. Siour, S. Turquety, M. Valari, R. Vautard and M.G. Vivanco (2013) CHIMERE 2013: a model for regional atmospheric composition modelling, *Geoscientific Model Development*, 6, 981-1028, doi:10.5194/gmd-6-981-2013.

Monahan, E., Spiel, D., and Davidson, K. (1986) A model of marine aerosol generation via white caps and wave disruption, in: Oceanic whitecaps, edited by: Monahan, E. and Mac Niochaill, G., 167–193, Dordrecht: Reidel, The Netherlands.

Nenes, A., Pilinis, C., and Pandis, S. N. (1999) Continued development and testing of a new thermodynamic aerosol module for urban and regional air quality models, *Atmos. Environ*., 33, 1553–1560.

Odman, M. T., Hu, Y., Russell, A. G., Hanedar, A., Boylan, J. W., and Brewer, P. F. (2009) Quantifying the sources of ozone, fine particulate matter, and regional haze in the Southeastern United States, *J. Environ. Manage.*, 90, 3155–3168.

Pay, M.T., Jiménez-Guerrero. P., Baldasano, J.M. (2012) Assessing sensitivity regimes of secondary inorganic aerosol formation in Europe with the CALIOPE-EU modeling system. *Atmospheric Environment*, 51, 146-164.

Renner, E. and 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.

Riahi, K., Dentener, F., Gielen, D., Grubler, A., Jewell, J., Klimont, Z., Krey, V., McCollum, D., Pachauri, S., Rao, S., van Ruijven, B., van Vuuren, D. P., and Wilson, C. (2012) Energy Pathways for Sustainable Development, in: Global Energy Assessment: Toward a Sustainable Future, IIASA, Laxenburg, Austria and Cambridge University Press, Cambridge, United Kingdom and New York, NY.

Sauter, F., van der Swaluw, E., Manders-Groot, A., Wichink Kruit, R., Segers, A., Eskes, H. (2012) LOTOS-EUROS v 1.8 Reference Guide, *TNO report* TNO-060-UT-2012-01451, 2012. http://www.lotos-euros.nl/doc/LOTOS-EUROS-v18-reference-guide.pdf.

Schaap, M., Sauter, F., Timmermans, R.M.A., Roemer, M., Velders, G., Beck, J. and Builtjes, P.J.H. (2008) The LOTOS-EUROS model: description, validation and latest developments. *Int. J. Environment and Pollution*, Vol. 32, No. 2, 270–290.

Schaap, M., Manders, A.M.M., Hendriks, E.C.J., Cnossen, J.M., Segers, A.J.S., Denier van der Gon, H.A.C., Jozwicka, M., Sauter, F., Velders, G., Matthijsen, J., Builtjes, P.J.H. (2009). Regional modelling of particulate matter for the Netherlands. *PBL Report* 500099008. http://www.pbl.nl/bibliotheek/rapporten/500099008.pdf

Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Flechard, C. R., Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyíri, A., Richter, C., Semeena, V. S., Tsyro, S., Tuovinen, J.-P., Valdebenito, Á., and Wind, P. (2012) The EMEP MSC-W chemical transport model – technical description. *Atmos. Chem. Phys*., 12, 7825–7865.

Thunis P., C. Cuvelier, P. Roberts, L. White, L. Post, L. Tarrason, S. Tsyro, R. Stern, A. Keschbaumer, L.Rouil, B. Bessagnet, R. Bergstrom, M. Schaap, G. Boersen, P. Builtjes (2008) EuroDelta-II, Evaluation of a Sectorial Approach to Integrated Assessment Modelling including the Mediterranean Sea. *JRC Scientific and Technical Reports* – EUR 23444 EN 2008.

Troen, I. and L. Mahrt (1986) A simple model of the atmospheric boundary layer: Sensitivity to surface evaporation. *Bound.-Layer Meteorol.*, **37**, 129-148.

Tsimpidi, A.P., Karydis, V.A., and Pandis, S.N., (2007) Response of inorganic fine particulate matters to emission changes of SO_2 and NH_3: the Eastern United States as a case study, *J. Air Waste Manage*., 57, 1489–1498.

Tsyro, S., Aas, W., Soares, J., Sofiev, M., Berge, H., and Spindler, G. (2011) Modelling of sea salt concentrations over Europe: key uncertainties and comparison with observations, *Atmos. Chem. Phys.*, 11, 10367–10388, doi:10.5194/acp-11-10367-2011.

Tuovinen, J.-P., Ashmore, M., Emberson, L., and Simpson, D. (2004) Testing and improving the EMEP ozone deposition module, *Atmos. Environ*., 38, 2373–2385.

UNECE (2012) Decision 2012/2 Amendment of the text of and annexes II to IX to the 1999 Protocol to Abate Acidification, Eutrophication and Ground-level Ozone and the addition of new annexes X and XI: http://www.unece.org/env/lrtap/multi_h1.html

van Leer B. (1984) Multidimensional explicit difference schemes for hyperbolic conservation laws, in Computing Methods in Applied Sciences and Engineering VI , edited by R. Growinski and J. L. Lions (Elsevier, Amsterdam, 1984).

van Zanten, M. C., Sauter, F. J.,Wichink Kruit, R. J., Van Jaarsveld, J. A., and Van Pul, W. A. J.: Description of the DEPAC module (2010) Dry deposition modelling with DEPAC GCN2010, RIVM report 680180001/2010, Bilthoven, The Netherlands, 74 pp.

Vautard, R., B. Bessagnet, M. Chin, and L. Menut (2005) On the contribution of natural Aeolian sources to particulate matter concentrations in Europe: testing hypotheses with a modelling approach*, Atmos. Environ*., 39, 3291–3303.

Venkatram, A. and Pleim, J. (1999) The electrical analogy does not apply to modelling dry deposition of particles, *Atmos. Environ*., 33, 3075-3076.

Vestreng, V. , G. Myhre, H. Fagerli, S. Reis, and L. Tarrason (2007) Twenty-five years of continuous sulphur dioxide emission reduction in Europe, *Atmos. Chem. Phys*., 7, 3663–3681.

Walcek, C. J. and Aleksic, N. M. (1998) A simple but accurate mass conservative peak-preserving, Mixing ratio bounded advection algorithm with fortran code, *Atmos. Environ*., 32, 3863–3880.

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 PM10 and PM2.5 in the Netherlands; measurement and modelling results*. PBL Report nr. 500099006*. ISSN: 1875-2322 (print) ISSN: 1875-2314 (on line).

Wichink Kruit, R.J., W.AJ. van Pul, F.J. Sauter, M. van den Broek, E. Nemitz, M.A. Sutton, M. Krol and A.A.M. Holtslag (2010) Modeling the surface-atmosphere exchange of ammonia. *Atmos. Environ*., Vol. 44, 7: 877-1004.

ANNEX 1: 2020 emissions ceilings according to the Gothenburg protocol

SO2

Country	Emission 2009 (kT)	Emission 2005 (kT)	Reduction (%)	Ceiling 2020 (kT)
Austria	17,4	27	26	20,0
Belarus	157,8	79	20	63,2
Belgium	76,7	145	43	82,7
Bulgaria	440,4	777	78	170,9
Croatia	59,3	63	55	28,4
Cyprus	17,9	38	83	6,5
Czech Republic	173,5	219	45	120,5
Denmark	14,3	23	35	15,0
Estonia	54,8	76	32	51,7
Finland	59,2	69	30	48,3
France	289,3	467	55	210,2
Germany	434,7	517	21	408,4
Greece	424,5	542	74	140,9
Hungary	79,7	129	46	69,7
Ireland	32,6	71	65	24,9
Italy	232,1	403	35	262,0
Latvia	4,1	6,7	8	6,2
Lithuania	29,5	44	55	19,8
Luxembourg	2,2	2,5	34	1,7
Malta	8,0	11	77	2,5
Netherlands	37,4	65	28	46,8
Norway	14,5	24	10	21,6
Poland	861,7	1224	59	501,8
Portugal	74,3	177	63	65,5
Romania	459,9	643	77	147,9
Slovakia	64,1	89	57	38,3
Slovenia	10,6	40	63	14,8
Spain	462,6	1282	67	423,1
Sweden	29,6	36	22	28,1
Switzerland	12,4	17	21	13,4
United Kingdom of Great Britain and Northern Ireland	397,3	706	59	289,5

NOx

Country	Emission 2009 (kT)	Emission 2005 (kT)	Reduction (%)	Ceiling 2020 (kT)
Austria	187,1	231	37	145,5
Belarus	189,2	171	25	128,3
Belgium	207,5	291	41	171,7
Bulgaria	117,3	154	41	90,9
Croatia	75,9	81	31	55,9
Cyprus	19,6	21	44	11,8
Czech Republic	251,4	286	35	185,9
Denmark	131,5	181	56	79,6
Estonia	30,2	36	18	29,5
Finland	154,7	177	35	115,1
France	1105,5	1430	50	715,0
Germany	1320,9	1464	39	893,0
Greece	381,9	419	31	289,1
Hungary	166,9	203	34	134,0
Ireland	87,4	127	49	64,8
Italy	973,5	1212	40	727,2
Latvia	32,0	37	32	25,2
Lithuania	53,5	58	48	30,2
Luxembourg	43,9	19	43	10,8
Malta	8,9	9,3	42	5,4
Netherlands	280,3	370	45	203,5
Norway	178,7	200	23	154,0
Poland	822,1	866	30	606,2
Portugal	199,0	256	36	163,8
Romania	252,0	309	45	170,0
Slovakia	84,2	102	36	65,3
Slovenia	45,7	47	39	28,7
Spain	943,7	1292	41	762,3
Sweden	153,1	174	36	111,4
Switzerland	79,5	94	41	55,5
United Kingdom of Great Britain and Northern Ireland	1143,3	1580	55	711,0

NH3

Country	Emission 2009 (kT)	Emission 2005 (kT)	Reduction (%)	Ceiling 2020 (kT)
Austria	63,4	63	1	62,4
Belarus	150,0	136	7	126,5
Belgium	69,1	71	2	69,6
Bulgaria	52,5	60	3	58,2
Croatia	36,9	40	1	39,6
Cyprus	5,2	5,8	10	5,2
Czech Republic	73,0	82	7	76,3
Denmark	75,1	83	24	63,1
Estonia	10,0	9,8	1	9,7
Finland	37,2	39	20	31,2
France	656,2	661	4	634,6
Germany	575,7	573	5	544,4
Greece	62,1	68	7	63,2
Hungary	67,9	80	10	72,0
Ireland	108,4	109	1	107,9
Italy	392,7	416	5	395,2
Latvia	16,5	16	1	15,8
Lithuania	28,3	39	10	35,1
Luxembourg	4,7	5	1	5,0
Malta	1,5	1,6	4	1,5
Netherlands	125,1	141	13	122,7
Norway	22,7	23	8	21,2
Poland	273,4	270	1	267,3
Portugal	47,4	50	7	46,5
Romania	187,7	199	13	173,1
Slovakia	25,1	29	15	24,7
Slovenia	17,7	18	1	17,8
Spain	354,7	365	3	354,1
Sweden	50,1	55	15	46,8
Switzerland	62,8	64	8	58,9
United Kingdom of Great Britain and Northern Ireland	283,0	307	8	282,4

NMVOC

Country	Emission 2009 (kT)	Emission 2005 (kT)	Reduction (%)	Ceiling 2020 (kT)
Austria	120,9	162	21	128,0
Belarus	362,0	349	15	296,7
Belgium	105,1	143	21	113,0
Bulgaria	91,3	158	21	124,8
Croatia	77,4	101	34	66,7
Cyprus	11,4	14	45	7,7
Czech Republic	151,2	182	18	149,2
Denmark	89,1	110	35	71,5
Estonia	36,7	41	10	36,9
Finland	111,4	131	35	85,2
France	865,6	1232	43	702,2
Germany	930,6	1143	13	994,4
Greece	212,1	222	54	102,1
Hungary	128,1	177	30	123,9
Ireland	47,7	57	25	42,8
Italy	1131,2	1286	35	835,9
Latvia	60,5	73	27	53,3
Lithuania	66,2	84	32	57,1
Luxembourg	9,4	9,8	29	7,0
Malta	2,6	3,3	23	2,5
Netherlands	152,2	182	8	167,4
Norway	138,4	218	40	130,8
Poland	634,1	593	25	444,8
Portugal	179,7	207	18	169,7
Romania	432,7	425	25	318,8
Slovakia	64,3	73	18	59,9
Slovenia	34,0	37	23	28,5
Spain	672,0	809	22	631,0
Sweden	197,0	197	25	147,8
Switzerland	90,7	103	30	72,1
United Kingdom of Great Britain and Northern Ireland	822,4	1088	32	739,8

PM2.5

Country	Emission 2009 (kT)	Emission 2005 (kT)	Reduction (%)	Ceiling 2020 (kT)
Austria	19,4	22	20	17,6
Belarus	51,8	46	10	41,4
Belgium	15,8	24	20	19,2
Bulgaria	29,0	44	20	35,2
Croatia	10,5	13	18	10,7
Cyprus	2,3	2,9	46	1,6
Czech Republic	20,4	22	17	18,3
Denmark	25,4	25	33	16,8
Estonia	18,6	20	15	17,0
Finland	38,2	36	30	25,2
France	251,4	304	27	221,9
Germany	105,7	121	26	89,5
Greece	62,8	56	35	36,4
Hungary	27,8	31	13	27,0
Ireland	8,5	11	18	9,0
Italy	168,6	166	10	149,4
Latvia	28,3	27	16	22,7
Lithuania	8,6	8,7	20	7,0
Luxembourg	2,2	3,1	15	2,6
Malta	1,4	1,3	25	1,0
Netherlands	15,9	21	37	13,2
Norway	43,9	52	30	36,4
Poland	123,3	133	16	111,7
Portugal	57,2	65	15	55,3
Romania	115,1	106	28	76,3
Slovakia	27,4	37	36	23,7
Slovenia	15,9	14	25	10,5
Spain	75,2	93	15	79,1
Sweden	27,7	29	19	23,5
Switzerland	9,7	11	26	8,1
United Kingdom of Great Britain and Northern Ireland	67,0	81	30	56,7

PMcoarse & CO

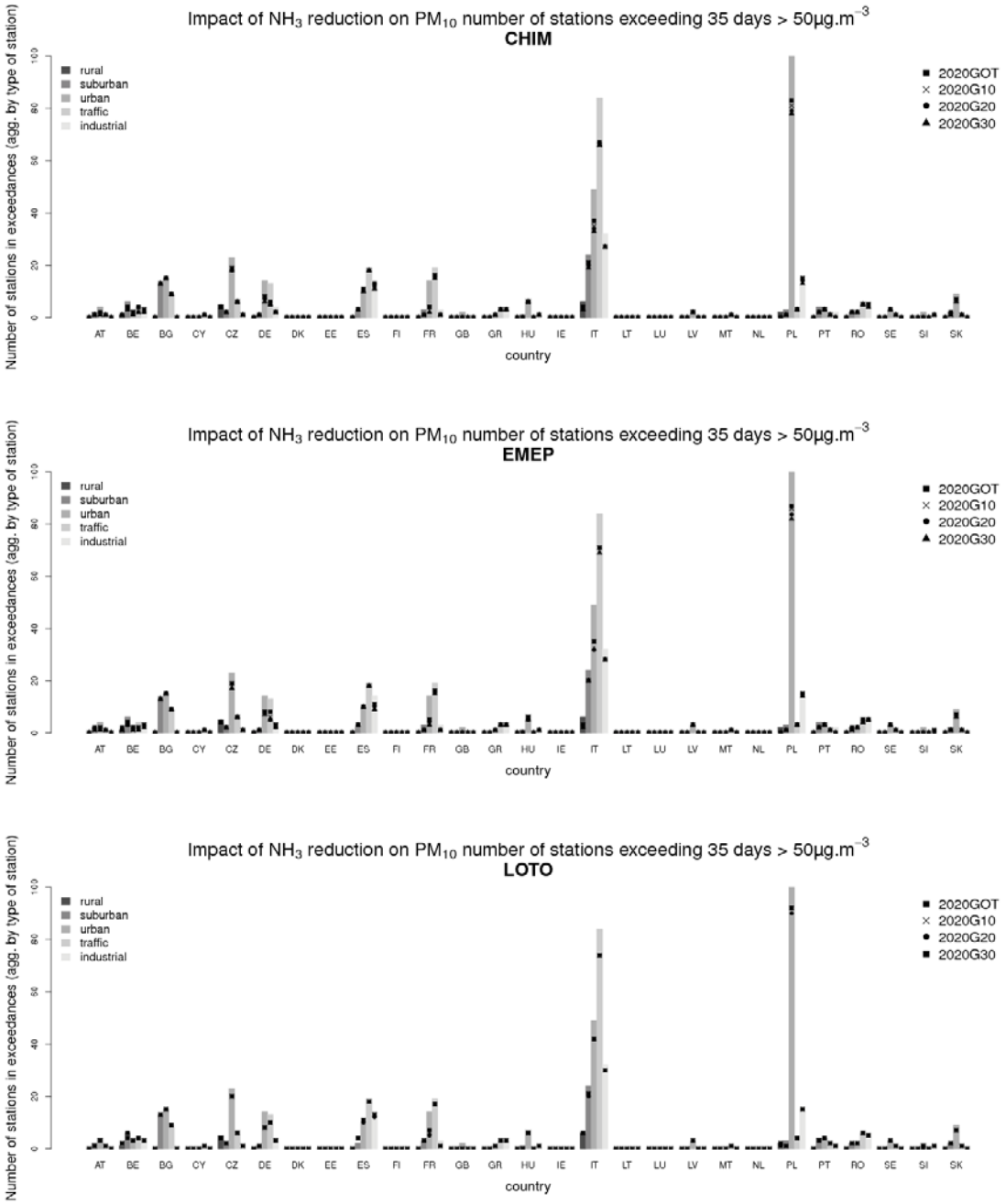
	PMcoarse	PMcoarse	CO	CO
Country	**Emission 2009 (kT)**	**Emission 2020 (kT)**	**Emission 2009 (kT)**	**Emission 2020 (kT)**
Austria	15,3	12,0	631,8	694,6
Belarus	12,8	7,8	990,0	951,1
Belgium	6,7	18,5	380,9	286,5
Bulgaria	19,9	27,7	253,7	392,9
Croatia	4,3	5,4	285,2	514,4
Cyprus	1,3	2,1	20,3	85,0
Czech Republic	15,9	14,5	403,5	437,8
Denmark	6,0	9,8	406,8	309,2
Estonia	4,7	2,4	168,2	105,3
Finland	13,4	6,4	465,5	602,1
France	112,8	86,0	3666,4	4575,6
Germany	81,3	87,0	3011,0	3999,5
Greece	37,2	18,0	591,3	1120,4
Hungary	19,9	12,9	312,8	486,5
Ireland	4,3	6,1	151,9	191,7
Italy	28,9	50,6	2725,3	3085,4
Latvia	4,6	1,9	266,9	133,0
Lithuania	2,4	3,6	169,2	155,5
Luxembourg	0,9	1,2	37,6	37,0
Malta	0,8	0,0	30,8	0,0
Netherlands	13,8	22,8	579,5	678,4
Norway	6,0	5,1	319,8	1542,1
Poland	125,3	51,4	2777,9	3068,3
Portugal	25,8	11,0	486,5	1810,4
Romania	20,9	34,1	1349,3	845,4
Slovakia	3,4	7,8	207,8	231,4
Slovenia	3,0	3,7	150,6	203,5
Spain	32,5	51,7	1686,5	3175,6
Sweden	11,6	9,5	612,2	598,3
Switzerland	10,6	5,3	256,5	331,1
United Kingdom of	46,9	50,3	2317,3	1809,7

ANNEX 2: Acronyms of EU countries

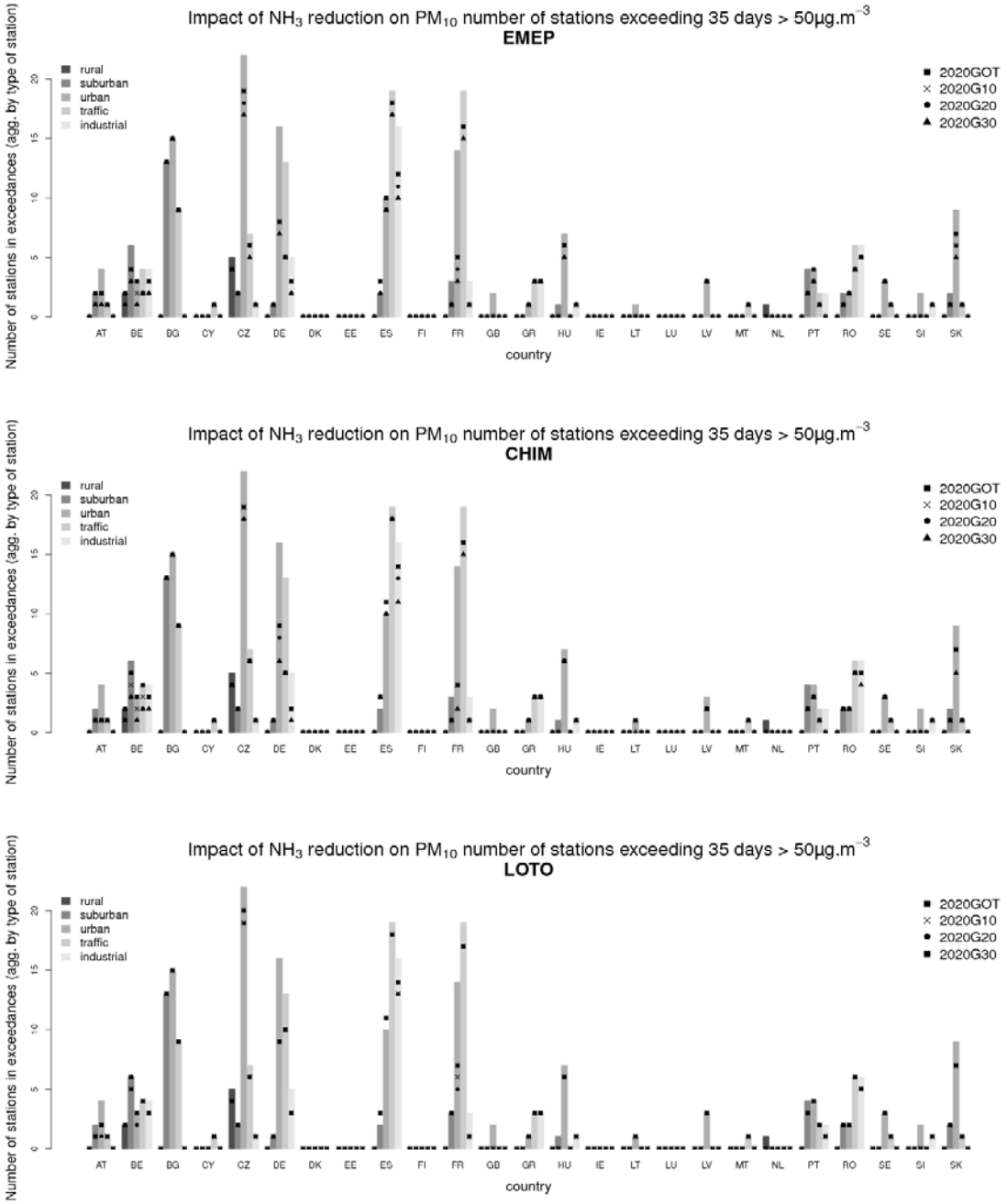
AT	Austria
BE	Belgium
BG	Bulgaria
CY	Cyprus
CZ	Czech Republic
DE	Germany
DK	Denmark
EE	Estonia
ES	Spain
FI	Finland
FR	France
GB	United Kingdom
GR	Greece
HU	Hungary
IE	Ireland
IT	Italy
LT	Lithuania
LU	Luxembourg
LV	Latvia
MT	Malta
NL	The Netherlands
PL	Poland
PT	Portugal
RO	Romania
SE	Sweden
SI	Slovenia
SK	Slovakia

ANNEX 3: Impact of NH3 emission reductions on the daily PM_{10} limits

Number of PM_{10} stations exceeding more than 35 days the daily limit value of 50 µg m^{-3} for the base year 2009, impact of the various scenarios for all EU countries with CHIMERE, EMEP and LOTOS-EUROS

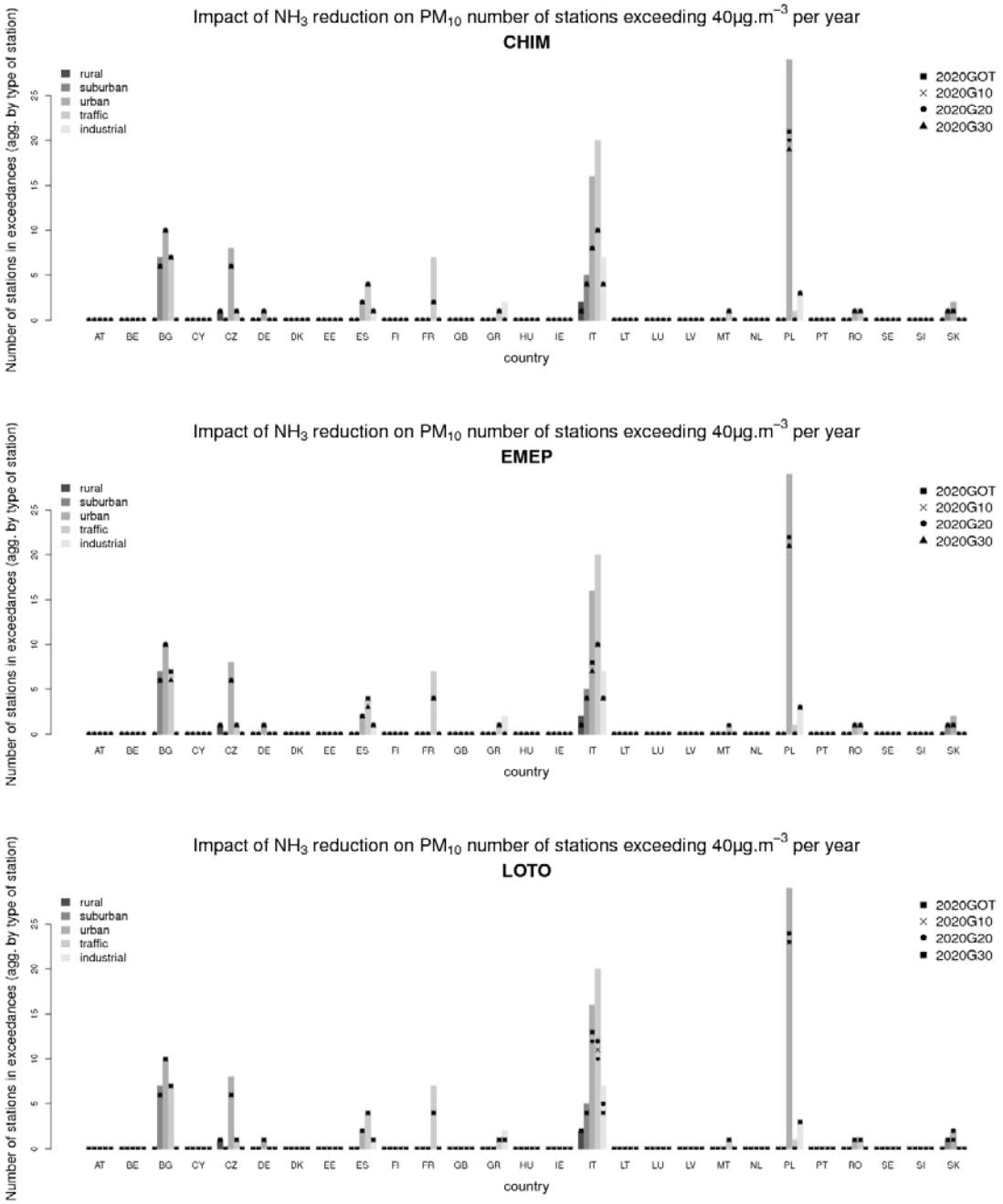


Number of PM_{10} stations exceeding more than 35 days the daily limit value of 50 µg m^{-3} for the base year 2009, impact of the various scenarios for all EU countries without IT and PL, with CHIMERE, EMEP and LOTOS-EUROS

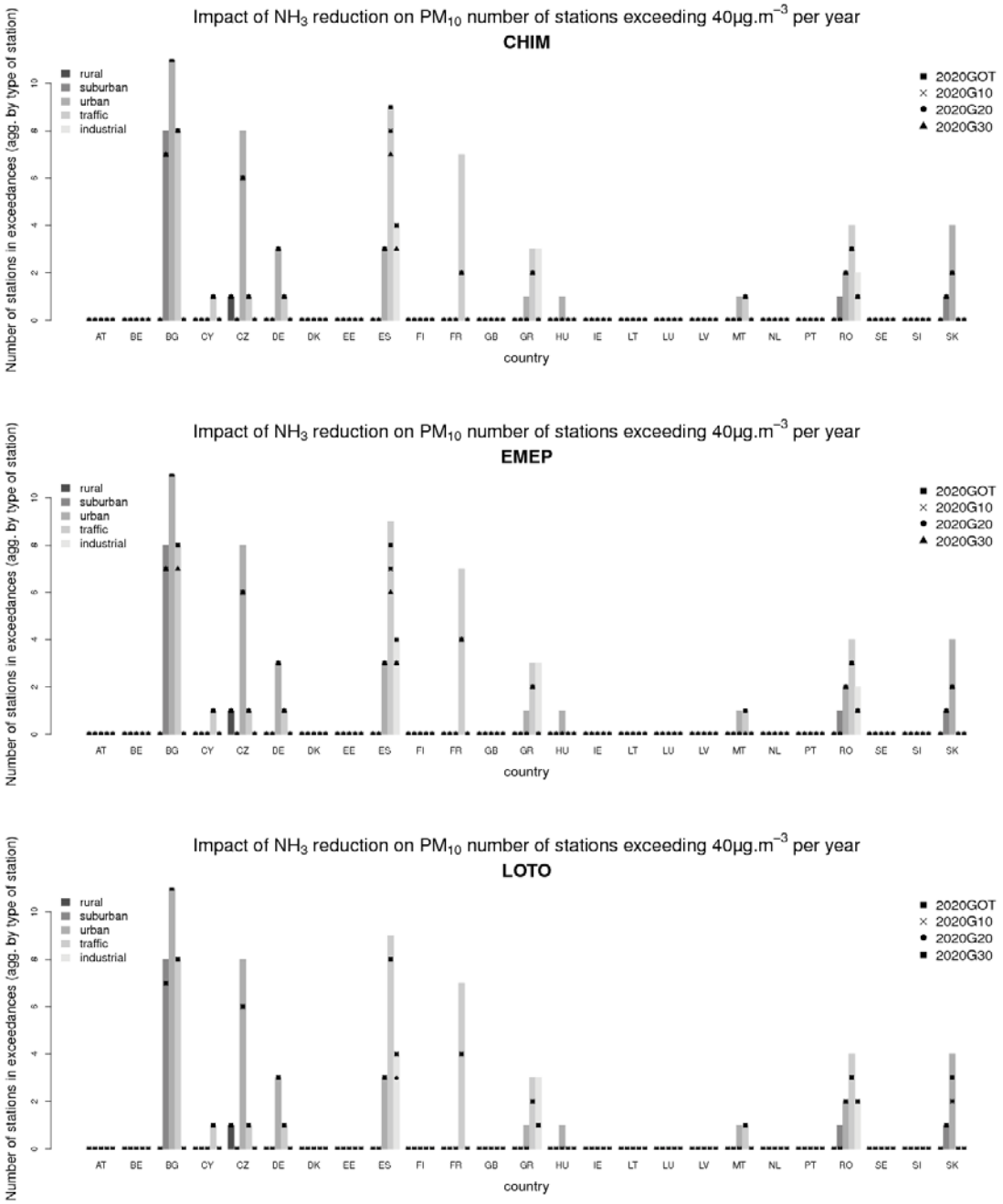


ANNEX 4: Impact of NH3 emission reductions on the annual PM10 limits

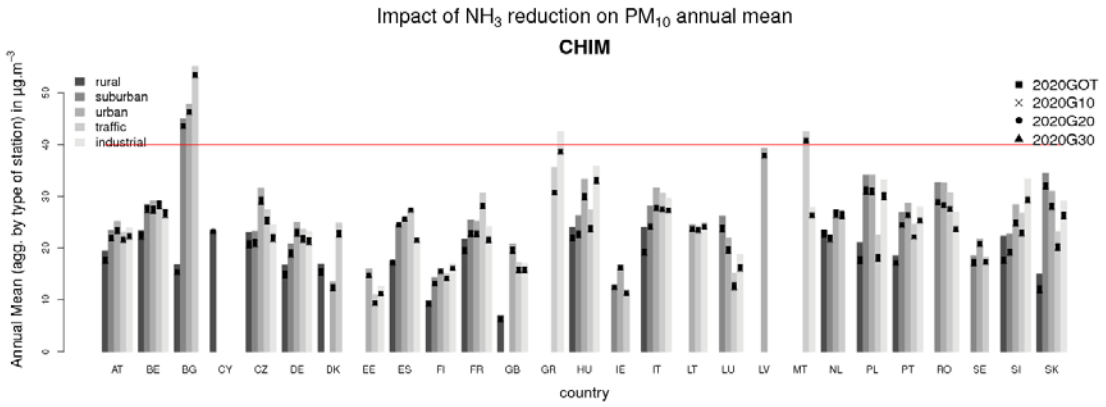
Number of PM_{10} stations exceeding the annual mean limit value of 40 µg m^{-3} for the base year 2009, impact of the various scenarios for all EU countries with CHIMERE, EMEP and LOTOS-EUROS

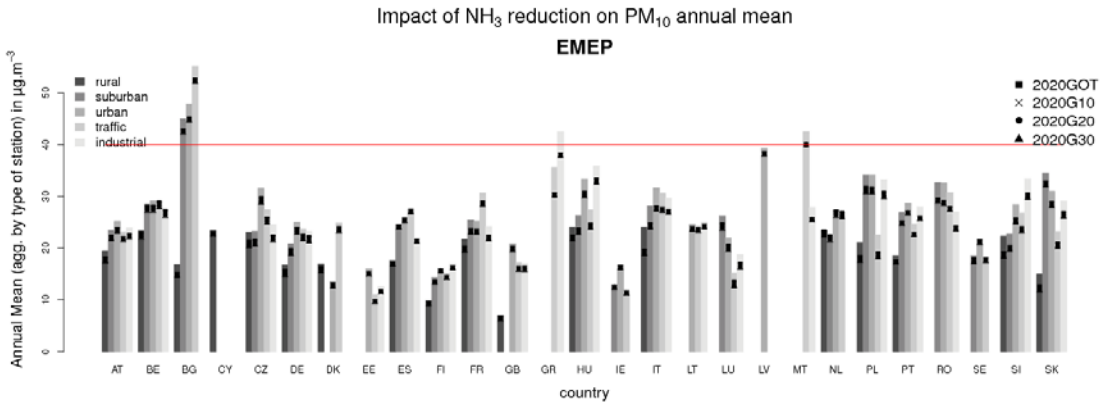


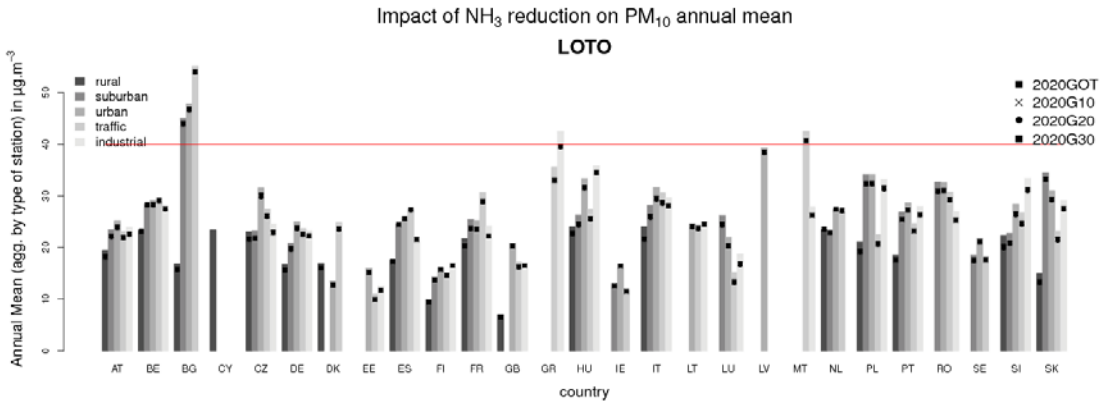
Number of PM_{10} stations exceeding annual mean limit value of 40 µg.m^{-3} for the base year 2009, impact of the various scenarios for all EU countries without IT and PL, with CHIMERE, EMEP and LOTOS-EUROS



ANNEX 5: Impact of NH_3 emission reductions scenarios on exceedance of the average annual PM_{10} limit value

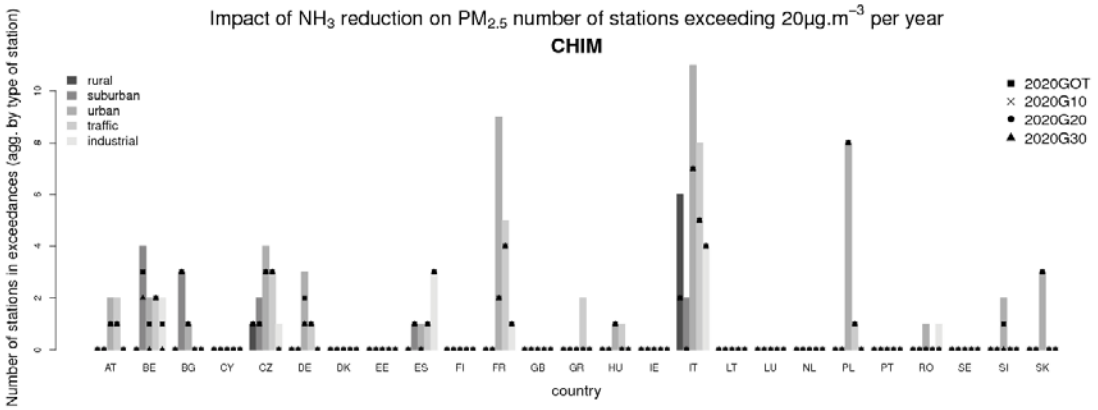


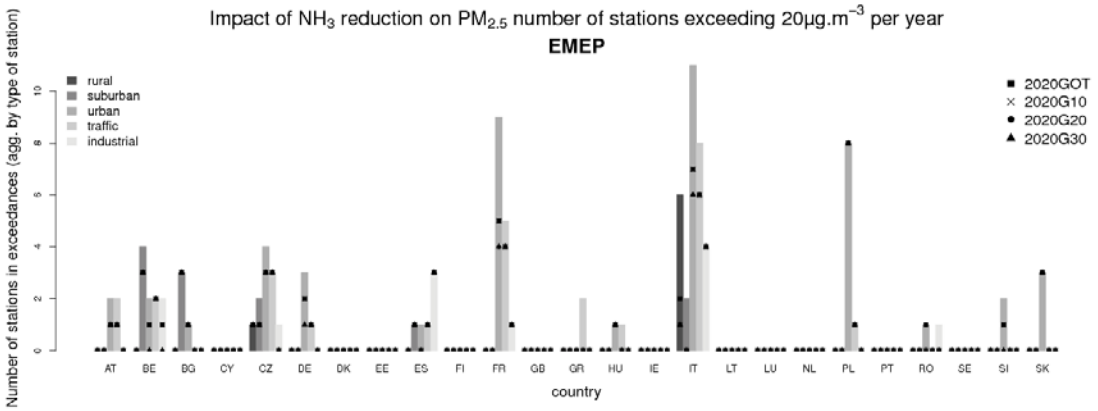


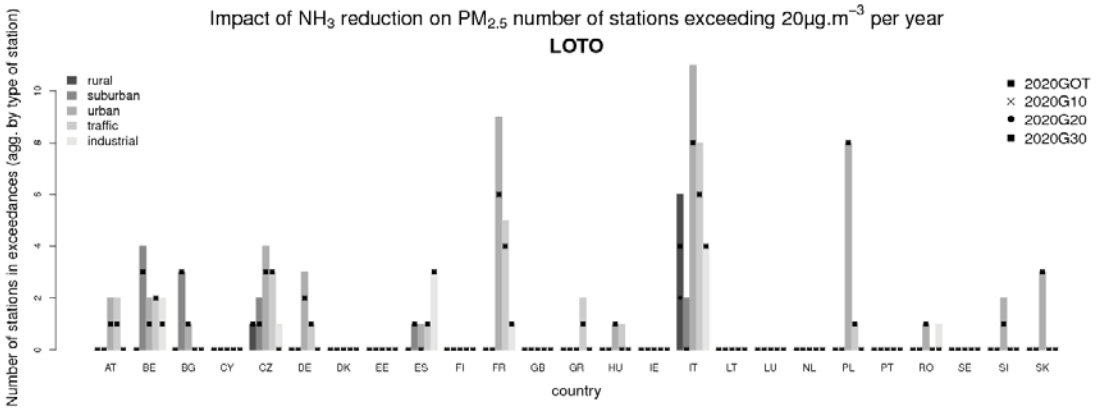


ANNEX 6: Impact of NH3 emission reduction scenarios on the annual mean $PM_{2.5}$ limit value

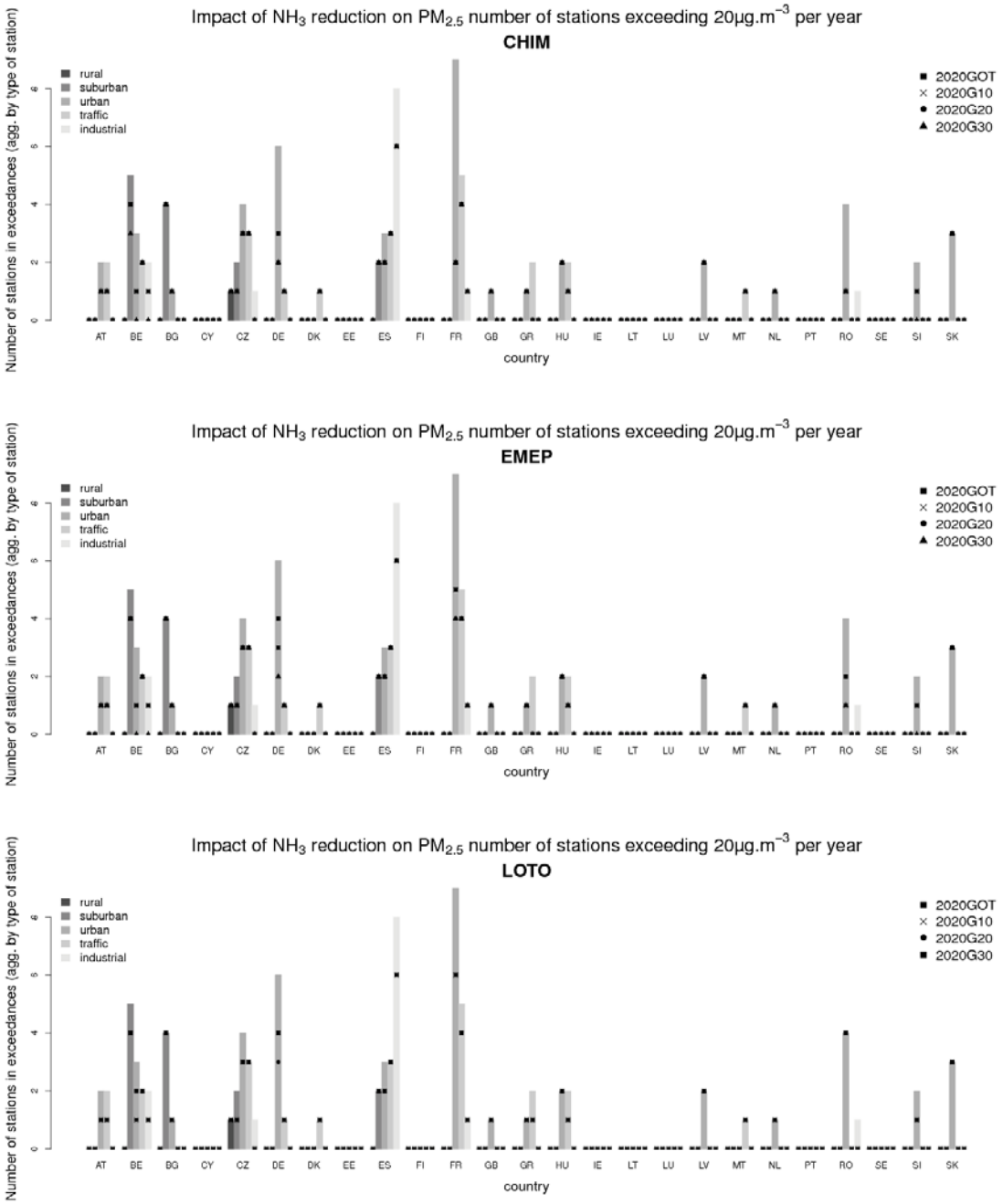
Number of $PM_{2.5}$ stations exceeding annual mean limit value of 20 µg.m^{-3} for the base year 2009, impact of the various scenarios for all EU countries with CHIMERE, EMEP and LOTOS-EUROS







Number of $PM_{2.5}$ stations exceeding annual mean limit value of 20 µg.m^{-3} for the base year 2009, impact of the various scenarios for all EU countries without IT and PL, with CHIMERE, EMEP and LOTOS-EUROS



ANNEX 7: Impact of Gothenburg Protocol and further NH_3 emission reductions on exceedances of the average annual $PM_{2.5}$ limit value