

CAMS Service Evolution



D6.1 Uncertainties introduced by non-linear chemistry in the CAMS source receptor products and consistency with CLRTAP/EMEP products.

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1 Executive Summary

CAMS currently offers a number of products to support decision and policy makers in regard to mitigation of air pollution at the European scale (<https://policy.atmosphere.copernicus.eu/>). These policy products provide information about the causes and main drivers of air pollution, so-called source receptor relationships (SR), and their potential evolution in the future.

One of the sources of uncertainty in the CAMS source receptor products are the methods used for accounting SR relationships. In the current CAMS policy service, 3 regional models perform source receptor calculations with different methodologies (answering slightly different questions), which for species formed through non-linear atmospheric chemistry yield different results.

In this task, we have explored and quantified the uncertainties associated with non-linear chemistry for regional source receptor CAMS products. We have defined two different experiments where we compare results from different regional models (EMEP, LOTOS-EUROS, CHIMERE) and source attribution methodologies (brute force (BF), local fraction (LF), labelling, non-linear surrogate modelling) for 1) country contributions to cities and 2) emission sector contributions to cities.

We found the largest differences in the city SRs from EMEP and LOTOS-EUROS (when using the same source attribution method, BF) to be in primary PM, while the SRs for the secondary inorganics (SIA) were more similar. This was contrary to what we expected, as chemistry is introducing additional uncertainties in modelling of SIA compared to modelling of primary PM. A similar issue was found in the sector SRs, where large differences in the residential biomass contributions between LOTOS-EUROS and EMEP was found for cities which have large residential heating emissions. We attribute these differences at least partly to differences in the depth of the surface layer in LOTOS-EUROS and EMEP. The results for the CHIMERE model are more in line with the EMEP model.

In theory, the choice of source attribution methodology does not matter for primary species. In practice, we see some small differences due to numerical implementation of methods, but the magnitude of differences are so small that they can be ignored. BF and LF methodologies are both methods that give potential impact of emission reductions and thus they give very similar results, even for source apportionment of secondary inorganic aerosols.

The labelling method traces where the pollution comes from (not what the impacts of emission reductions are) and is a fundamentally different method. The differences between BF and labelling for source apportionment of secondary inorganic aerosols can be significant, especially on a shorter time scale (e.g. daily). For the emission sector contributions, this is mostly connected to the agricultural and traffic exhaust sectors, and for some cities with large contributions from other sources of NO_x to these sectors as well.

When comparing the magnitude of differences for city SRs between model formulations (i.e. EMEP versus LOTOS-EUROS) and due to source attribution methodology (i.e. BF versus LF and BF versus labelling for city SRs), as defined by the root mean square error, we find that RMSE for the secondary inorganic aerosols (SIAs) are rather large due to SA methodologies and of the same magnitude as the differences due to model formulations for nitrate and sulfate. For PM_{2.5} the model differences (EMEP versus LOTOS-EUROS) are clearly larger than differences due to SA methodology. Here, the RMSE for PM_{2.5} is a combination of RMSE for primary PM (large differences due to model formulation, no differences due to SA

methodology) and SIA (comparable differences due to model formulation and SA methodology). For LF versus BF SA methodology, the differences are even smaller for SA methodology, and model differences totally dominant.

The comparison of the three CAMS source attribution systems for sector contributions underline the conclusions from the comparison for country contributions. On an annual basis the differences in source attribution results are mostly due to differences between the models instead of differences in the applied source attribution methods. The models show largest differences for the primary components from the residential combustion sector which may be attributed to differences in vertical mixing, and the depth of the surface layer in the models. Going to smaller timescales the impact of non-linearity on the results becomes more apparent and differences between brute force and tagging approaches grow larger. This is mostly associated with the formation of ammonium nitrate and the sources of its precursors, which are the agricultural sector for ammonia and traffic exhaust for NO_x. On shorter timescales it is therefore important to consider the purpose of the source attribution when choosing a source attribution method, i.e. tagging for the provision of contributions and brute force based approaches for the provision of potential impacts of emission reductions.

At present, the CAMS Policy Support Service separates the source attribution into different products based on the different methods: potential impact of emission reductions (BF, done with the EMEP model for spatial allocation, and ACT/CHIMERE for sectoral allocation) and contribution (labelling, done with LOTOS-EUROS for spatial allocation). For users, this is confusing, as many users do not understand the difference between contribution and potential impacts. In principle we could combine source attribution of primary PM from the different models (EMEP, LOTOS-EUROS and also CHIMERE) and methodologies into a mini-ensemble. However, for SIA (and to a lesser extent SOA) it is less clear how this can be done from a conceptual perspective, although in practice the differences due to model formulations are as large as differences in SA methodology.

Presentation of non-linearities in the CAMS Policy Support Service

At present, the forecasting of city SRs presents the impact of reducing emissions in countries and the city itself. The contributions are calculated by performing 15% reductions in the sources and scale up to 100%. In order to close the gap between the contributions shown and the total concentration, a term 'Others' is defined as the difference between the total concentration and the sum of the contributions shown.

We recommend that in the CAMS2_71 service for country-to-city and/or city-to- itself (impacts of emission reduction), the 'Other' term is replaced by 'Other sources' (meaning all other sources/regions that are quantified but not displayed) and a 'non-linearity' or 'closure' term which is quantified as the difference between the total concentration and the contributions of of all the different terms, which in theory would make up the concentrations if the model system was linear. If the non-linearity term is large, this means that you should be careful when interpreting how your emission reductions would impact PM_{2.5}/PM₁₀, because the impact will not scale with the size of your reduction.

Can we approximate the local contribution of PM_{2.5} in cities?

The local contribution to PM in cities is often approximated to primary PM_{2.5} (e.g. in the integrated assessment model GAINS), assuming that local production of SIA takes time and

matters essentially at regional scale. We show that although primary PM_{2.5} is indeed the largest local contribution for all the 79 European cities investigated, local production of SIA is important in some cities. For the 79 cities investigated, local SIA production are normally below 5 percent, but up to 20-30% of the local contribution for a few cities, for instance Athens, Madrid, Sofia and Milan.

On the consistency and comparability of Source-Receptor relationships in the LRTAP Convention and CAMS

A comparison between the Source-Receptor relationships calculated with the EMEP model for CAMS Policy service (CAMS_SR) and those for the Convention for Long Range Transboundary Air Pollution annual assessment (CLRTAP_SR) was carried out, where we analyzed source contributions to annual mean PM and ozone in 80 cities and 31 countries. In general, we find the results in terms of top-three pollution contributors from CLRTAP_SR and CAMS_SR to be reasonably consistent, given differences in the computational settings, model version and emissions. The deviations are well within 30% for PM. Somewhat larger differences for ozone are not surprising due to rather small magnitudes of the contribution from European countries compared to ozone pollution originated beyond Europe and ozone formed from biogenic precursors. Unfortunately, due to rather different model versions and input data, it was not feasible to discriminate the effect of different execution of emission reductions (all emissions in the same run in CAMS_SR vs individual emission reductions in separate CLRTAP_SR runs) on SR products. Given a fair general consistency between CAMS_SR in CLRTAP_SR products (especially for PM), some possibilities of their complimentary usage are suggested. Preliminary SR estimates, based on CAMS_SR products, could be provided to CLRTAP members 1.5-2 years earlier than SR tables are published in EMEP Status reports. CLRTAP_SR could possibly be used in CAMS Annual Assessment Reports since both assess the pollution situation 2 years back. Based on the latest version of EMEP model, meteorology from IFS reanalysis and EMEP emissions for the actual year, CLRTAP_SR results would provide quite relevant information, identifying the major sources of pollution and estimating potential impacts of emission reductions.

Table of Contents

1	Executive Summary	2
2	Introduction	6
2.1	Background.....	6
2.2	Scope of this deliverable	6
2.2.1	Objectives of this deliverables.....	6
2.2.2	Work performed in this deliverable.....	7
2.2.3	Deviations and countermeasures.....	7
2.2.4	CAMEO Project Partners:	7
3	Uncertainties related to non-linearity in chemistry and methodological approach	9
3.1	Introduction	9
3.2	Description of the setup of the experiment	10
3.3	Evaluation of the EMEP and LOTOS-EUROS models	14
3.4	City source receptor calculations for PM	17
3.4.1	Results.....	17
3.4.2	Quantification of differences due to model formulation and source attribution methodology	24
3.5	Results for sector source receptor calculations	27
3.6	Visualisation of non-linearities in the city SR forecast	33
3.7	Local contributions in cities from non-linear species.....	38
3.8	Conclusions and recommendations	42
4	Consistency and comparability between EMEP source-receptor calculations for CLRTAP and CAMS Policy Service	45
4.1	Introduction	45
4.2	Description of SR in CAMS and CLRTAP	45
4.3	Methodology for comparison.....	47
4.4	Results.....	49
4.4.1	PM _{2.5} and PM ₁₀	52
4.4.2	Ozone	53
4.4.3	Why is the match between CAMS_SR and CLRTAP_SR 1st contributor worse for ozone than PM?.....	54
4.5	Conclusions and recommendations	56
4.5.1	Potential use of CAMS_SR in CLRTAP_SR activities	57
4.5.2	Potential use of CLRTAP_SR in CAMS Policy Service.....	57
	Annex A - A simplified explanation of the NH ₃ -(NH ₄) ₂ SO ₄ -HNO ₃ -H ₂ SO ₄ non-linearities	59

2 Introduction

2.1 Background

Monitoring the composition of the atmosphere is a key objective of the European Union's flagship Space programme Copernicus, with the Copernicus Atmosphere Monitoring Service (CAMS) providing free and continuous data and information on atmospheric composition.

The CAMS Service Evolution (CAMEO) project will enhance the quality and efficiency of the CAMS service and help CAMS to better respond to policy needs such as air pollution and greenhouse gases monitoring, the fulfilment of sustainable development goals, and sustainable and clean energy.

CAMEO will help prepare CAMS for the uptake of forthcoming satellite data, including Sentinel-4, -5 and 3MI, and advance the aerosol and trace gas data assimilation methods and inversion capacity of the global and regional CAMS production systems.

CAMEO will develop methods to provide uncertainty information about CAMS products, in particular for emissions, policy, solar radiation and deposition products in response to prominent requests from current CAMS users.

CAMEO will contribute to the medium- to long-term evolution of the CAMS production systems and products.

The transfer of developments from CAMEO into subsequent improvements of CAMS operational service elements is a main driver for the project and is the main pathway to impact for CAMEO.

The CAMEO consortium, led by ECMWF, the entity entrusted to operate CAMS, includes several CAMS partners thus allowing CAMEO developments to be carried out directly within the CAMS production systems and facilitating the transition of CAMEO results to future upgrades of the CAMS service.

This will maximise the impact and outcomes of CAMEO as it can make full use of the existing CAMS infrastructure for data sharing, data delivery and communication, thus supporting policymakers, business and citizens with enhanced atmospheric environmental information.

2.2 Scope of this deliverable

2.2.1 Objectives of this deliverables

The first objective of this deliverable is to **explore and quantify** uncertainties associated with non-linear chemistry for regional source receptor CAMS products. We **explain their causes** and investigate the **potential for operational provision** of information related to chemical non-linearity.

A second objective of this deliverable is to analyse the consistency and comparability between the annual SR information for European countries (and cities) from CAMS and the SR matrices that are produced within EMEP and used for policy development within the LRTAP Convention (one of the major differences between source receptor calculations in CAMS and LRTAP is that emissions of SO_x, NO_x, NH₃, NMVOC and PPM are reduced together in one run in CAMS, but in individually in LRTAP).

We identify the potential for exploitation of EMEP/CLRTAP SR matrices in future operational CAMS products and vice versa (D6.1).

2.2.2 Work performed in this deliverable

In this deliverable the work as planned in the Description of Action (DoA, WP6 T6.1.1 and 6.1.2) was performed.

2.2.3 Deviations and countermeasures

No deviations have been encountered.

2.2.4 CAMEO Project Partners:

ECMWF	EUROPEAN CENTRE FOR MEDIUM-RANGE WEATHER FORECASTS
Met Norway	METEOROLOGISK INSTITUTT
BSC	BARCELONA SUPERCOMPUTING CENTER-CENTRO NACIONAL DE SUPERCOMPUTACION
KNMI	KONINKLIJK NEDERLANDS METEOROLOGISCH INSTITUUT-KNMI
SMHI	SVERIGES METEOROLOGISKA OCH HYDROLOGISKA INSTITUT
BIRA-IASB	INSTITUT ROYAL D'AERONOMIE SPATIALEDE BELGIQUE
HYGEOS	HYGEOS SARL
FMI	ILMATIETEEN LAITOS
DLR	DEUTSCHES ZENTRUM FUR LUFT - UND RAUMFAHRT EV
ARMINES	ASSOCIATION POUR LA RECHERCHE ET LE DEVELOPPEMENT DES METHODES ET PROCESSUS INDUSTRIELS
CNRS	CENTRE NATIONAL DE LA RECHERCHE SCIENTIFIQUE CNRS
GRASP-SAS	GENERALIZED RETRIEVAL OF ATMOSPHERE AND SURFACE PROPERTIES EN ABREGE GRASP
CU	UNIVERZITA KARLOVA
CEA	COMMISSARIAT A L ENERGIE ATOMIQUE ET AUX ENERGIES ALTERNATIVES
MF	METEO-FRANCE
TNO	NEDERLANDSE ORGANISATIE VOOR TOEGEPAST NATUURWETENSCHAPPELIJK ONDERZOEK TNO
INERIS	INSTITUT NATIONAL DE L ENVIRONNEMENT INDUSTRIEL ET DES RISQUES - INERIS
IOS-PIB	INSTYTUT OCHRONY SRODOWISKA - PANSTWOWY INSTYTUT BADAWCZY

CAMEO

FZJ	FORSCHUNGSZENTRUM JULICH GMBH
AU	AARHUS UNIVERSITET
ENEA	AGENZIA NAZIONALE PER LE NUOVE TECNOLOGIE, L'ENERGIA E LO SVILUPPO ECONOMICO SOSTENIBILE

3 Uncertainties related to non-linearity in chemistry and methodological approach

3.1 Introduction

CAMS currently offers a number of products to support decision and policy makers in regard to mitigation of air pollution at the European scale (<https://policy.atmosphere.copernicus.eu/>). These policy products provide information about the causes and main drivers of air pollution, so-called source receptor relationships (SR), and their potential evolution in the future. Quantitative uncertainty information is needed if the products are to be used by policy makers to prioritise measures in different activity sectors and to gauge the scale of actions that must be targeted when designing air quality policies with short or long term perspectives.

One of the sources of uncertainty in the CAMS source receptor products are the methods used for accounting SR relationships. In the current CAMS policy service, 3 regional models perform source receptor calculations with different methodologies (answering slightly different questions), which for species formed through non-linear atmospheric chemistry yield different results.

At present, the following models and methodologies are being applied within CAMS2_71:

- **EMEP model:** Country-to-city SR and city-to-city SR with brute force (BF) methodology (i.e. reducing emissions of NO_x , SO_x , NH_3 , VOC, PPM 15% (at the same time) for countries and cities and scaling the effect up to 100%). This method gives the effect of emission reductions (Potential impacts, e.g. Clappier et al., 2022), for instance: What happens to PM if you reduce NH_3 ? In principle the result could be zero (at some specific place) as NH_3 could be in excess of H_2SO_4 and no HNO_3 available for formation of NH_4NO_3 . This method can also give negative contributions, for instance reducing NO_x in a city might lead to increased ozone.
- **LOTOS-EUROS model:** Country-to-country SR with **labelling** (Kranenburg et al. 2013). This method traces where the pollution comes from. It will never give zero contributions. For instance, even if NH_3 is in excess and reductions of NH_3 gives little impact on NH_4^+ , it will trace the amount of NH_3 in the $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 that is present.
- **CHIMERE model:** Sector-to-city contributions and scenarios with a parametrized concentration-emission response function updated on a daily basis (the parameterization is based on doing several brute force runs with different combinations of reductions in the different components, Colette et al, 2022). The results in the sector contribution is based on 100% reductions.

In addition to these methods, a new method for calculating source receptor relationships has been developed at MET Norway - the local fraction (LF) methodology, also referred to as sensitivities (Wind et al. 2020, Wind et al. 2024). The LF method can be considered as a kind of brute force methodology - but the emission reductions are very small (It calculates the derivative dC/dE at the current concentration). Leveraging on the experience with the LF method within the work of the CAMEO project, it is planned to use the LF method within the CAMS policy user service within the next few months, and therefore we include this method in our analysis.

In principle, all these methods; BF, tagging, ACT (non-linear surrogate modelling) and LF give the same results for primary species such as primary PM. In practice, however, there can be (small) differences. For instance, the LF and BF methods are slightly different due to the Bott

advection scheme (Wind et al. 2024). The ACT surrogate was demonstrated to be within 2% of sensitivity simulations with the full CTM CHIMERE.

For the secondary species such as nitrate, sulfate, ammonium, secondary organic aerosols or ozone, chemical non-linearities lead to differences in the results.

Clearly, differences in the results also arise from differences in the model formulations themselves (for instance in chemistry, deposition and/or advection schemes) and in the setup of models (for instance the use of different emissions, meteorological drivers, boundary conditions, etc). Here, we are interested to understand whether differences that arise due to differences in methodology are of the same magnitude (or larger/smaller) than differences due to different model formulations.

3.2 Description of the setup of the experiment

In order to be able to quantify the differences in the SRs (country-to-city or sector-to-city) that result from differences in methodology (e.g. BF vs LF vs tagging) versus differences in model formulations, we first agreed to a detailed specification of the model setups (see Table 3.2.1).

Table 3.2.1: Overview of the model setups for EMEP, LOTOS-EUROS and CHIMERE in the operational CAMS2_71 service (in 2023, when the CAMEO exercise started) and in the CAMEO exercise.

	CAMS2_71: EMEP (in 2023)	CAMS2_71: LOTOS-EUROS (in 2023)	CAMS2_71: CHIMERE (in 2023)	Agreed for the CAMEO experiment
Forest fires	GFAS v1.2	GFAS v1.2	GFAS v1.2	GFAS v1.2
Soil NO _x	CAMS-GLOB-SOIL v2.3	Novak and Pierce (1993).	MEGAN V2.10	each model their own set-up
Model resolution (meteorology resolution)	0.25x0.125 (MACC14 grid). IFS forecast meteor, 12 UTC	{0.4x0.2} [0.2x0.1], 12UTC	0.25x0.25	0.2x0.1
Domain	lon : -30 to 45 by 0.25 degrees east lat : 30 to 76 by 0.125 degrees north	{lon:-25 to 45. lat: 30 to 71.7} [lon: -10 to 30. Lat: 35 to 65]	lon: -25 to 45 by 0.25 degrees east lat : 30 to 70 by 0.25 degrees north	lon: -24.9 to 44.9 by 0.2 degrees east lat : 30.05 to 71.95 by 0.1 degrees north
Emission resolution	0.1x0.05	0.1x0.05	0.1x0.05	0.1x0.05
Emission version	CAMS v5.1 (Ref 2.0.1) for 2018 (CAMS2_40 U5)	CAMS v5.1 (Ref 2.0.1) for 2018	CAMsv5.1 for 2018 (before may 2023)	CAMsv6.1 for 2019 and 2018

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	CAMS2_71: EMEP (in 2023)	CAMS2_71: LOTOS-EUROS (in 2023)	CAMS2_71: CHIMERE (in 2023)	Agreed for the CAMEO experiment
			CAMSv2.2.1 for 2015)	
Emission time factors	GENEMIS	TNO profiles, HDD for GNFR C, PPM	TNO profiles	CAMS TEMPO v4.1 HDD for RWC (for LE: TNO diurnal profile)
City definitions	3x3 grid cells on the MACC14 grid	average closest 4 grid cells	Nearest Neighbour (one grid)	Core city masks: cameo_city- core_masks_v2. nc The city masks correspond to city-cores, as defined on the Urban Audit 2021. City masks and area weights for 79 of the 80 cities covered on our CAMS2_71 SR forecast (London was not included in Urban Audit 2021).
Initial conditions/ restart file	From corresponding SR forecast for the previous day	From the previous day forecast. Initially IFS-COMPO	From the previous day forecast. Initially IFS-COMPO	Two weeks spinup, starting from a 3d interpolated IFS global
Boundary conditions	From VRA 2019 (45r1 whole year) CAMS-GLOBAL 12 UTC forecast for previous day	CAMS-Global 12 UTC forecast for previous day (for PM only dust, no SS, no SO ₄ ²⁻ etc) For dust we use our SDS-WAS	CAMS-Global IFS	VRA2019 (and 2018) from IFS45r1 Boundary conditions for ozone are NOT reduced for

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	CAMS2_71: EMEP (in 2023)	CAMS2_71: LOTOS-EUROS (in 2023)	CAMS2_71: CHIMERE (in 2023)	Agreed for the CAMEO experiment
		run instead of IFS		LOTOS-EUROS or EMEP for the city SR. For sector SR, ozone has been reduced in EMEP model runs.
Source allocation method	Brute force, 15% reductions of NO _x , SO _x , VOC, NH ₃ , CO, PM at the same time (country, and pairs of cities).	Tagging, source sectors and/or countries	ACT surrogate model based on brute force reductions	EMEP: Sensibilities (LF) and brute force (15%) LOTOS- EUROS: Tagging and brute force (15%) CHIMERE: 15% and 100% emission reduction results from ACT
Species	PM _{2.5} , PM ₁₀ , NO ₂ MDA8, O₃	PM _{2.5} , PM ₁₀	PM _{2.5} , PM ₁₀ , NO ₂ , MDA8O3, O ₃	PM _{2.5} , PM ₁₀ , PM components, NO ₂

For the source sector runs, a more detailed version of the CAMSv6.1 anthropogenic emissions for 2018 and 2019 has been made available to the modeling teams. This version includes fuel specific information for certain GNFR sectors.

The source sector attribution with the LOTOS-EUROS and EMEP models has been performed for the sectors as provided in Table 3.2.2.

Table 3.2.2 Overview of source sector setup in LOTOS-EUROS and EMEP in the CAMEO exercise.

Attributed sectors LOTOS-EUROS / EMEP	GNFR (sub) sectors
Agriculture	K_AgriLivestock L_AgriOther
Industry Biomass	B_Industry
Industry Other	B_Industry
Industry Other Combustion	B_Industry

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Power Plant Biomass	A_PublicPower
Power Plant Other	A_PublicPower
Residential Biomass	C_OtherStationary Comb (residential only)
Residential Other	C_OtherStationary Comb (residential only)
Traffic Exhaust	F1/2/3_RoadTransport _exhaust
Traffic Non-exhaust	F4_RoadTransport _non-exhaust
Shipping	G_Shipping
Other Sectors	D_Fugitive; I_Offroad; H_Aviation; J_Waste; E_Solvent; M_other
Natural	Not applicable; (Sea Salt and Dust)
Wildfire	Not applicable; gfas-hourly
boundary and initial conditions (BIC)	Not applicable

The source sector attribution with the CHIMERE model and its ACT tool has been performed for the sectors as provided in Table 3.2.3.

Table 3.2.3 Overview of source sector setup in CHIMERE in the CAMEO exercise.

Attributed sectors CHIMERE	GNFR (sub) sectors
Agriculture	K_AgriLivestock; L_AgriOther
Industry	A_PublicPower, B_Industry; D_Fugitive
Residential	C_OtherStationary Comb
Traffic	F_RoadTransport
Shipping	G_Shipping
Other Sectors	I_Offroad; H_Aviation; J_Waste; E_Solvent; M_other

3.3 Evaluation of the EMEP and LOTOS-EUROS models

In the following sections, country-to-city and sector-to-city SRs for EMEP and LOTOS-EUROS (and partly for CHIMERE) will be compared. As a first step in this comparison, we evaluated the results from the EMEP and LOTOS-EUROS models against observations. A full evaluation is available from the website

<https://aeroval.met.no/pages/evaluation/?project=cameo&experiment=CAMEO+2019-reporting>

Figure 3.3.1 shows the comparison of PM_{10} , $PM_{2.5}$ and its chemical components towards EMEP observations for 2019. Both of the models have a very small negative bias of $PM_{2.5}$ towards observations (-6% for EMEP and -2% for LOTOS-EUROS). Figure 3.3.2 shows the bias of EMEP and LOTOS-EUROS results for $PM_{2.5}$ towards all EEA observations for 2019, showing a similar picture. However, LOTOS-EUROS overestimates nitrate (38%), EC (46%) and sea salt in $PM_{2.5}$ (79%), and is slightly less underestimated than the EMEP model for sulfate and organic carbon. Overall, it is higher for all chemical components. The reason for the similar comparison to $PM_{2.5}$, despite all chemical components being higher in LOTOS-EUROS, is probably related to the inclusion of PM water in the EMEP model results, whilst $PM_{2.5}$ in LOTOS-EUROS is dry. The reference measurement method for EMEP observations is gravimetric where mass is collected on filters that are conditioned to relative humidity 50% and the EMEP model calculates associated PM water at 50% relative humidity. For the EEA measurements, it is less clear to which extent the $PM_{2.5}$ mass includes water. However, if the CTM does not include PM water, this should in principle lead to an underestimation

In Figure 3.3.3, a comparison of the seasonal cycles in the two models against observations is presented. LOTOS-EUROS underestimates $PM_{2.5}$ in summer, but this is compensated by an overestimation in winter. The underestimation in summer is at least partly due to the lack of SOA in the LOTOS-EUROS model, and the comparison of OA in LE to observations of OA clearly demonstrates this underestimation in summer (RI-urbans project - not shown). New developments with inclusion of a VBS routine for SOA production in LOTOS-EUROS show a decrease of OA in winter and a small increase in summer (private communication).

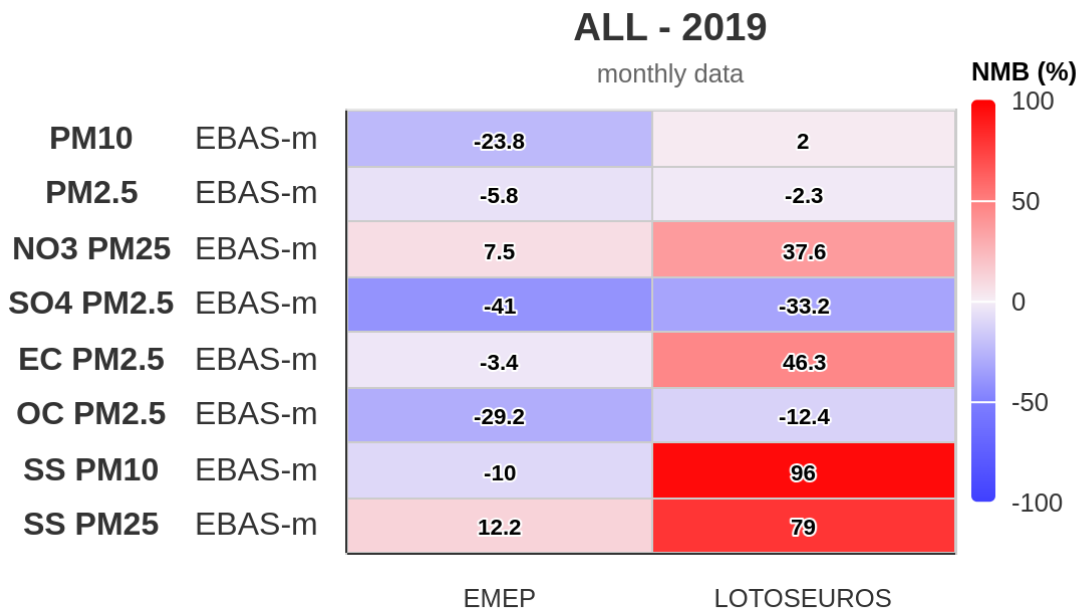
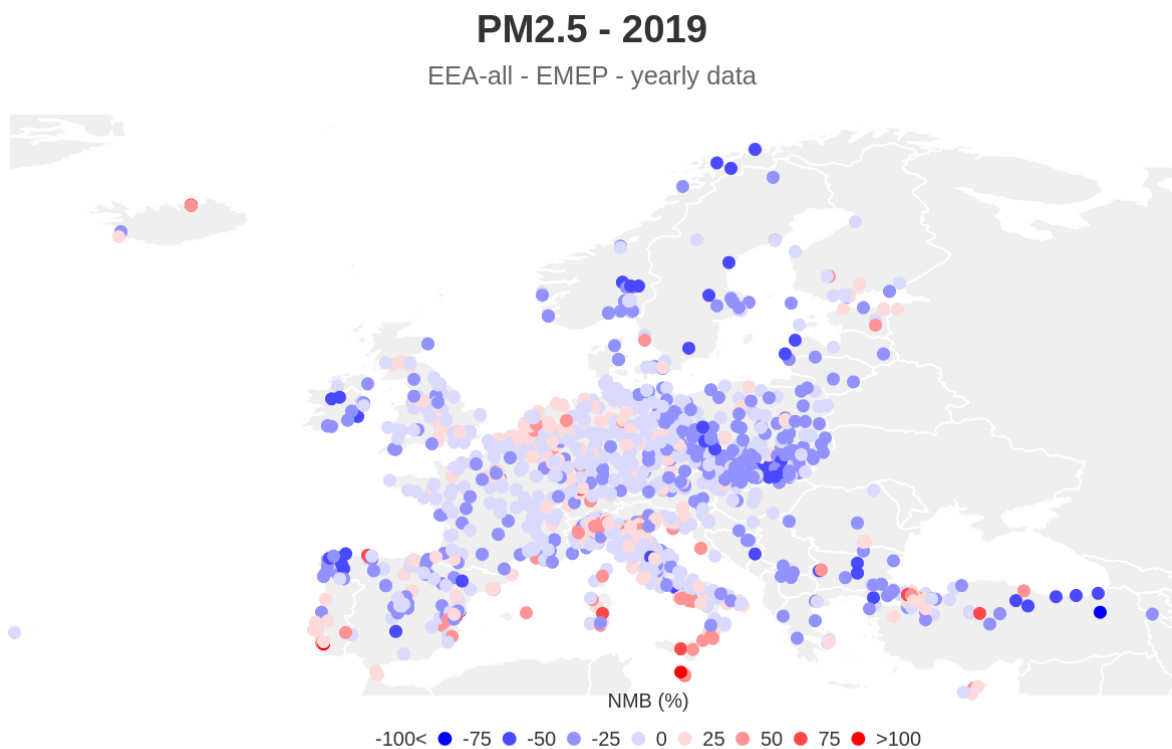


Figure 3.3.1. Heatmap (NMB, %) of the comparison of EMEP and LOTOS-EUROS model results to EMEP observations for 2019.



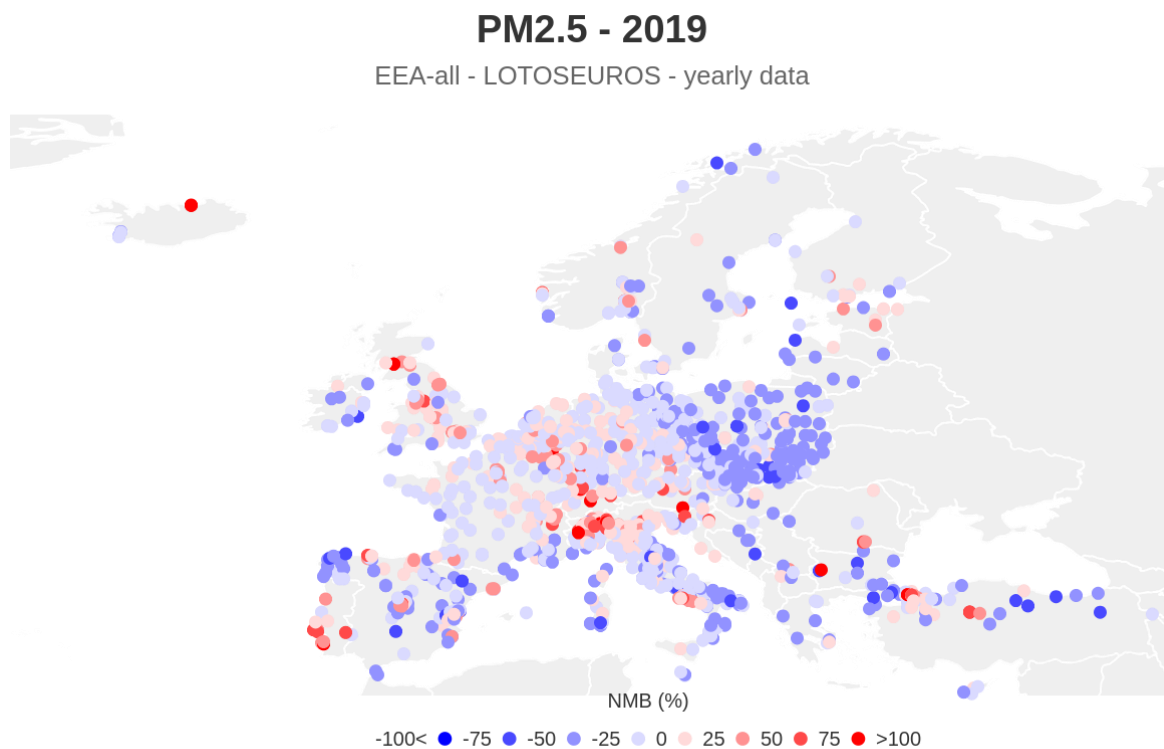


Figure 3.3.2. Bias (%) in PM_{2.5} towards all EEA observations of PM_{2.5}. Top:EMEP model results and bottom: LOTOS-EUROS model results

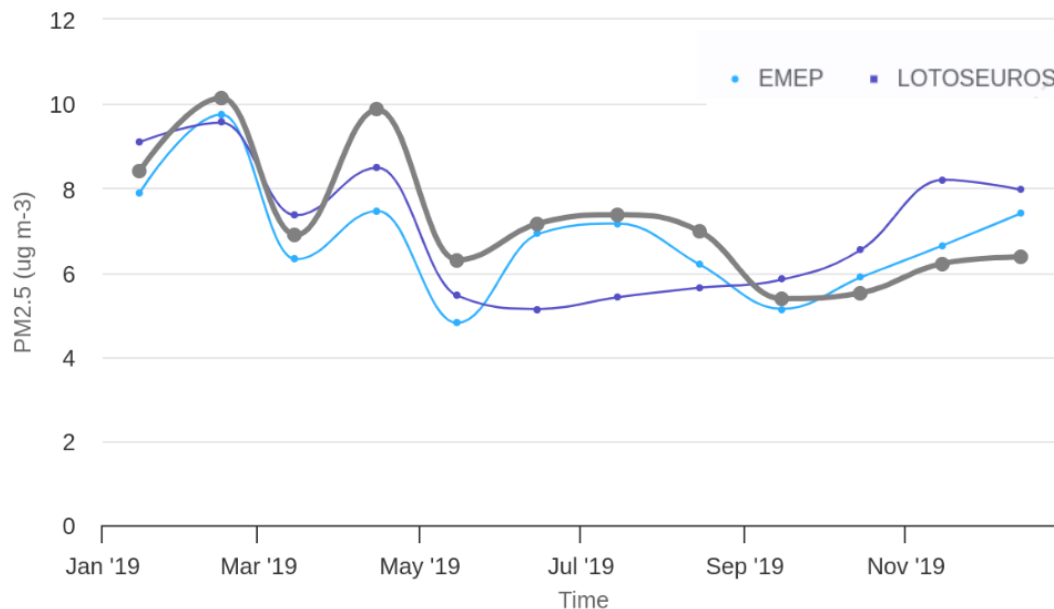


Figure 3.3.3. Comparison of (monthly average) EMEP observations, EMEP (light blue) model and LOTOS-EUROS (dark blue) model for PM_{2.5} for 2019.

3.4 City source receptor calculations for PM

In the CAMS Policy Service, the EMEP and LOTOS-EUROS models are used to infer spatial allocation. Their simulation setup described in Section 3.2 has been used to quantify the contributions of emissions from different countries to 79 cities in Europe for 2019.

Two different methodologies have been used for each model:

- EMEP Model : brute force (BF) and local fractions (LF)
- LOTOS-EUROS: BF and labelling

Since the two models are set up in (almost) exactly the same way, differences between EMEP BF and LOTOS-EUROS BF are due to different **model formulations**, whilst differences between EMEP LF and BF, or LOTOS-EUROS BF and labelling, are due to different **source apportionment methodology**.

3.4.1 Results

In Figure 3.4.1 we show results for 8 cities for country contributions to PM_{2.5} concentrations in 2019. The host country's contribution to the city is similar between different SA methodologies (i.e. EMEP BF versus EMEP LF, and LOTOS-EUROS BF versus LOTOS-EUROS labelling), but it varies between the models. For instance, in Madrid the contribution from Spain is significantly higher in LOTOS-EUROS than in the EMEP model. However, total PM_{2.5} is also higher in LOTOS-EUROS. In Figure 3.4.2 we present contributions in % of the total concentrations, and although the results between the models are more similar, LOTOS-EUROS still shows a higher host country contribution than EMEP.

In Figure 3.4.3 we compare the first, second and third largest contribution (as decided by EMEP BF) to 79 cities for dry PM_{2.5}. The left plots (a and d) compares results for EMEP BF and LF and shows that the results are almost identical between the two SA methodologies. The plots to the right (c and f) shows a comparison of LOTOS-EUROS BF and labelling methodology. Although not as indistinguishable as EMEP BF and LF, the results are similar. The middle plots (b and e) compares results from EMEP and LOTOS-EUROS BF and show larger differences. LOTOS-EUROS BF has significantly higher host country contributions than EMEP BF, up to 20 percentage points higher contributions. For second and third largest contributions, the differences are smaller.

Figure 3.4.4 presents the comparison of the highest daily contributions. The spread is larger than for the yearly means in Fig 3.4.3, but similar to the yearly comparison EMEP BF and LF are very similar. LOTOS-EUROS BF compares well to LOTOS-EUROS labelling, whilst LOTOS-EUROS BF is consistently larger than EMEP BF.

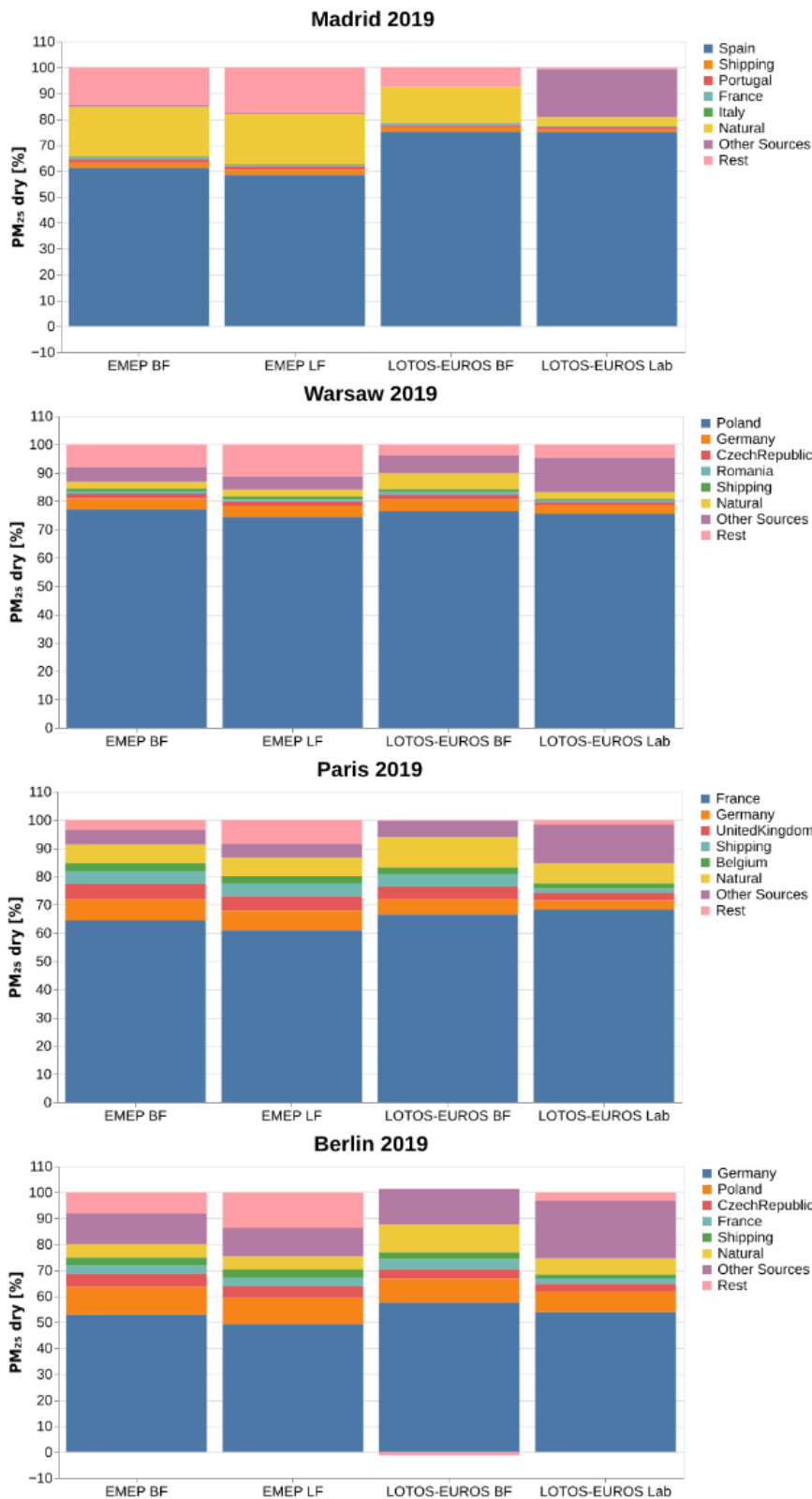


Fig 3.4.1 Country contributions ($\mu\text{g}/\text{m}^3$) to $\text{PM}_{2.5}$ air pollution (dry PM) in Madrid, Warsaw, Paris, Berlin, Oslo, Zagreb, Bremen, Århus, calculated with EMEP brute force (BF), EMEP local fraction (LF), LOTOS-EUROS BF and LOTOS-EUROS labelling (Lab). Note that for TNO Lab, 'Other' also includes natural contributions from BIC (as it was not possible to separate out).

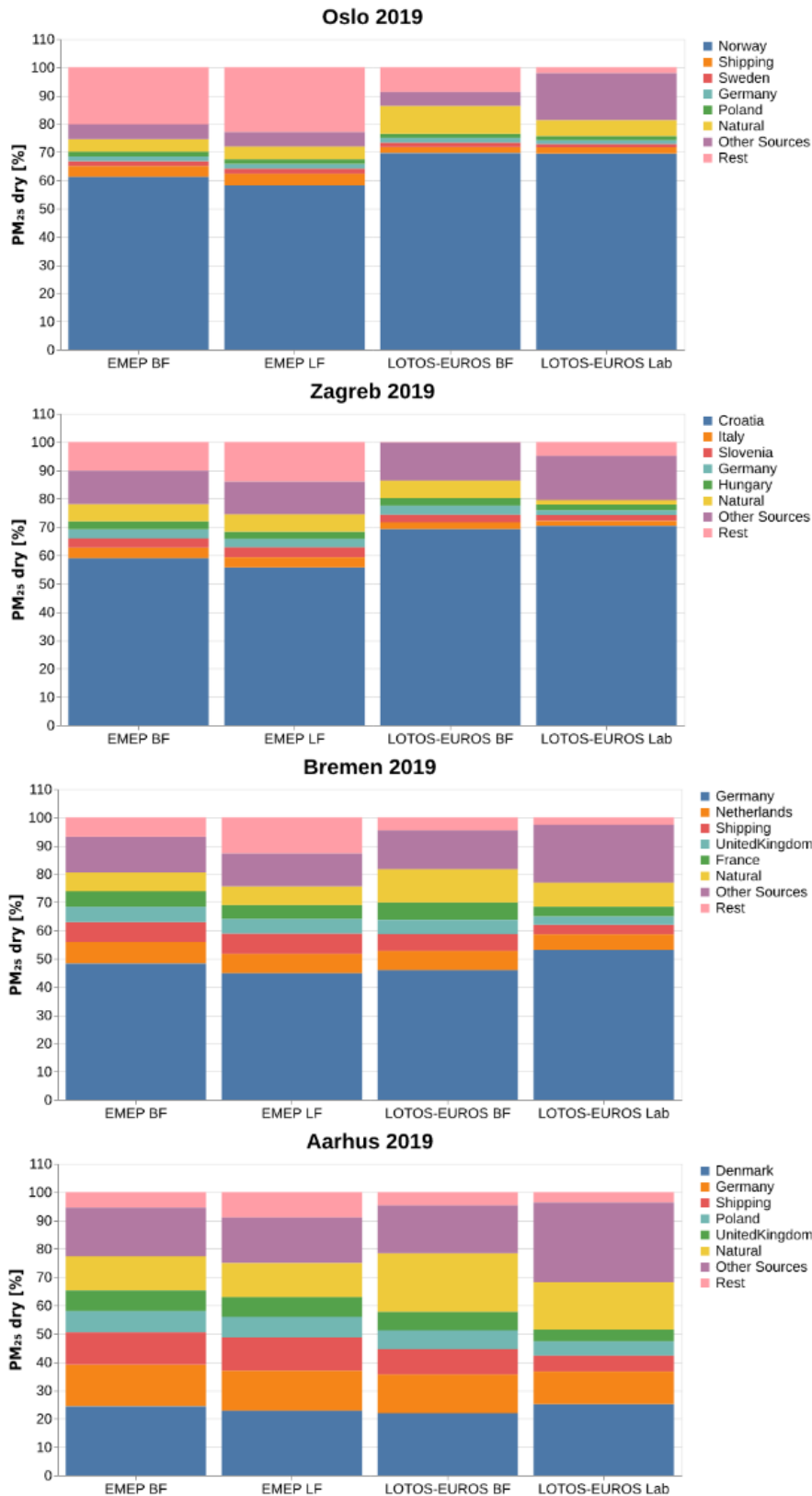


Fig 3.4.1 continued: Country contributions ($\mu\text{g}/\text{m}^3$) to $\text{PM}_{2.5}$ air pollution (dry PM) in Madrid, Warsaw, Paris, Berlin, Oslo, Zagreb, Bremen, Århus, calculated with EMEP brute force (BF), EMEP local fraction (LF), LOTOS-EUROS BF and LOTOS-EUROS labelling (Lab). Note that for TNO Lab, 'Other' also includes natural contributions from BIC (as it was not possible to separate out).

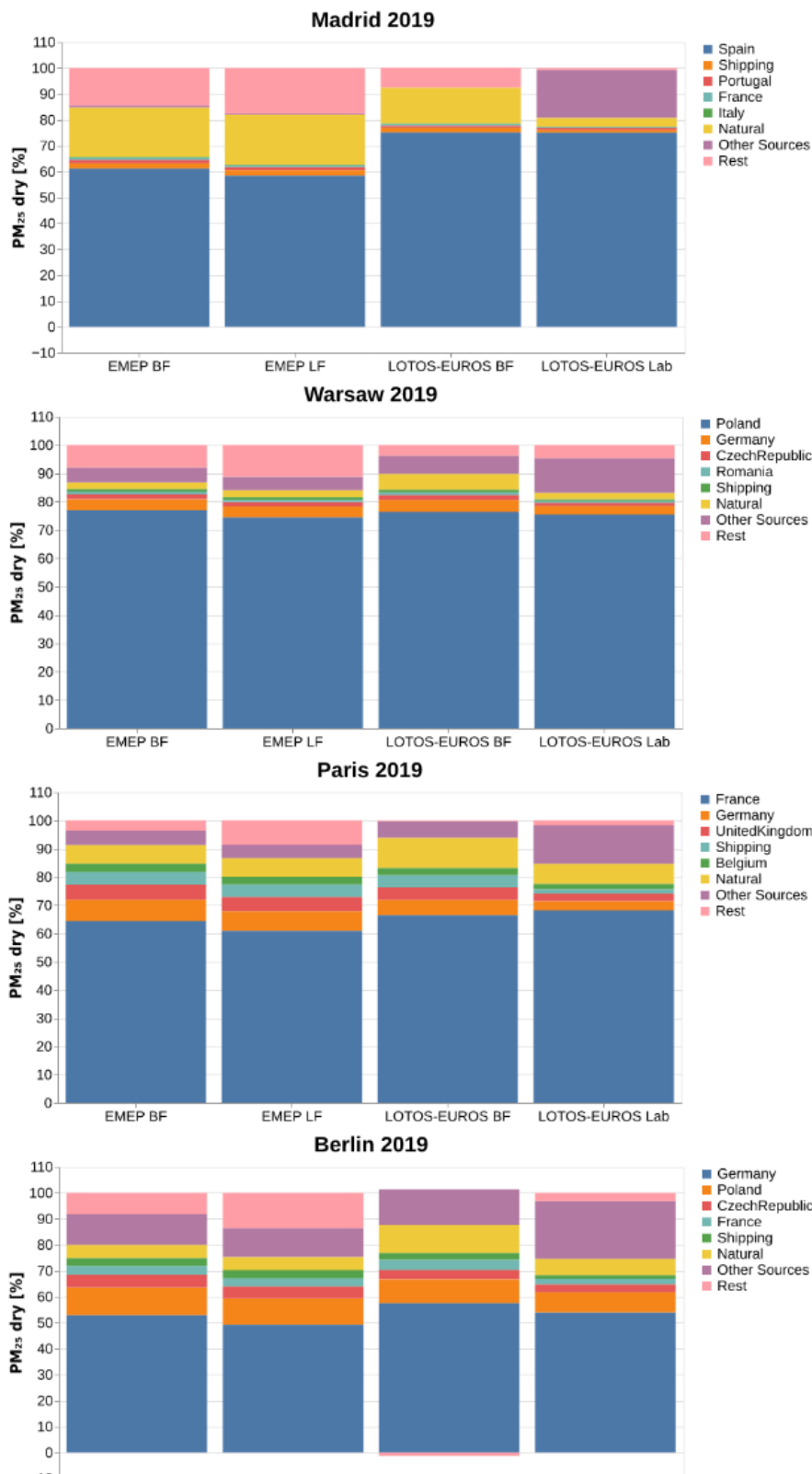


Fig 3.4.2: Relative country contributions(%) to PM_{2.5} air pollution (dry PM) in Madrid, Warsaw, Paris, Berlin, Oslo, Zagreb, Bremen, Århus, calculated with EMEP brute force (BF), EMEP local fraction (LF), LOTOS-EUROS BF and LOTOS-EUROS labelling (Lab). Note that for TNO Lab, 'Other sources' also includes natural contributions from BIC (as it was not possible to separate out)

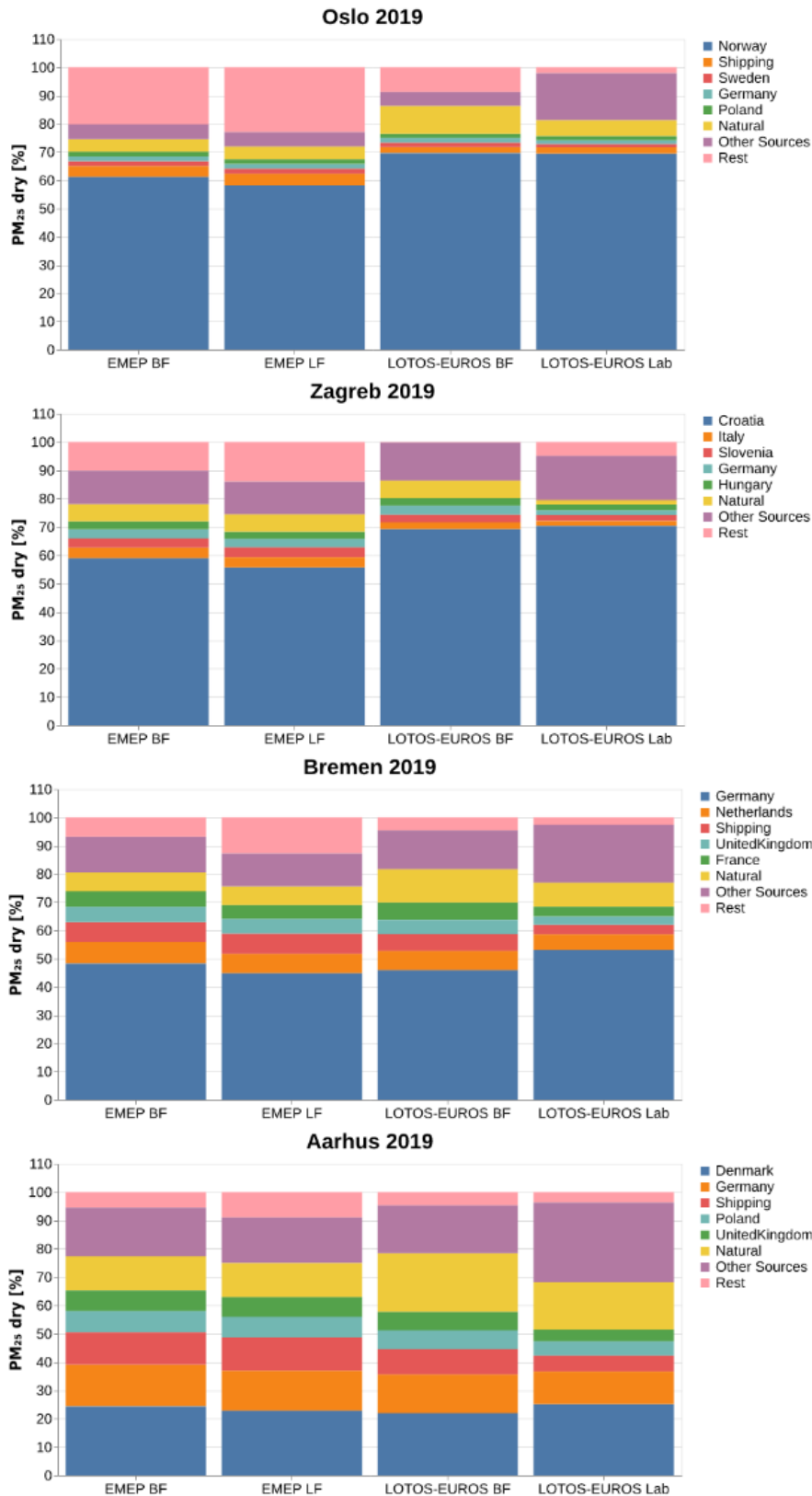


Fig 3.4.2 continued: Relative country contributions(%) to PM_{2.5} air pollution (dry PM) in Madrid, Warsaw, Paris, Berlin, Oslo, Zagreb, Bremen, Århus, calculated with EMEP brute force (BF), EMEP local fraction (LF), LOTOS-EUROS BF and LOTOS-EUROS labelling (Lab). Note that for TNO Lab, 'Other sources' also includes natural contributions from BIC (as it was not possible to separate out)

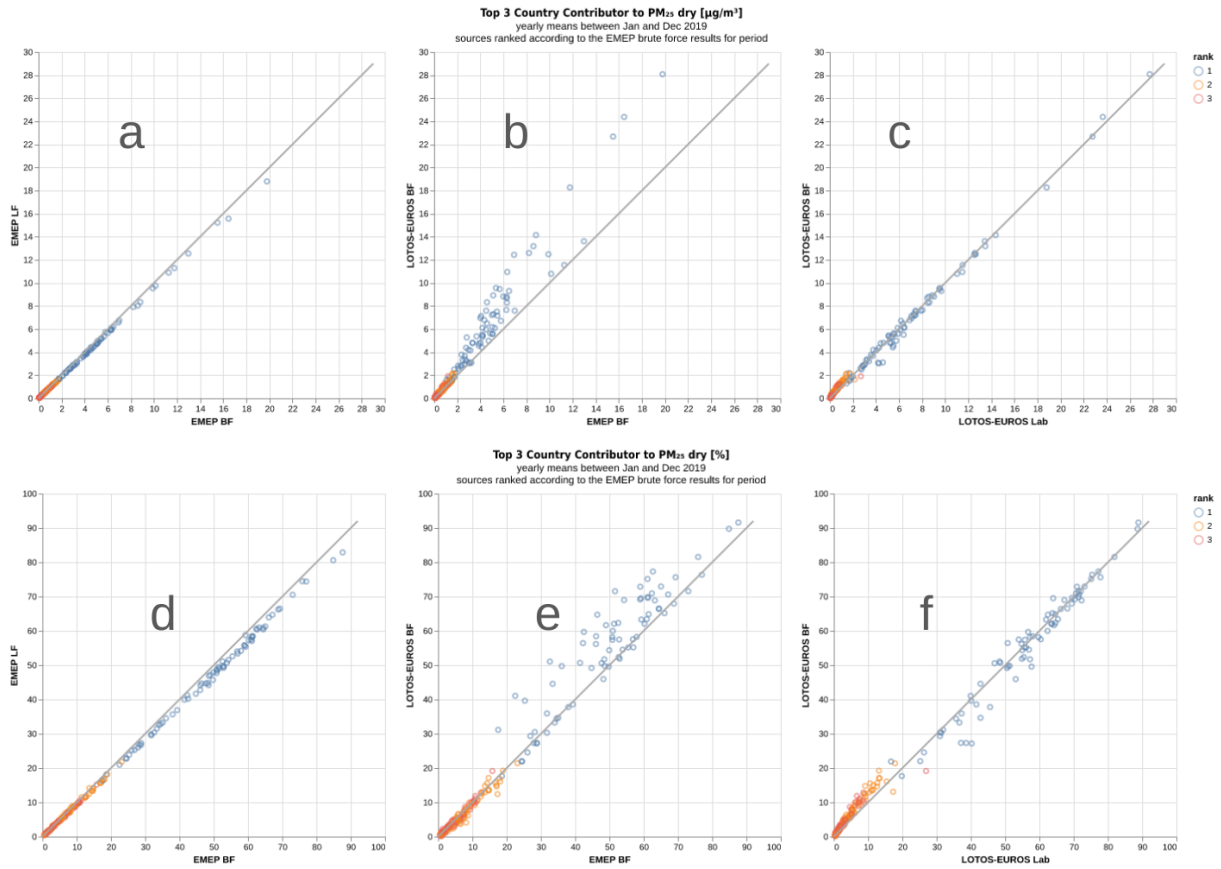


Figure 3.4.3: Comparison of the 3 largest country contributions to PM_{2.5} (top: $\mu\text{g}/\text{m}^3$, bottom: %) in 79 cities in Europe as decided by the EMEP BF model. EMEP BF versus LF (a, d), EMEP BF versus LOTOS EUROS BF (b, e), LOTOS EUROS Labelling versus BF (c, f).

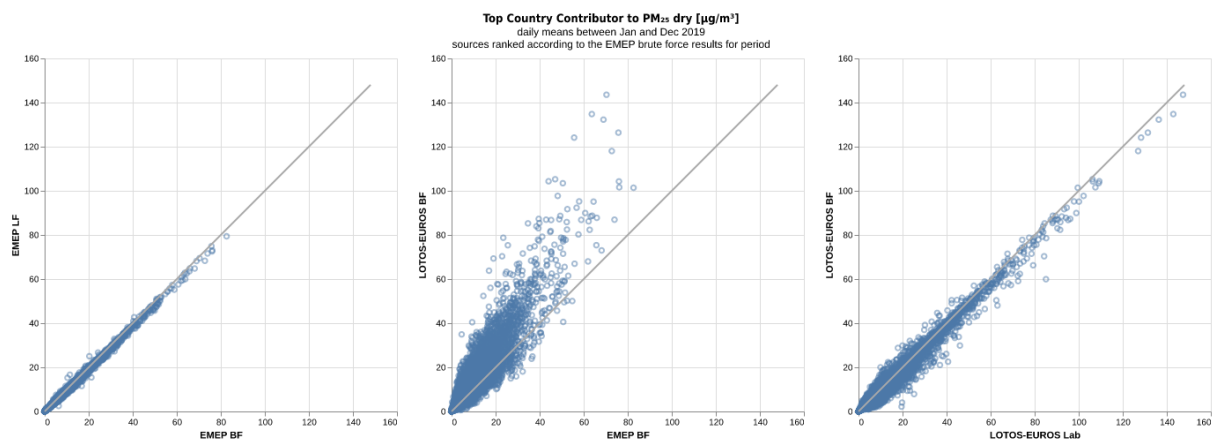


Figure 3.4.4: Comparison of the largest (anthropogenic) contribution to PM_{2.5} in 79 cities in 2019, daily means.

In order to understand better why EMEP and LOTOS-EUROS BF results differ significantly, we have analyzed contributions from different chemical components. In Figure 3.4.5, we compare BF results for the 3 largest country contributors (as decided by EMEP BF) to concentrations in each of the 79 cities for different components; Primary organic matter (POM), Elemental carbon (EC), ammonium (NH_4^+), nitrate (NO_3^-) and sulfate (SO_4^{2-}).

While the secondary inorganic components nitrate and ammonium (and to some extent sulfate) are well correlated and of the same magnitude, the contributions from primary components (EC and POM) are significantly higher in LOTOS-EUROS.

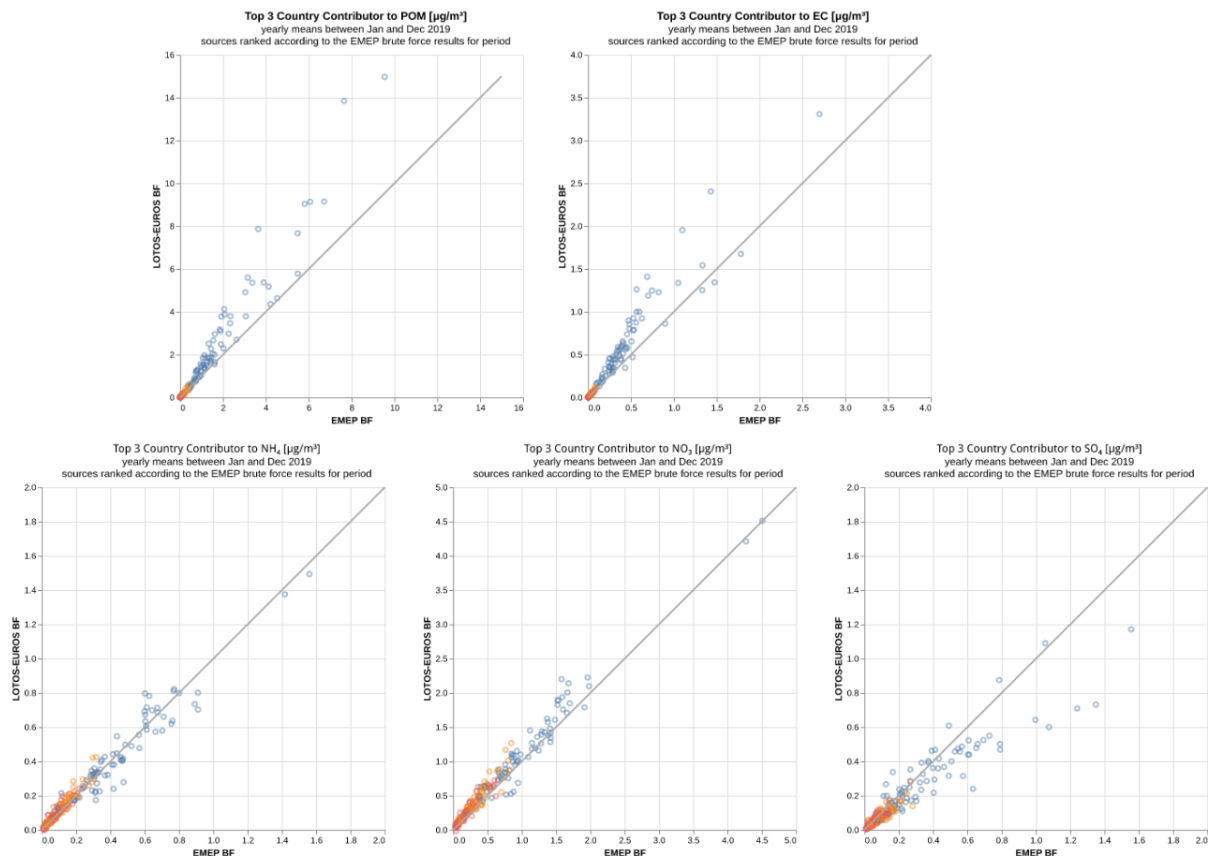


Figure 3.4.5 Comparison of EMEP and LOTOS-EUROS BF results for 79 cities ($\mu\text{g}/\text{m}^3$). The 3 largest country contributors (as decided by EMEP BF) to concentrations in each of the 79 cities for different components; Primary organic matter (POM), Elemental carbon (EC), ammonium, nitrate and sulfate.

In Section 3.5, we show that these results are consistent with the results on sectoral source apportionment, where the LOTOS-EUROS model shows consistently larger contributions from residential heating. This is consistent with the fact that the largest differences seen in host country contributions in Figure 3.4.1 and 3.4.2 are for cities which have large residential heating emissions. A possible reason for the difference between EMEP and LOTOS-EUROS results for the residential heating contribution is the difference in the depth of the lowest layer, which is 20 meter in LOTOS-EUROS and 50 meter in EMEP. The residential heating emissions are released into the lowest layer and assumed to be instantaneously well mixed. With a 20 meter layer, concentrations are higher than when instantaneously mixed within a 50 meter layer, and although emissions are vertically mixed this process is not equally fast. In Chapter 4, where

we discuss the difference between SRs done with the EMEP model in CAMS and in the LRTAP Convention, we also hypothesize that the difference in the surface layer between the two model versions of the EMEP model plays a key role for the differences found in the results.

3.4.2 Quantification of differences due to model formulation and source attribution methodology

In order to quantify the differences in source attribution (SA) in cities due to different methodologies, we have calculated the root mean square error (RMSE) of (daily mean) EMEP LF versus EMEP BF, and the RMSE of (daily mean) LOTOS-EUROS BF versus labelling. These two RMSEs give an estimate of the differences due to different SA methodologies. Similarly, we have quantified the difference due to model formulations by calculating the RMSE of (daily mean) EMEP BF versus LOTOS-EUROS BF. The RMSEs for the SA methodologies can be compared to the RMSE for model formulations in order to answer what is the most important reason for the differences: SA methodology or model formulation. The results of this analysis are shown in Figure 3.4.6 for different components. Only the results for RMSE of model formulations (EMEP BF versus LOTOS-EUROS BF) compared to RMSE of LOTOS-EUROS BF versus labelling are shown, as the results for EMEP BF and LF are very similar.

The two top plots show POM (top left) and EC (top right) respectively. As expected, the RMSE due to SA methodology is very small (in principle it should be zero, since the primaries are linear and thus the different SA methodologies should give similar results) and differences are only due to numerical implementation of the methods. On the contrary, RMSE for the SIAs are rather large due to SA methodologies (the x-axis) and of the same magnitude as the differences due to model formulations for nitrate and sulfate.

In Figure 3.4.7, we compare RMSE (x-axis: LOTOS-EUROS labelling versus BF, y-axis: EMEP versus LOTOS-EUROS BF) based on daily means between January and December 2019 for $PM_{2.5}$ (left) and PM_{10} (right). It is clear that especially for the largest contributor, RMSE is much larger for model differences (EMEP versus LOTOS-EUROS) than differences in SA methodology. For LF versus BF SA methodology, the differences are even smaller (not shown).

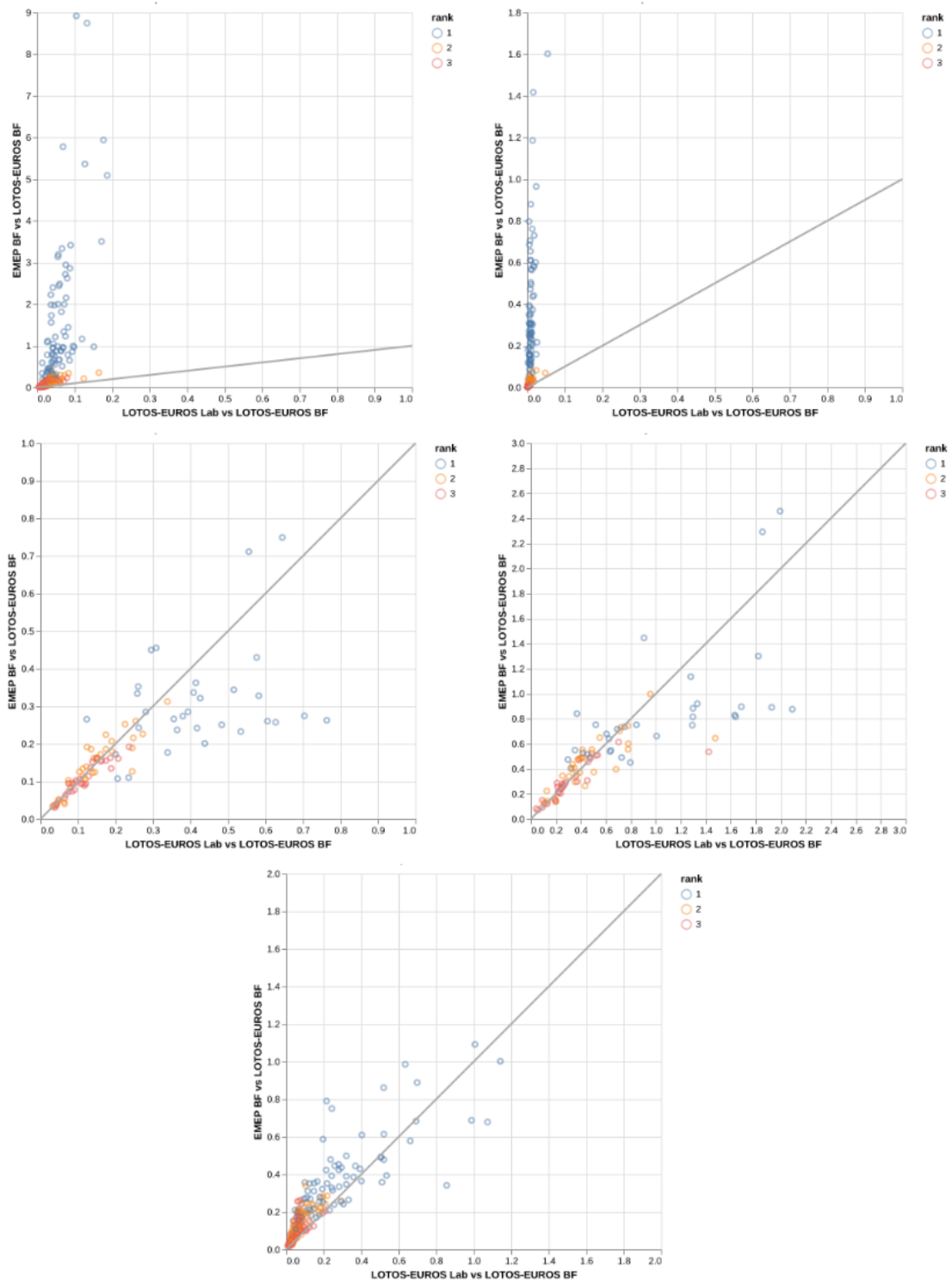


Figure 3.4.6: Comparison of RMSE (x-axis: LOTOS-EUROS labelling versus BF, y-axis: EMEP versus LOTOS-EUROS BF) based on daily means between January and December 2019 for POM (top left), EC (top right), NH₄⁺ (middle left), NO₃⁻ (middle right), SO₄²⁻ (bottom)

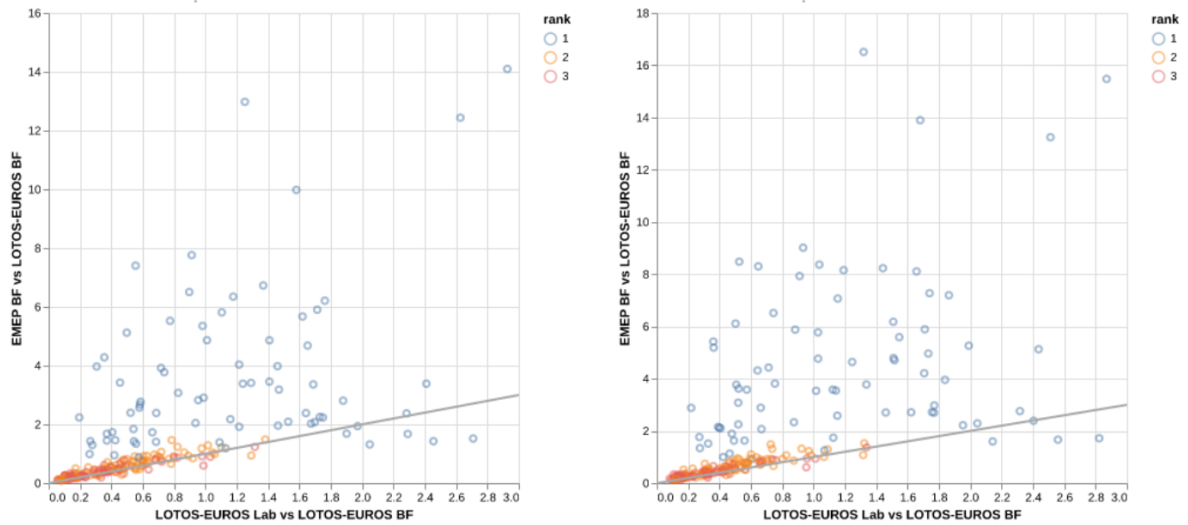


Figure 3.4.7 : Comparison of RMSE (x-axis: LOTOS-EUROS labelling versus BF, y-axis: EMEP versus LOTOS-EUROS BF) based on daily means between January and December 2019 for PM_{2.5} (left) and PM₁₀ (right).

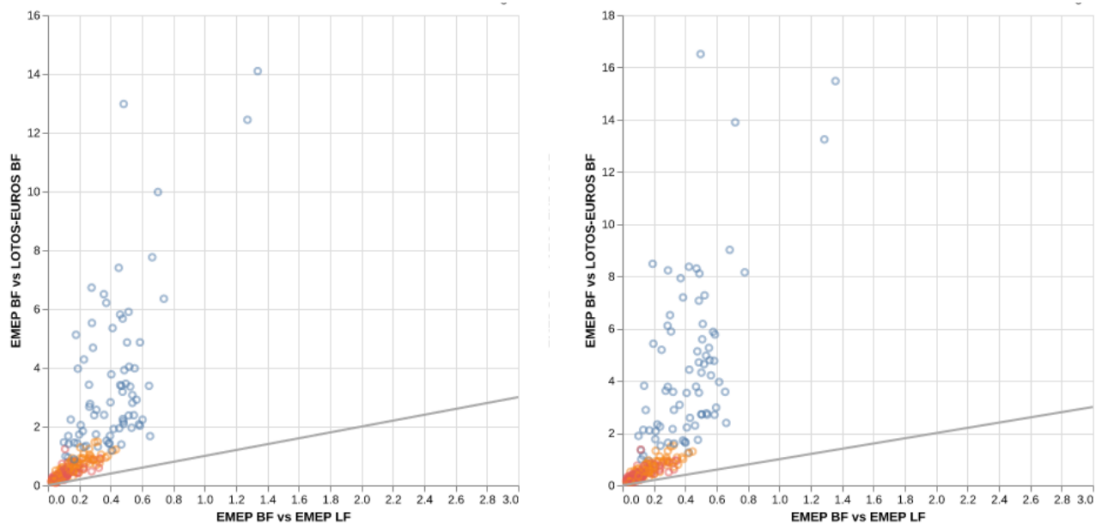
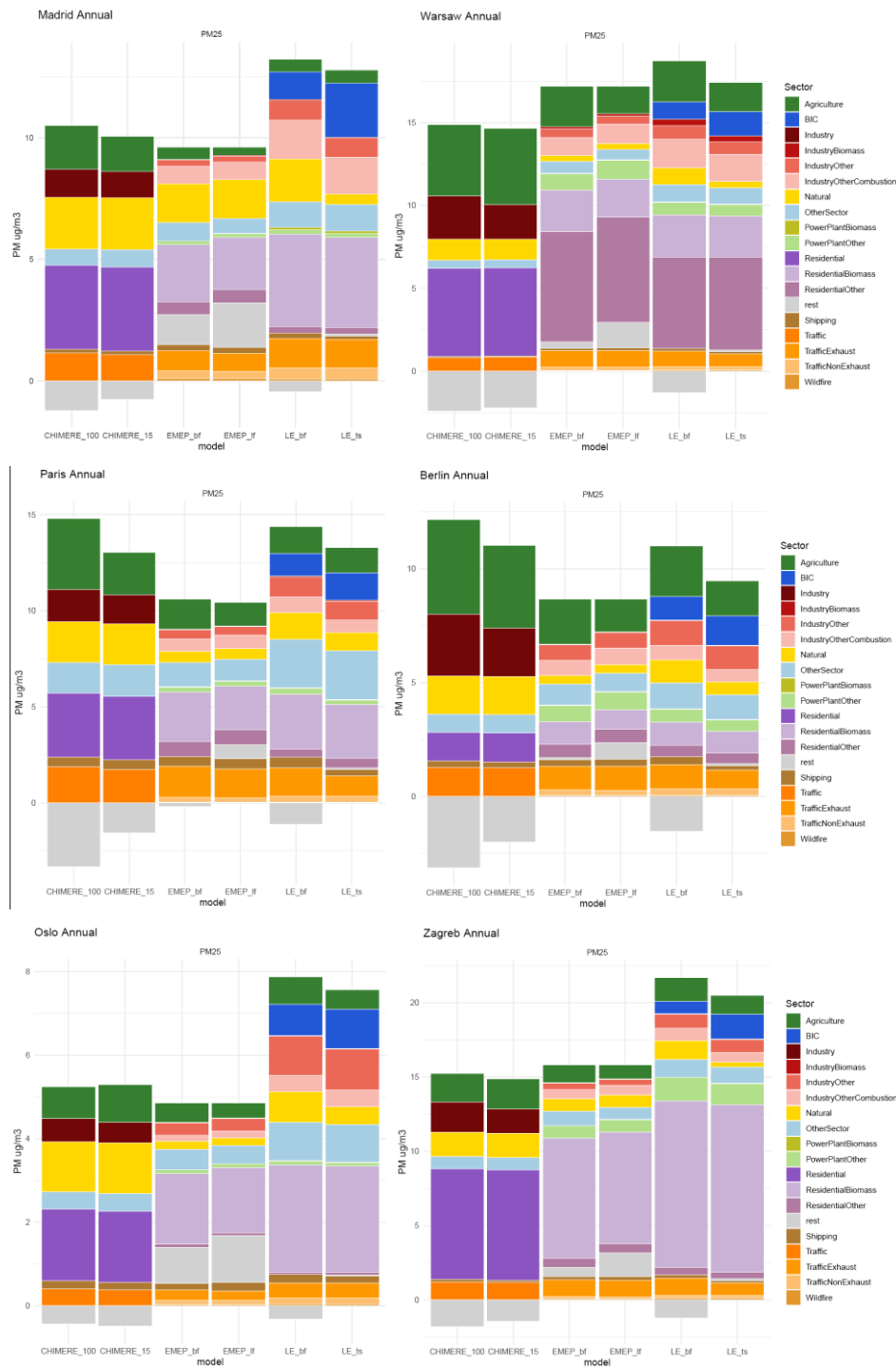


Figure 3.4.8 : Comparison of RMSE (x-axis: EMEP BF versus LF, y-axis: EMEP versus LOTOS-EUROS BF) based on daily means between January and December 2019 for PM_{2.5} (left) and PM₁₀ (right).

3.5 Results for sector source receptor calculations



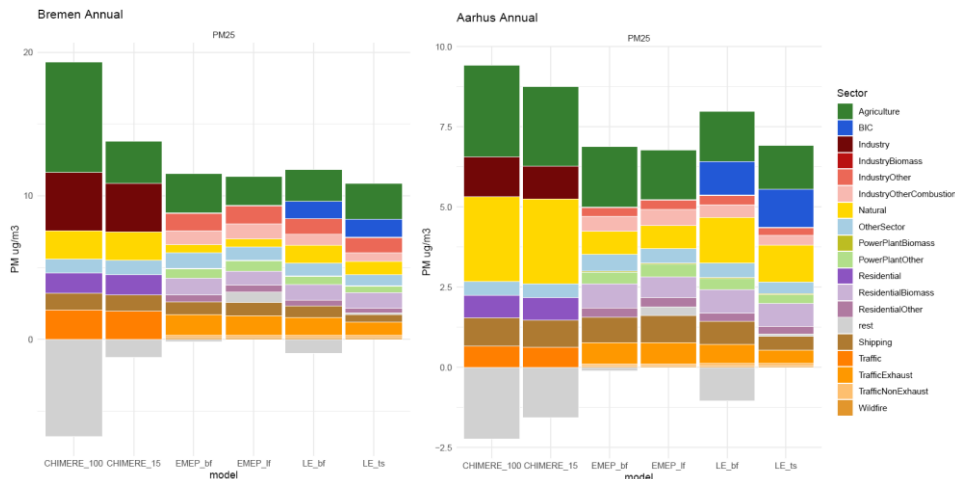


Fig 3.5.1: Sector contributions ($\mu\text{g}/\text{m}^3$) to $\text{PM}_{2.5}$ air pollution (dry PM) in Madrid, Warsaw, Paris, Berlin, Oslo, Zagreb, Bremen, Århus, calculated with CHIMERE-ACT 100% and 15% reductions (CHIMERE_100 and CHIMERE_15), EMEP brute force (EMEP_bf), EMEP local fraction (EMEP_lf), LOTOS-EUROS brute force (LE_bf) and LOTOS-EUROS labelling (LE_ts). Note that for LOTOS-EUROS labelling, 'BIC' also includes natural contributions from BIC (as it was not possible to separate out). Note that for CHIMERE industry also contains the powerplant contributions.

In Figure 3.5.1 we show results for 8 cities for sector contributions to annual $\text{PM}_{2.5}$ concentrations. For most cities the differences between the three models is larger than the difference between the source attribution methods applied within one model. Note that the EMEP BF and LF results have excluded the BIC contributions, due to large values from changes in the ozone boundary conditions. New runs will be performed with fixed ozone to determine the BF BIC contribution. Within the LOTOS-EUROS labelling results 'BIC' also includes natural contributions from the boundaries (as they were not explicitly separated into a fine and coarse fraction).

Some of the main differences seen between the models are:

- LOTOS-EUROS is giving larger contributions from Industry in Warsaw, Oslo and Madrid than EMEP and CHIMERE. This has been related to a small difference in the applied altitude profile of industrial emissions. Within LOTOS-EUROS the emissions from industrial sources are identified as area sources instead of point sources and inserted in the model's surface layer, while within the EMEP and CHIMERE models these emissions are distributed over different altitudes similar to the point sources.
- Natural contributions show large differences for some of the cities. In general CHIMERE and LOTOS-EUROS show larger contributions than EMEP. Note that part of the natural contributions is contained in the BIC contribution for LOTOS-EUROS. As mentioned before LOTOS-EUROS is modeling higher sea salt concentrations over land than the EMEP model. The deposition scheme for particles in the LOTOS-EUROS model is currently being reviewed as a possible cause for the overestimation of sea salt concentrations.
- Large differences in the residential biomass contributions between the EMEP and LOTOS-EUROS models can be seen for some of the cities (Madrid, Zagreb, Oslo). This is connected to large differences in the modelled primary PM as shown in section 3.4.1. The largest differences are for cities which have large residential heating emissions. Residential emissions are inserted into the surface layer of each model.

Since these emissions are usually taking place during cold conditions with low vertical mixing, the resulting concentrations are very sensitive to the depth of the models surface layer, which is 20m in LOTOS-EUROS and 50m in EMEP. This could well explain a large part of the differences in this modelled contribution. Also differences in vertical mixing parametrisations in both models can influence the results. The results for the CHIMERE model are more in line with the EMEP model for Zagreb and Oslo.

The comparison to CHIMERE ACT is influenced by the lower level of details in the attributed sectors. Note that the results for CHIMERE act based on 100% emission reductions for the sectors lead to large contributions of for example the agriculture sector for some of the cities, which is not consistent with the 15% emission reductions applied in the BF calculations. For example in Bremen the agricultural contribution from CHIMERE-ACT for the 100% reduction is nearly three times as large as for the CHIMERE-ACT 15% reduction. This ACT 100% reduction results are closer to a zeroing-out experiment where the PM fraction is assessed which would remain if all agricultural emissions were removed. The ACT 100% reduction results are thus closer to an estimate of the potential impact of (total) agricultural emissions on PM, although it is quite unlikely (and even probably not desirable) that such emissions can be totally removed. Note that the step-by-step evaluation of each sector leads to some double counting of possible impacts from sources which together contribute to the formation of secondary inorganic aerosols. This is solved by complementing with a closure simulation where all emissions are removed. The residual (closure) between the latter and sum of each individual sector is labelled "rest" in the figure. That residual can be large and negative in some cases, such as in Bremen, which explains some of the differences noted compared to the EMEP and LOTOS-EUROS and CHIMERE-ACT 15% reductions source apportionment.

The different source attribution methods applied within one model (BF versus LF in EMEP, BF versus labelling in LOTOS-EUROS) provide similar main contributors to the annual concentrations. However when considering shorter averaging time periods, the differences between the source attribution methods become larger because of the larger influence of non-linearity. When comparing the average contributions to the monthly average $PM_{2.5}$ concentration in Utrecht for April (Figure 3.5.2) from the brute force versus the labelling in LOTOS-EUROS, one can see that the brute force contribution for agriculture is smaller than from labelling and the other way around for the traffic exhaust contribution. This is related to the non-linear process of ammonium nitrate formation from NO_x sources (mainly traffic) and NH_3 sources (mainly agriculture) as can be seen in the bottom plot of Figure 3.5.2. In April in the Netherlands there is an excess of NH_3 from agricultural activities, therefore 15% emission reductions will have a smaller impact than reductions of NO_x emissions. This is included in the brute force calculations showing the potential impact of emission reductions. In the labelling routine the actual contributions are determined instead of the impact of emission reductions, and the ammonium nitrate is equally assigned to the agricultural NH_3 source and the NO_x sources. The differences between the brute force and local fraction within the EMEP model is smaller because they are both representing impacts of emission reductions.

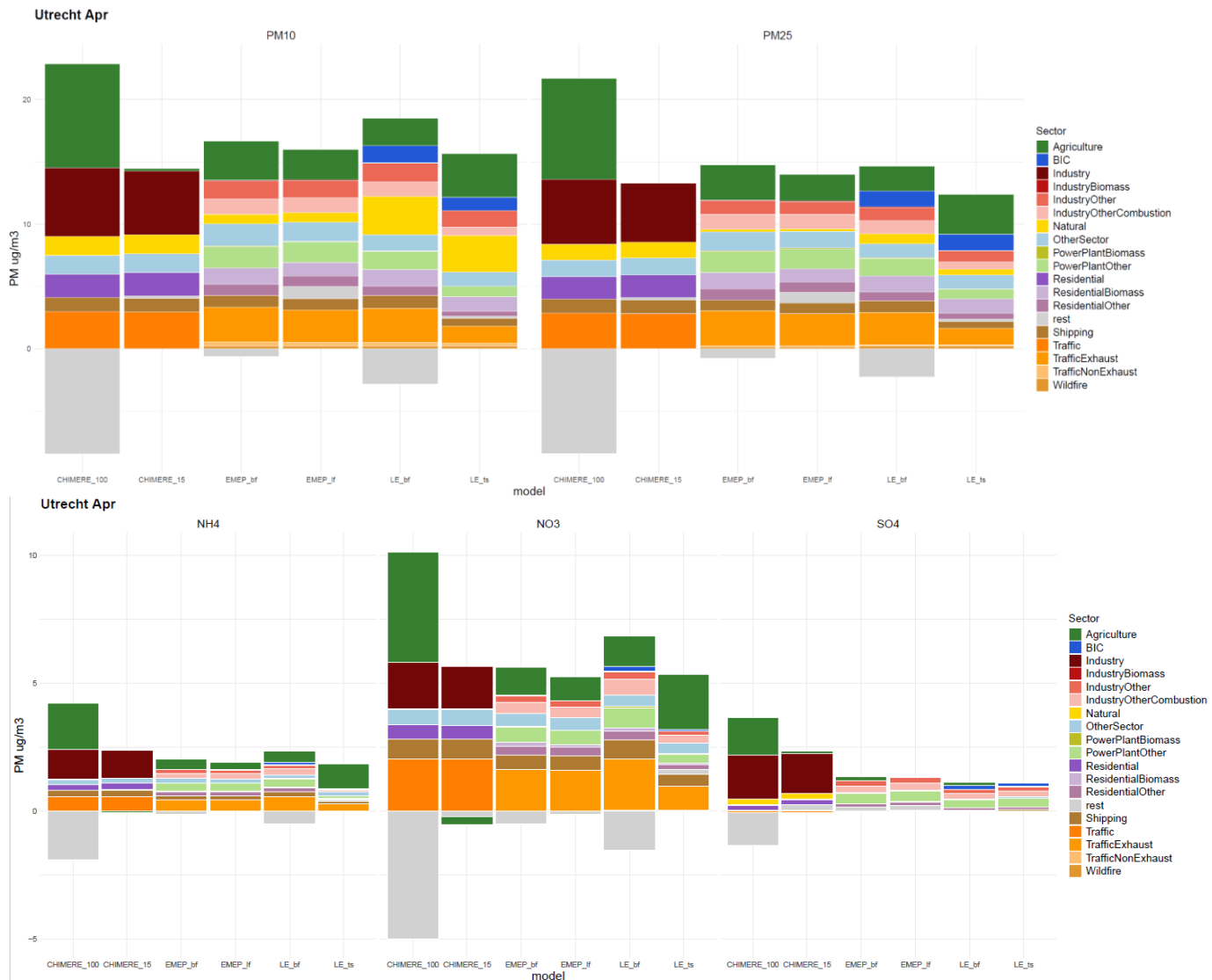


Fig 3.5.2: Sector contributions ($\mu\text{g}/\text{m}^3$) to PM_{10} and $\text{PM}_{2.5}$ air pollution (dry PM) (top plot) and Secondary Inorganic Aerosol (SIA) components NH_4^+ , NO_3^- and SO_4^{2-} (bottom plot) for Utrecht in April 2019 calculated with CHIMERE ACT 100% and 15% reductions (CHIMERE_100 and CHIMERE_15), EMEP brute force (EMEP_bf), EMEP local fraction (EMEP_lf), LOTOS-EUROS brute force (LE_bf) and LOTOS-EUROS labelling (LE_ts). Note that for LOTOS-EUROS labelling, 'BIC' also includes natural contributions from BIC (as it was not possible to separate out). Note that the CHIMERE industry also contains the powerplant contributions.

The impacts of non-linearity on source attribution results become more apparent when analysing daily values and are mostly affecting the sectors with high NO_x and NH_3 emissions. Figure 3.5.3 shows the differences between the brute force and labelling results in LOTOS-EUROS on a daily basis for three sectors. For the residential biomass sector the differences are very small due to the mostly primary nature of its emissions (EC, OC and other PPM). For agriculture and traffic however there are large differences between the two methods with variability depending on the location and day of the year related to the chemical regimes as explained above.

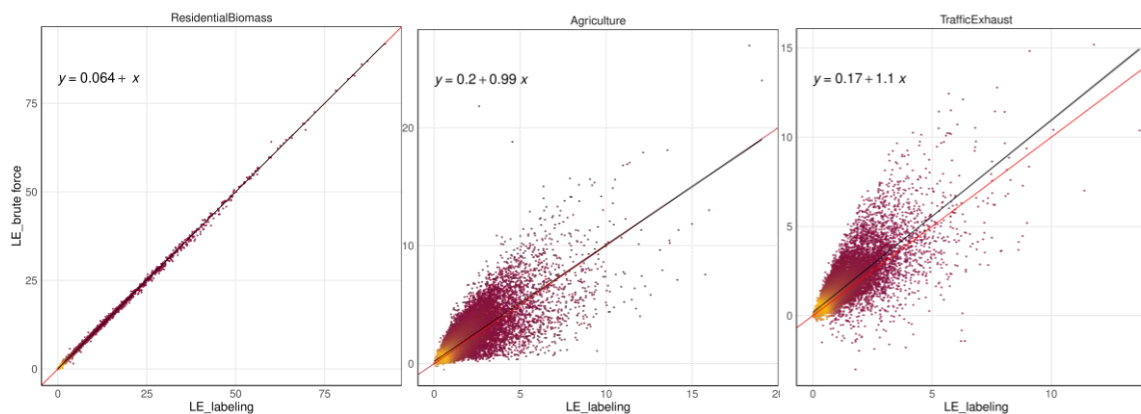


Fig. 3.5.3 Comparison of daily $PM_{2.5}$ contributions from LOTOS-EUROS BF versus LOTOS-EUROS tagging for 79 cities ($\mu\text{g}/\text{m}^3$) for sectors residential biomass (left column), agriculture (middle column) and traffic exhaust (right column).

In some cases the difference in the NH_3 sources contributions is compensating for the difference in the NO_x sources contribution, however the non-linear behaviour of secondary aerosol formation and chemistry also is the cause of the brute force attribution method not fulfilling the property of completeness (i.e. the sum of all source contributions adds up to the total concentration), as reflected in the closure term 'rest' in Figures 3.5.1 and 3.5.2.

This can also be seen in the time series of the difference in LOTOS-EUROS between the sum of contributions from labelling versus the sum of contributions from brute force (excluding the closure term) for Berlin in Figure 3.5.4. In Berlin the upscaling of the brute force reductions to 100% leads to a larger explained concentration than the actual modelled concentration for most days. The largest differences are seen during spring and fall as the summer temperatures are too high to sustain significant ammonium nitrate levels. This behavior can be explained through the formation of secondary inorganic aerosols. When there is no strong limiting regime, reducing either the agricultural NH_3 emissions or combustion related emissions will both lower the NH_4NO_3 levels. By upscaling the impacts of both NO_x and NH_3 emission reductions one is effectively double counting the impact. Simultaneous reduction of the respective sources will however not lead to an additive impact, and the sum of the contributions calculated separately will therefore be larger than the contribution when reduced together. For Utrecht in winter the upscaling of the BF results often leads to lower explained mass concentration than the total concentration. The Netherlands has relatively large NO_x emissions compared to Berlin in the winter months. This is largely explained by the traffic sector as shown in Figure 3.5.5, the traffic sector is the main contributor to NO_3 concentration. During stagnant conditions in winter when ozone levels are depleted a small reduction in NO_x emissions will hardly impact nitrate formation as there is no ozone available to perform the NO_x oxidation. Hence, the sensitivity run will show a small impact although a substantial level of nitrate formed earlier/elsewhere may be present.

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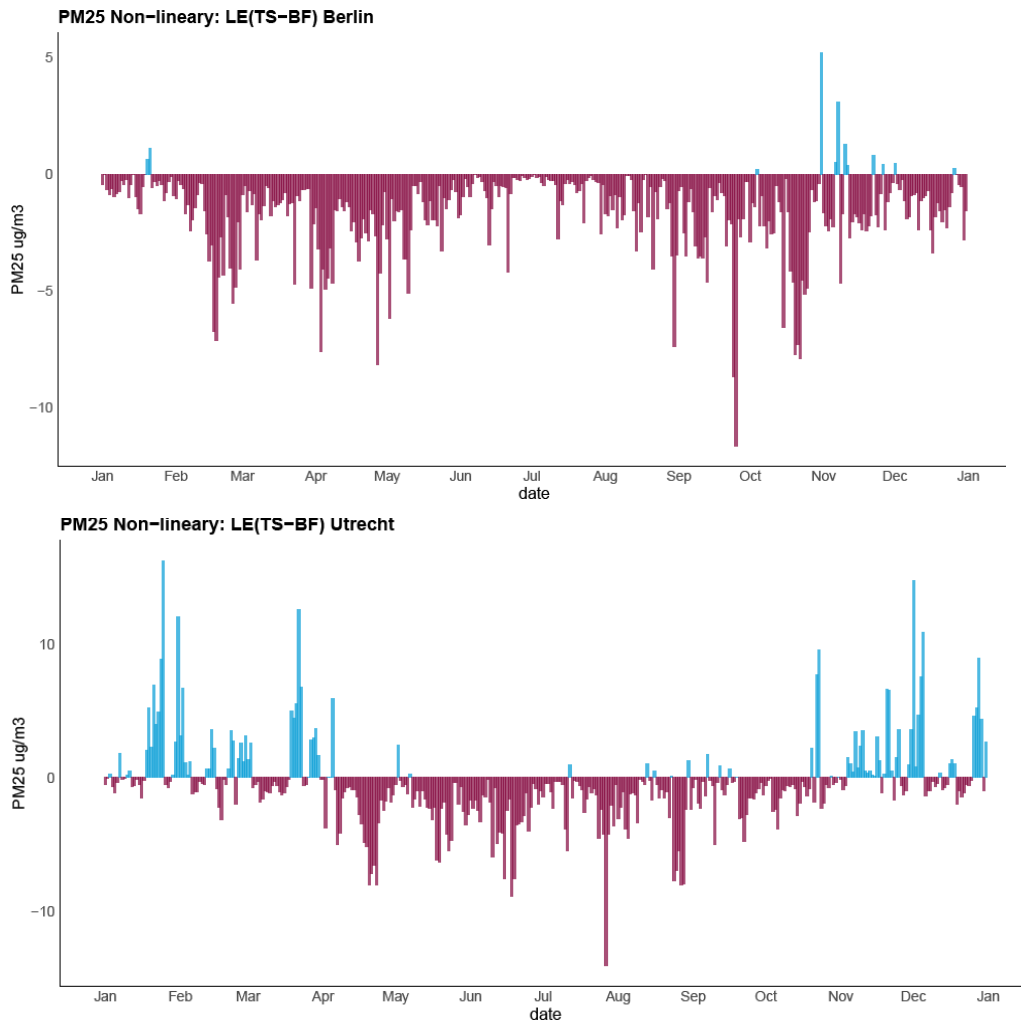


Fig. 3.5.4 Comparison of total sum of PM_{2.5} contributions ($\mu\text{g}/\text{m}^3$) from LOTOS-EUROS labelling (LE_TS) - LOTOS-EUROS BF (LE_BF) for Berlin and Utrecht.

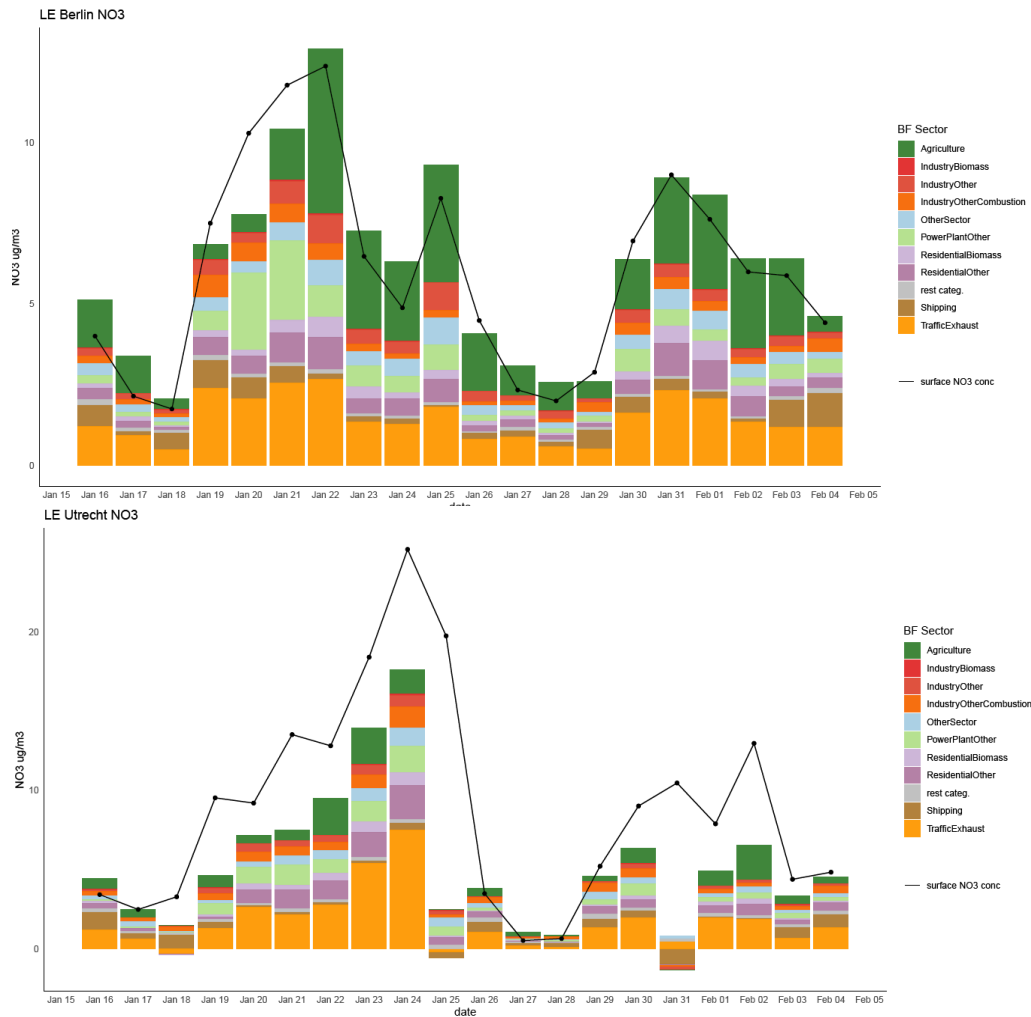


Fig. 3.5.5 The time-series from 15th of January to 5th of February of NO₃⁻ contributions (µg/m³) from LOTOS-EUROS BF and NO₃⁻ surface concentration for Berlin and Utrecht.

3.6 Visualisation of non-linearities in the city SR forecast

At present, the forecasting of city SRs presents the impact of reducing emissions in countries and the city itself, see Figure 3.6.1. The contributions are calculated by performing 15% reductions in the sources and scale up to 100%. The contributions from the different countries are stacked, and the total concentration is also shown. In order to close the gap between the contributions shown and the total concentration, a term 'Others' is defined as the difference between the total concentration and the sum of the contributions shown. This term can be divided into the remaining contribution from all anthropogenic and natural sources **plus the non-linear interactions**, which can be either positive or negative.

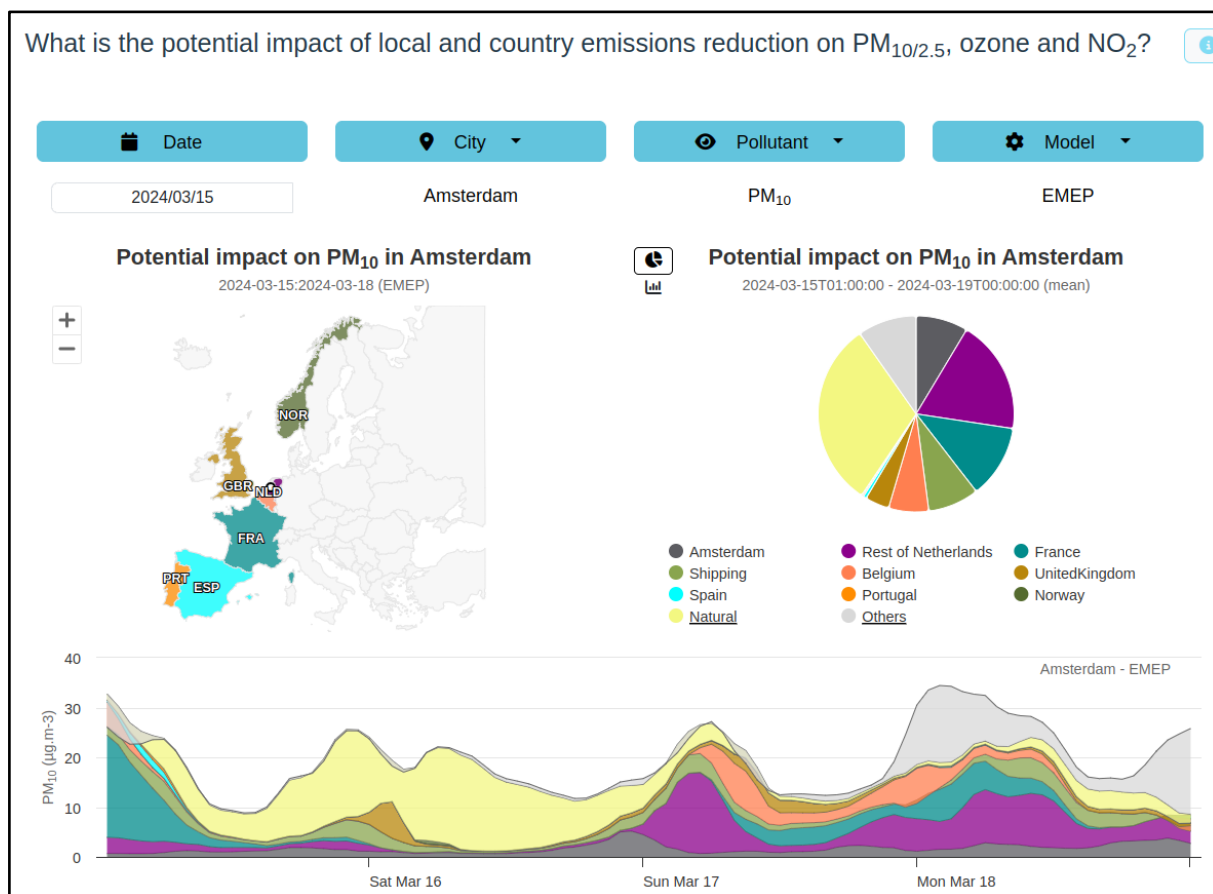


Figure 3.6.1: Example of forecast for PM10 from the CAMS Policy Support Service

For primary PM (PPM), which is inert (i.e. concentrations do not change due to chemical reactions), the model system is linear. That means that if one does a 15% reduction of PPM emissions, the changes in concentrations will be 15% of what it would be if one did a 100% reduction in emissions. Therefore, it does not matter what percentage reduction is done in the modelling; the effect is linear.

For species that undergo chemical reactions, the system is not linear. For instance, if one reduces NH₃, the concentration of PM_{2.5} (or (NH₄)₂SO₄ and NH₄NO₃) may change a lot or very little depending (among others) on the relative levels of NH₃, HNO₃ and H₂SO₄. If NH₃ is in excess (i.e., **not all NH₃ is already bound in (NH₄)₂SO₄, and NH₃ is available for participation in the equilibrium reaction with HNO₃ to form NH₄NO₃, see supplement**), the reduction of NH₃ emissions is much less efficient for reducing PM_{2.5} than if it is not in excess. The secondary inorganic aerosol (SIA) concentration decrease is not linear with the ammonia emission reduction and it may become significantly larger as the percent reduction of ammonia emission rises, especially in the cases/regions of excessive NH₃ (Pinder et al., 2007; Bessagnet et al., 2014). Thus, one cannot accurately quantify how much a 100% reduction in NH₃ will affect PM_{2.5} by doing a 15% reduction and then scaling by 100/15.

We define a measure of non-linearity of a model source allocation performed with 15% brute force calculations as:

$$\text{Non-linearity}(i,j) = \text{Concentration}(i,j) - \text{Sum_of_contributions_to}(i,j)$$

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where

Concentration(i,j) is the pollutant concentration obtained in the base run; Sum_of_contributions_to(i,j) = sum over all sources to grid cell i,j (country sources, forest fires, natural sources, and boundary conditions).

The non-linearity term might be positive or negative:

- It is positive if the sum of reductions by 15% (scaled to 100%) gives a contribution smaller than the concentration.
- It is negative if the sum of reductions by 15% (scaled to 100%) gives a contribution larger than the concentration.

In reality, the individual contributions from different components (or even from different countries) in PM_{2.5}/PM₁₀ can be positive or negative, so that the sum of contributions also might cancel out some non-linearities. In order to account for that, we considered three different possibilities:

- Approximate non-linearity to be the closure term (where closure = concentration minus all anthropogenic and natural contributions). This means accepting some cancelling out of non-linearity.
- Taking the absolute value when adding up the 'non-linearity term' from each component (e.g. NO₃⁻, NH₄⁺, SO₄²⁻):

$$\text{Non-linearity}(i,j) = |\text{contrNO3}(i,j)| + |\text{contrNH4}(i,j)| + |\text{contrSO4}(i,j)| + \dots$$

where *contrXX*(i,j) is the closure term for species XX (= NO₃⁻, NH₄⁺, SO₄²⁻) at grid cell i,j.

- Interpreting the non-linearity as uncertainty and thus derive it as a square root of the sum of the squares of the non-linearity terms:

$$\text{Non-linearity}(i,j) = \sqrt{\text{contrNO3}(i,j)^2 + \text{contrSO4}(i,j)^2 + \text{contrNH4}(i,j)^2 \dots}$$

We have chosen the first option, as the concept is easier to explain and visualize, and there is no exact way of defining uncertainty due to non-linearities.

If the **non-linearity term is large compared to the concentration** it means that that system is very non-linear, i.e. that one should be careful when interpreting how intended emission reductions would impact PM_{2.5}/PM₁₀, because **the impact will not scale with the size of the reduction**. When the non-linearities are **negative**, the intended reductions would be **more efficient than** expected from linearity assumptions, and finally, when they are **positive**, the effect of emission reductions is **less efficient** than expected.

Here, we have calculated the non-linearity for PM₁₀ as:

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$$\text{Non-linearity}(i,j) = \text{Concentration_PM10}(i,j) - \text{bSOA}(i,j) - \text{OM_bgnd} - \text{Seasalt_PM10}(i,j) - \text{Dust_PM10}(i,j) - \text{Forest Fire_PM10}(i,j) - \sum_k \text{contr}(i,j)$$

where $\text{contr}(i,j)$ is the sum of contributions in grid i,j from anthropogenic emissions in countries k . bSOA is the SOA resulting from BVOC and OM_bgnd is the background OM (organic matter) field ($0.4 \mu\text{g}/\text{m}^3$ at the surface) assumed in the EMEP model. There are some sources that are not accounted for here (e.g. soil NO_x , lightning, non-PM forest fires, DMS and volcanoes: these sources might contribute to SIA formation indirectly) that will end up in the non-linearity term, but this contribution is only minor in most cases.

For some cities, specifically those close to the borders of CAMS domain, e.g., Nicosia or Athens, affected by emission sources not reduced (e.g. from Africa or Russia), some anthropogenic contributions end up in the closure term. This can be solved by adding an extra run reducing all anthropogenic contributions and subtracting this term from closure. In an operational setting, also some of the sources mentioned above, in particular volcanoes and DMS, need to be included.

Figure 3.6.2 shows an example for PM_{10} in Amsterdam in February 2019. The non-linearity term (= the closure term) is mostly positive (the bottom time series), meaning that 15% reductions of emissions would result in less than 15% reduction of the anthropogenic part of PM_{10} . In the bottom plot of Figure 3.6.2, the total non-linearity term for PM_{10} is split into non-linearities due to different chemical components, in particular discriminating those for NO_3^- , SO_4^{2-} and NH_4^+ . Clearly, NH_4NO_3 is dominating the non-linearity term.

In Figure 3.6.3, we show results for Bratislava in July 2019. Here, non-linearities are mostly negative, meaning that a 15% reduction in emissions would give more than a 15% reduction in the anthropogenic part of PM_{10} (see Annex A for explanation of the non-linearities in different situations).

In Figure 3.6.4, we show an example from Berlin where we have compared the non-linearity/closure term using the EMEP BF and LF calculations. Because BF calculations in general are hampered by the numerical differences introduced by the Bott scheme when doing emission reductions for countries, total contributions are overall a little larger in BF than LF. Therefore, the closure terms for the LF calculations are overall somewhat smaller negative, or larger positive (and more correct). The similarity between the closure terms in those two approaches, which to a large extent represent calculation of non-linearities when i) starting from a very small reduction and ii) when starting from a 15% reduction, are encouraging and suggest that the calculation of the non-linearity term is rather robust in those first 15% reductions.

For the CAMS Policy Support Service for country-to-city and/or city-to- itself (impacts of emission reduction), we recommend that the 'Other' term is replaced by 'Other sources' (meaning all other sources that are quantified but not displayed) and a 'non-linearity' term which is quantified as the difference between the total concentration and the contributions of all the different terms. If the non-linearity term is large, this means that one should be careful when interpreting how any emission reductions would impact $\text{PM}_{2.5}/\text{PM}_{10}$, because the impact will not scale with the size of the reduction.

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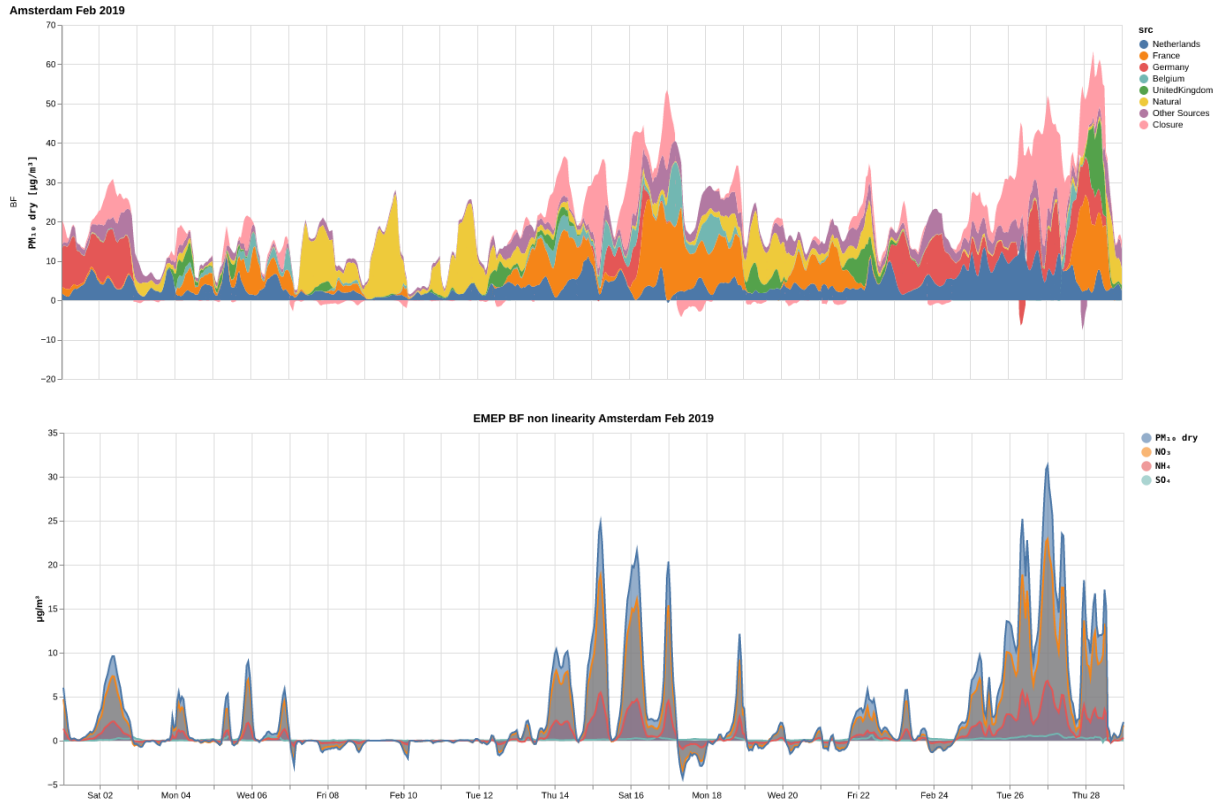
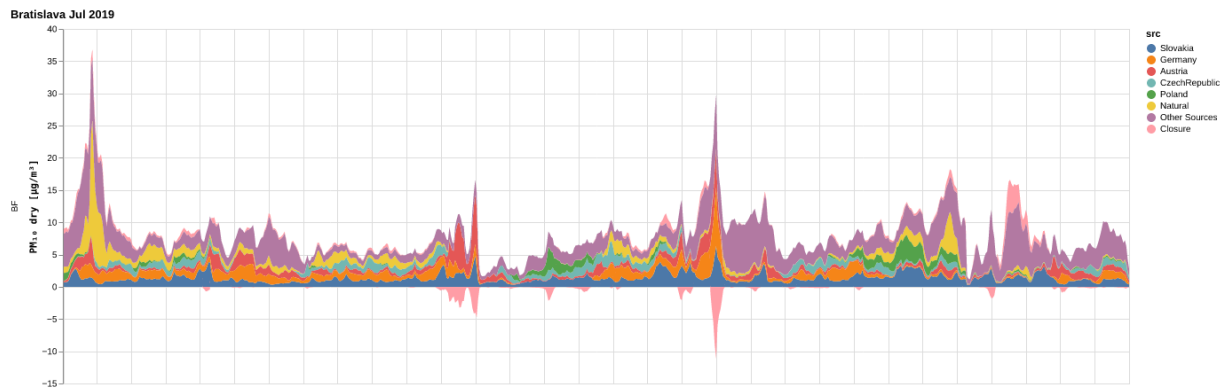


Figure 3.6.2 Top: Contributions to PM₁₀ pollution in Amsterdam in February 2019. Bottom: The non-linearity terms for NO₃⁻, SO₄²⁻, NH₄⁺ and PM₁₀ (overlaid, not stacked).



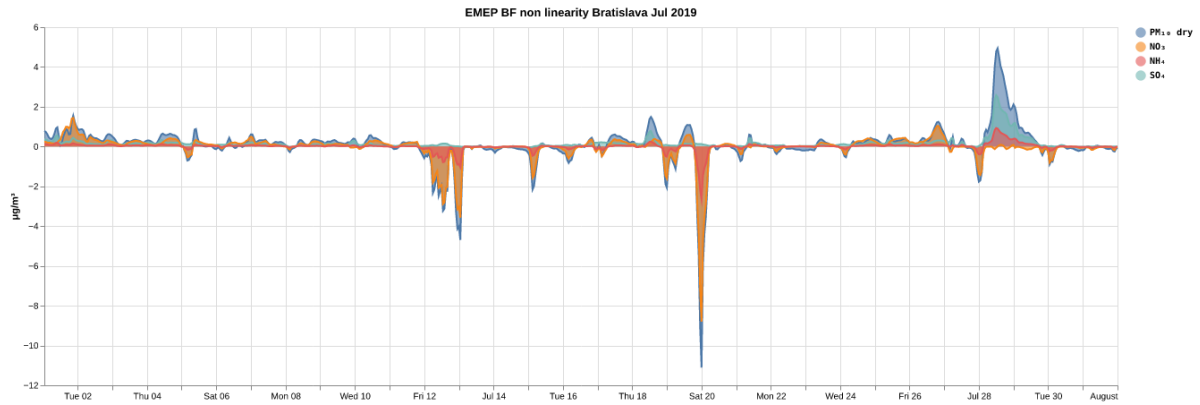


Figure 3.6.3 Top: Contributions to PM₁₀ pollution in Bratislava in July 2019. Bottom: The non-linearity terms for NO₃⁻, SO₄²⁻, NH₄⁺ and PM₁₀ (overlaid, not stacked).

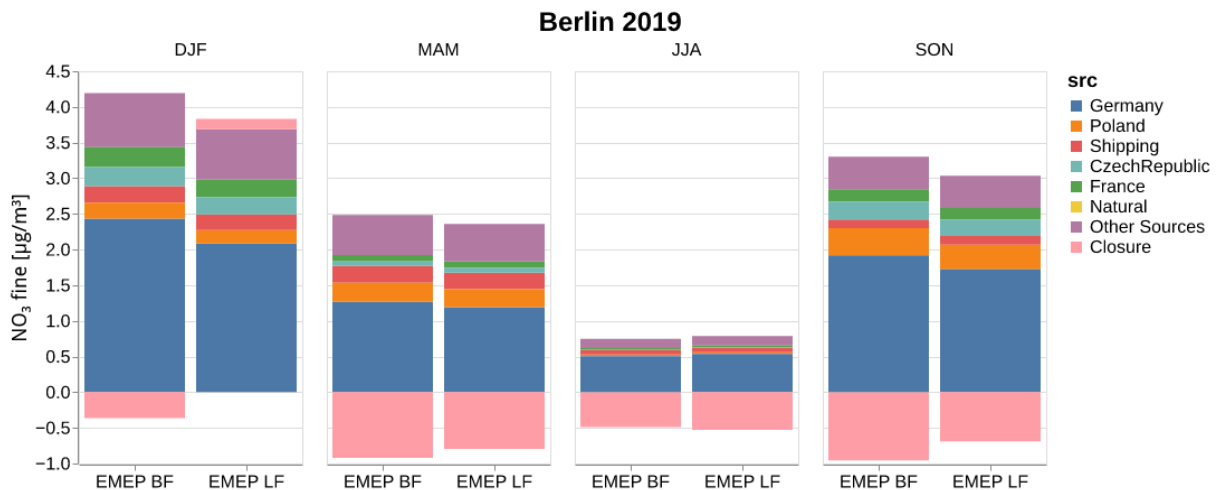


Figure 3.6.4: Contributions to NH₄NO₃ concentrations in Berlin in different seasons (from left to right: DJF, MAM, JJA, SON) using EMEP BF (left) and EMEP LF (right).

3.7 Local contributions in cities from non-linear species

In the CAMS policy support service that forecasts the potential impact of local and country emissions reduction on PM₁₀/PM_{2.5}, two different types of model runs are performed at present; 1) EMEP model runs where emissions are reduced in country by country and 2) Model runs where emissions are reduced in city by city. Due to the large amounts of CPU time that is required to run all of these model runs every day, some simplifications are done. For instance, emissions in cities that are far from each other (e.g. Vienna and Brussels, Oslo and Warsaw, etc.) are reduced simultaneously, and in this way we are reducing the number of runs.

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It has been proposed to use the local fraction methodology for calculation of country and city to city SR's, because this methodology is much more computationally efficient. In Section 3.4 of this report, as well as in EMEP Status Report 1/2024, it has been shown that the EMEP LF method can be used as a replacement of the BF methodology.

It has been proposed that the local contribution (from the city to the city itself) can be approximated by primary PM, and that the formation of SIA within the city is limited. This approximation is applied, e.g., in the integrated assessment model GAINS.

As part of the investigations of the LF method, we also analyzed whether it is reasonable to approximate the local contribution (i.e., the city to itself) by the PPM contribution. Figure 3.7.1 shows the contribution from the city to itself (and the non-local contribution) for 19 cities in Europe, where the total local contribution has been split into contributions from PPM, SIA and anthropogenic and biogenic SOA. From the Figure it can be seen that PPM is indeed the largest local contribution, and that in most cases it would be a good approximation to assume that the local contribution mostly consists of PPM. Furthermore, secondary organic aerosols result only very little from local production, as expected. However, local production of SIA is important in cities such as Athens, Madrid, Sofia and Milan, where the local SIA production can be 20-30% of the local contribution.

From this analysis, we conclude that local production of SIA can be important and should be included when analysing air pollution in cities. Local production of SOA can be neglected.

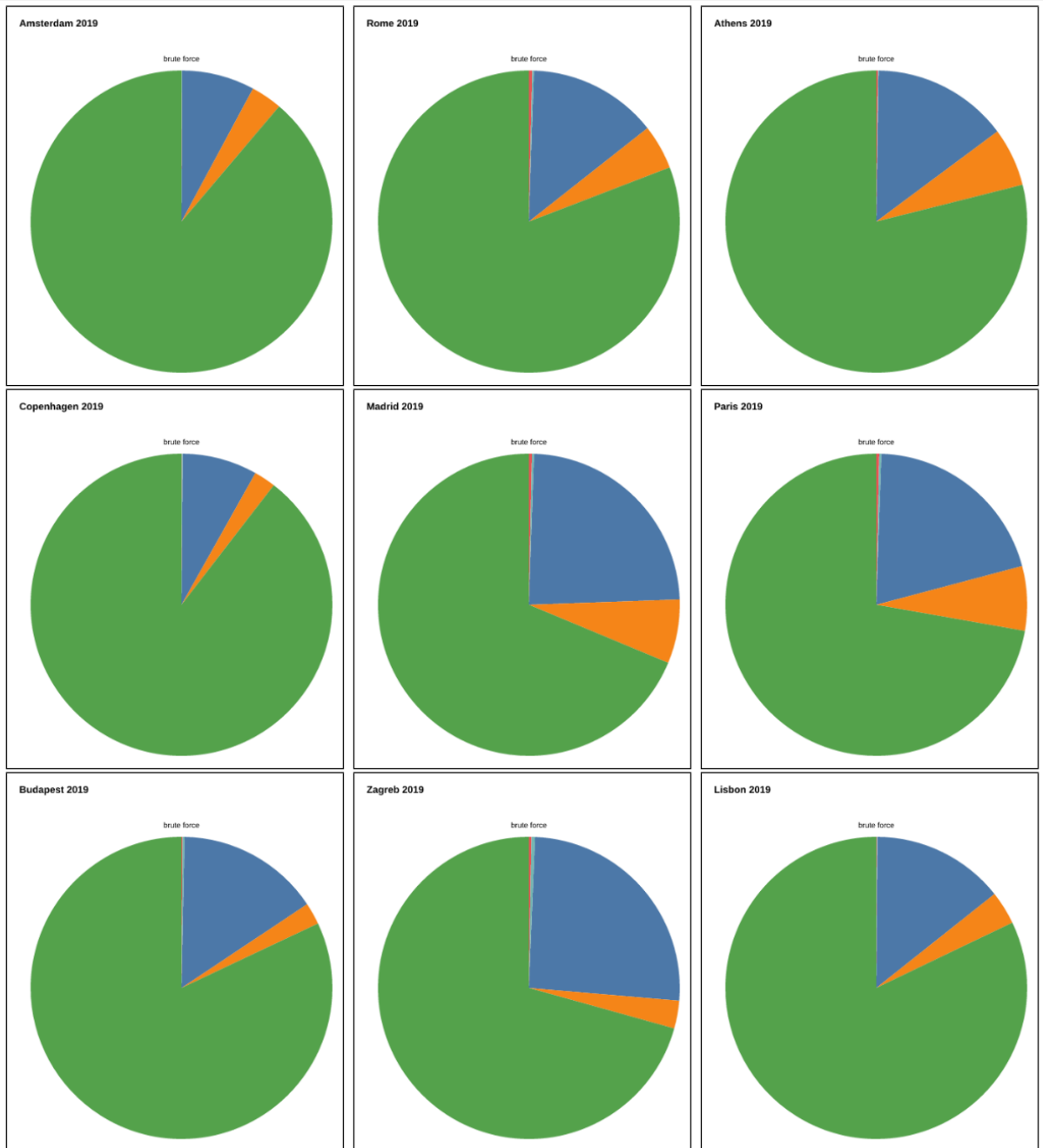


Figure 3.7.1: The local (city to itself) contribution from primary PM, secondary inorganic (SIA), anthropogenic secondary organic aerosol (aSOA) and biogenic secondary organic aerosol (bSOA) and the non-local contribution (rest PM25).

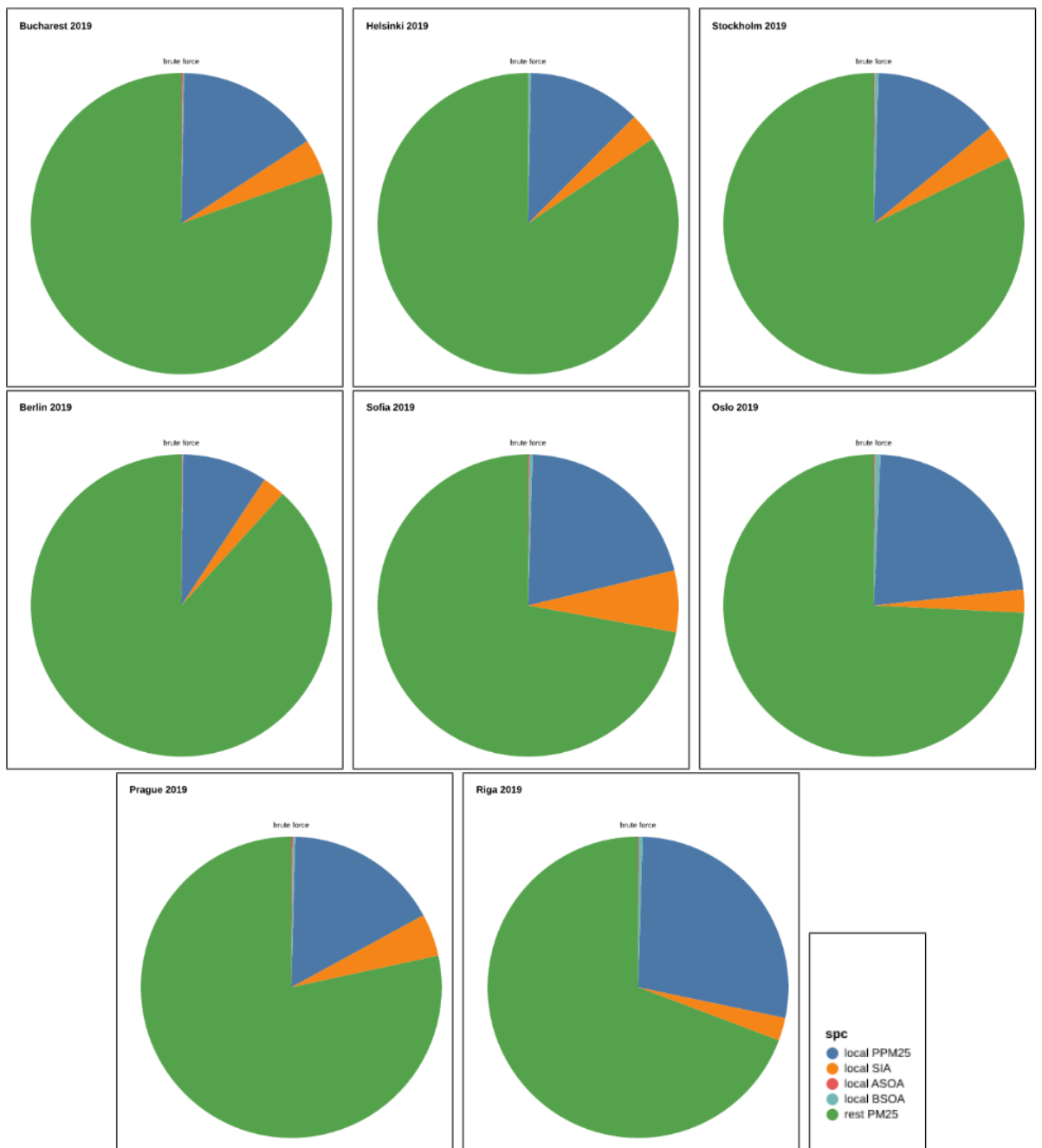


Figure 3.7.1 continued: The local (city to itself) contribution from primary PM, secondary inorganic (SIA), anthropogenic secondary organic aerosol (aSOA) and biogenic secondary organic aerosol (bSOA) and the non-local contribution (rest PM2.5).

3.8 Conclusions and recommendations

In this task, we have quantified the relative importance of the non-linearities for different chemical species and policy products. We have focused on source receptor products for emission contributions from different **countries** and **sectors** to **cities**, as those are the most prominent products on the CAMS Policy service today. Furthermore, we have suggested how non-linearities in source allocations can be calculated and presented for PM on the CAMS Policy Service.

Differences due to model formulations (same source apportionment methodology):

- Contrary to what we expected, we found the largest differences in the city SRs from EMEP and LOTOS-EUROS BF to be in primary PM, while the SRs for the secondary inorganics were more similar. A similar issue was found for the sector SRs, where large differences in the residential biomass contributions between LOTOS-EUROS and EMEP was found for cities which have large residential heating emissions (which are treated as primary emissions here). Residential emissions are inserted into the surface layer in both models. Since these emissions usually occur during cold conditions with low mixing heights and inefficient vertical mixing, the resulting concentrations are very sensitive to the depth of the models' surface layer, which is 20m in LOTOS-EUROS and 50m in EMEP. This could at least partly explain the differences in this modelled contribution. Also differences in vertical mixing parametrisations in the models influence the results. The results for the CHIMERE model are more in line with the EMEP model.
- We found the differences in SIA to be less significant. SIA is less sensitive to the formulation of the lowest layer, both because it is secondary (and needs some time to form) and also because it is formed in reaction between species from a mix of high and low level sources (e.g. industry, public power, agriculture, traffic etc.). Although the chemical schemes for SIA formation are not the same in the two models, the results for SIA are rather similar (also seen in the comparison to observations). From previous model intercomparison (e.g. a recent model (2023) intercomparison of the regional models in CAMS, see e.g. https://aeroyal.met.no/pages/intercomp/?project=cams2_40_p2&exp_name=wetdep&experiment=aq¶meter=concNno3pm25&station=ALL#) it is well known that this is not always the case, and it is likely that differences in SIA between other models would be larger.

Differences due to source apportionment methodologies (same model):

- In theory, the choice of source attribution methodology should not matter for primary species. In practice, we see some small differences due to numerical implementation of methods, but the magnitude of differences are so small that they can be ignored.
- BF and LF methodologies are the methods that give potential impact of emission reductions and thus they give very similar results, even for source apportionment of secondary inorganic aerosols
- The labelling method traces where the pollution comes from (not what the impacts of emission reductions are) and is a fundamentally different method. The differences between BF and labelling results for source apportionment of secondary inorganic aerosols are significant, especially on a shorter time scale (e.g. daily). For emission sector contributions, this is mostly connected to the agricultural and traffic exhaust sectors (and for some cities other large NO_x sources as well).

Comparing differences due to model formulations with those due to source apportionment methodology:

- We quantified the differences in country contributions to cities due to different methodologies as the root mean square error (RMSE) of (daily mean) EMEP LF versus EMEP BF, the RMSE of (daily mean) LOTOS-EUROS BF versus labelling. Similarly, we have quantified the difference due to model formulations by calculating the RMSE of (daily mean) EMEP BF versus LOTOS-EUROS BF.
- RMSE for the secondary inorganic aerosols (SIAs) are rather large due to SA methodologies and of the same magnitude as the differences due to model formulations for nitrate and sulfate.
- It is clear that particularly for the largest contributor, RMSE for PM_{2.5} is much larger for models' differences (EMEP versus LOTOS-EUROS) than differences in SA methodology. Here, the RMSE for PM_{2.5} is a combination of RMSE for primary PM (large differences due to model formulation, no differences due to SA methodology) and SIA (comparable differences due to model formulation and SA methodology).

For LF versus BF SA methodology, the differences are even smaller for SA methodology, and model differences are totally dominant.

At present, the CAMS Policy Support Service separates the source attribution methods for PM into two different products: Potential Impacts of emission reductions (BF, done with the EMEP model for the spatial allocation or with ACT/CHIMERE for the sectoral allocation) and Country Contributions (labelling, done with LOTOS-EUROS for the spatial allocation). For users, this is confusing, as many users do not understand why source allocations calculated with these two methods (contribution and potential impact) are different and which of them should be used. One of the reasons for the differences in SA produced by BF and labelling is different treatment of chemical non-linearities, but in this task we have shown that the differences between the models (i.e. between EMEP and LOTOS-EUROS) are larger than the differences in source attributions done with different methodologies.

It follows from the conclusions above, that in principle we could combine source attribution of primary PM from the different models (EMEP, LOTOS-EUROS and also CHIMERE) and methodologies into a mini-ensemble. For SIA (and to a lesser extent SOA) it is less clear how this can be done from a principal perspective, although in practice the differences due to model formulations are as large as differences in SA methodology.

Presentation of non-linearities in the CAMS Policy Service:

- We recommend that in CAMS2_71 source allocation products for country-to-city and/or city-to-itself (impacts of emission reduction), the term 'Others' should be split to 'Other sources' (meaning all other sources that are quantified but not displayed) and a 'Non-linearity' term which is quantified as the difference between the total concentration and the sum of all allocated contributions, which in theory would make up the concentrations if the model system was linear. A large non-linearity term means that you should be careful when interpreting how your emission reductions would impact PM_{2.5}/PM₁₀, because the impact will not scale with the size of your reduction. In such cases, the contribution of a country is going to be overestimated or underestimated by the source allocation.
- The closure terms (the non-linearity term) from the LF calculations are overall somewhat less negative, or larger positive (and more correct) than those from the BF

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calculations. The similarity between the closure terms in those two approaches, which are based on calculation of non-linearities i) starting from a very small reduction and ii) starting from a 15% reduction, is encouraging and suggests that the calculation of the non-linearity term using 15% emission reductions is quite robust. This also means that this method for calculating the non-linearity term can be used even if the BF calculations are replaced with LF in the CAMS Policy service.

Local contribution to PM in cities:

- The local contribution to PM in cities is often approximated to primary PM (e.g. in the integrated assessment model GAINS), assuming that local production of SIA and SOA takes time and matters more at regional scale. We show that although primary PM is indeed the largest local contribution for all cities investigated, local production of SIA is important in cities such as Athens, Madrid, Sofia and Milan, where it can comprise as much as 20-30% of the local PM contribution.

4 Consistency and comparability between EMEP source-receptor calculations for CLRTAP and CAMS Policy Service

4.1 Introduction

The EMEP model has been used for calculating source-receptor (SR) relationships for UNECE countries in the framework of the Convention for Long Range Transboundary Air Pollution (henceforth CLRTAP_SR) and also for allocation of pollution sources in major EU cities within the CAMS Policy service (CAMS_SR). In the both projects, the SR calculations are based on an emission perturbation (or brute force) method and performed by reducing emissions by 15%. The main difference between the SR setups in the two projects is that in CLTRAP_SR calculations, the emissions of different pollutants (SO_x, NO_x, NH₃, NMVOC, PPM) are reduced in separate model runs, while for CAMS_SR forecasts all emissions are reduced in a single run.

The first part of Task 6.1.2 consists in the analysis of “the consistency and comparability between the annual SR information for European countries (and cities) from CAMS and the SR matrices that are produced within EMEP and used for policy development within the LRTAP Convention”. Country-to-country and country-to-city SR calculations have been compared for 31 countries and 80 cities (with a population of 500 000 people and above). The second part of the Task is to investigate the “potential for exploitation of EMEP CLRTAP_SR matrices in future operational CAMS products and vice versa”. In the following sections, we explain the methodology for comparison of CLRTAP_SR and CAMS_SR, discuss the obtained results in terms of consistency and comparability, and finally outline main points with respect to possible complementarity of CLRTAP_SR and CAMS_SR information for both CLRTAP and CAMS Policy Service.

Originally, one of the intentions in the framework of Task 6.1.2 was to investigate the non-linearities in SR calculations with respect to reducing emissions of SO_x, NO_x, NH₃, NMVOC and PPM separately or together in one run (which is is one of the major differences between CLRTAP_SR and CAMS_SR). However, there turned out to be too many inconsistencies in the setup of these SR calculations, making the discrimination of this effect too difficult.

4.2 Description of SR in CAMS and CLRTAP

For comparison of CLRTAP_SR and CAMS_SR, we have used existing results from earlier SR calculations for the year 2022: the CLRTAP_SR were performed in 2024 and included in EMEP status Report 1/2024 and the CAMS_SR were obtained from CAMS2_71 source allocation forecasts back in 2022. It should be noted that in addition to the manner of SR calculations in terms of simultaneous or individual emission reductions (mentioned in Section 4.1), CAMS_SR and CLRTAP_SR differ in terms of model version and setup and input data (see Table 4.1 for technical details).

Here, we summarise the most relevant differences between the runs' setup, which are expected to make appreciable effects on model simulated pollutant fields and thus on SR results (those having the most significant effects are **highlighted in bold**).

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Emission perturbations:

- For CLRTAP_SR, five separate runs are made for 15% reduction of SO_x, NO_x, NH₃, NMVOCs, and PPM individually; in CAMS_SR runs 15% reductions are applied to all emissions (see documentation on https://policy.atmosphere.copernicus.eu/documentation/source_allocation.php).

Model versions: CLRTAP_SR used a much more recent version than CAMS_SR.

- **CLRTAP_SR uses ISORROPIA-Lite for gas/aerosol partitioning, which simulates more NO₃⁻ (including coarse NO₃⁻ on sea salt and mineral dust) and thus PM with respect to MARS in CAMS_SR.**
- CLRTAP_SR has updated photolysis rates calculated with Cloud-J v7.3e scheme, which produces somewhat more surface ozone
- Updates in sea salt parameterisation in CLRTAP_SR have some effects on NO₃⁻ and PM

Setup:

- Horizontal resolution - CLRTAP_SR are run on a coarser grid (0.3x0.2°) than CAMS_SR (0.2x0.1°)
- **Vertical resolution - CLRTAP_SR uses a thinner lowest layer (50 m) than CAMS_SR (92 m)**

Input data:

- **Emissions - CLRTAP_SR used the most recent official EMEP emission for 2022, whereas CAMS_SR used CAMS emissions for 2015**, which are to a large extent based on EMEP country/sector totals. Since EMEP emissions went down from 2015 to 2022 in EU countries, CAMS_SR are based on higher emissions. There are also differences in the spatial distribution of emissions, in particular for the residential heating sector.
- Meteorological driver - IFS reanalysis in CLRTAP_SR vs forecast in CAMS_SR. Note that **three-dimensional precipitation is not available in IFS forecast output, so that it needs to be recreated based on two-dimensional precipitation and three-dimensional liquid cloud water**
- Boundary conditions - pollutant concentration fields for actual dates obtained from CAMS IFS are used in CAMS_SR, whilst CLRTAP_SR runs are mostly based on tabulated “climatological” concentrations
- Temporal distribution of Residential emissions - CLRTAP_SR uses Heating Degree Days to adjust the temporal variation of Residential heating emissions, while CAMS_SR does not allow for their temperature dependence

Table 4.1 Technical details for EMEP SR runs for CLRTAP and CAMS

Technical details	CAMS_SR	CLRTAP_SR
Model version ¹	rv4.34 (OS 1.01.2020)	rv5.3
Horizontal resolution	0.25 x 0.125°	0.3 x 0.2°
Lowest layer	92 m	50 m
Emissions	CAMS REF2 v1 for 2015	EMEP for 2022
Forest Fires	GFAS v1.4 (hourly)	FINN modvrs_v25
Meteorology	12:00 UTC operational IFS forecast	ECMWF-IFS (cycle 48r1) reanalysis
Precipitation	3D recreated from 2D field	3D
Heating Degree Day	No	Yes
BIC	CAMS IFS	EMEP (climatology based, tabulated)
Results availability: temporal resolution	For current year (starting from 2019) Hourly/daily/seasonal/yearly	For 2 years back Yearly (monthly?)
Emission perturbation for SO _x , NO _x , NH ₃ , NMVOC, PPM	All emissions are reduced by 15% simultaneously in the same run	Emissions are reduced by 15% individually in separate runs
Components	PM _{2.5} , PM ₁₀ , O ₃ , O ₃ MDA8	PM _{2.5} , elemental carbon, primary PM _{2.5} , ozone indicators (MM- AOT40f, SOMO35, MDA8), deposition of oxidised sulphur, oxidised and reduced nitrogen - as reported in 2024

4.3 Methodology for comparison

CAMS_SR are focused on PM_{2.5}, PM₁₀ and O₃ concentrations, while CLRTAP_SR also calculates other pollutants, as well as depositions of oxidized and reduced nitrogen and oxidized sulphur to different ecosystems. For this comparison, we will focus only on concentrations of PM_{2.5}, PM₁₀ and O₃, which are calculated by both.

Before we can compare them, we must also consider the different temporal and spatial scales of the SR output. In CAMS_SR the receptors are European cities (since 2023, 80 cities with a population above 500 000 people). The SR calculations are used to quantify how much of the pollution concentration in the city can be attributed to emissions in the city itself and to emissions from each country in the European Economic Area and Switzerland (31 different

¹ See the text for more specifics

countries), from international shipping, and due to hemispheric transport for ozone. 4-day forecasts are run operationally every day, producing hourly SR output (https://policy.atmosphere.copernicus.eu/daily_source_attribution/country_impact.php?), and the first 24 hours of each forecast are used to compile seasonal/yearly average SR².

In contrast, in CLRTAP_SR the receptors are countries and sea areas. SR calculations are used to quantify how much of the pollution in 59 countries³ and 5 sea areas can be attributed to emissions from each of these same areas (country-to-country blame matrices). This product is calculated only once a year, and results are only for annual mean (EMEP Status Report 1/2024).

Since we only have annual mean data from CLRTAP_SR, the comparison is done for annual mean concentrations⁴. For a comparison, we must use the same receptors for both, i.e. calculate contributions to cities from CLRTAP output as in CAMS_SR, or calculate contributions to countries from CAMS output as in CLRTAP_SR. To do this, we need the gridded model output from each reduction run.

Concentrations in countries based on CAMS_SR model output were obtained by first calculating annual mean from the hourly SR output, then averaging each model run (i.e. the baseline and each emission reduction simulation) over each receptor country (simple average using area-weighted country mask, as is done in CLRTAP_SR).

Concentrations in cities based on CLRTAP_SR model output were obtained by averaging each model run over each receptor city. The city definition used in the CAMS Policy product is a 3x3 grid cell in the 0.25x0.125° grid used at the time. Since the CLRTAP runs used a different grid (0.3x0.2°), area-weights were applied for grid cells partly within the city. This method was also reapplied to CAMS_SR runs in order to include all 80 cities in the comparison, since some of the cities are not shown in the CAMS policy product for years prior to 2023 (for 2022, only 38 cities are shown).

In both cases, the contribution of a source (country or shipping) to a city/country is calculated as the difference in concentration between the baseline and the run where that source was reduced by 15 %. To represent source allocation, this difference is then scaled up to 100 % by multiplying by 100/15. In CLRTAP_SR, the calculated contributions from the runs reducing SO₂, NO_x, NH₃, NMVOCs, and PPM are added. The scaling up to 100 % is done in all cases for our comparison here in order to be consistent with how the contributions are shown in the CAMS Policy product. However, in the EMEP status report the blame matrices are mostly presented as the effect of a 15 % emission reduction (t.e. not scaled to 100 %).

In our comparison, we only include sources that are calculated in both CAMS_SR and CLRTAP_SR, i.e. 31 countries and international shipping. In CLRTAP_SR, the contribution from international shipping is calculated as the sum of the contributions from the 5 sea areas that are reduced individually. Since CLRTAP_SR does not have a reduction run for emissions in the city itself, this part of the CAMS_SR is included in the home country contribution when comparing.

Both SR products quantify natural sources. However, these are not included in the comparison because they are not the same sources: in CAMS_SR it is mineral dust, sea salt and forest fires, while in CLRTAP_SR it is Volcanoes and DMS.

² In fact, the CAMS Policy Service interface provides daily time series of country allocations, zoomable on any time period (https://policy.atmosphere.copernicus.eu/yearly_air_pollution_analysis/country_impact.php)

³ These are the parties to the CLRTAP convention.

⁴ The EMEP status report does not actually show blame matrices for annual mean PM₁₀ and ozone, but these concentrations were outputted by the model runs and can be used for this comparison.

4.4 Results

In this section we present and discuss CLRTAP_SR and CAMS_SR results in terms of top-three contributions to annual mean concentrations of PM₁₀, PM_{2.5} and ozone in 80 cities and 31 countries, based on the simulations for 2022, described above.

Figure 4.1 compares, for each city (a-c) and each country (d-f), the three largest contributions to PM_{2.5} calculated with CAMS_SR to the same contributors calculated with CLRTAP_SR. Note that in all cases, the top-three contributors are identified from CAMS_SR, so that the same contributor is compared between CAMS_SR and CLRTAP_SR. Figures 4.2-4.3 show the same comparison for PM₁₀ and ozone.

Here, we choose to show the contributions in $\mu\text{g}/\text{m}^3$ because the relative contributions would be biased due to differences in the PM₁₀, PM_{2.5} and ozone concentrations in the base runs: PM concentrations are higher in CLTRAP_SR than CAMS_SR run (mostly because of higher NO₃ and sea salt concentrations), and ozone concentrations are in general slightly higher partly due to the Cloud-J scheme, but also boundary conditions and emissions used plays a role (see Section 4.2).

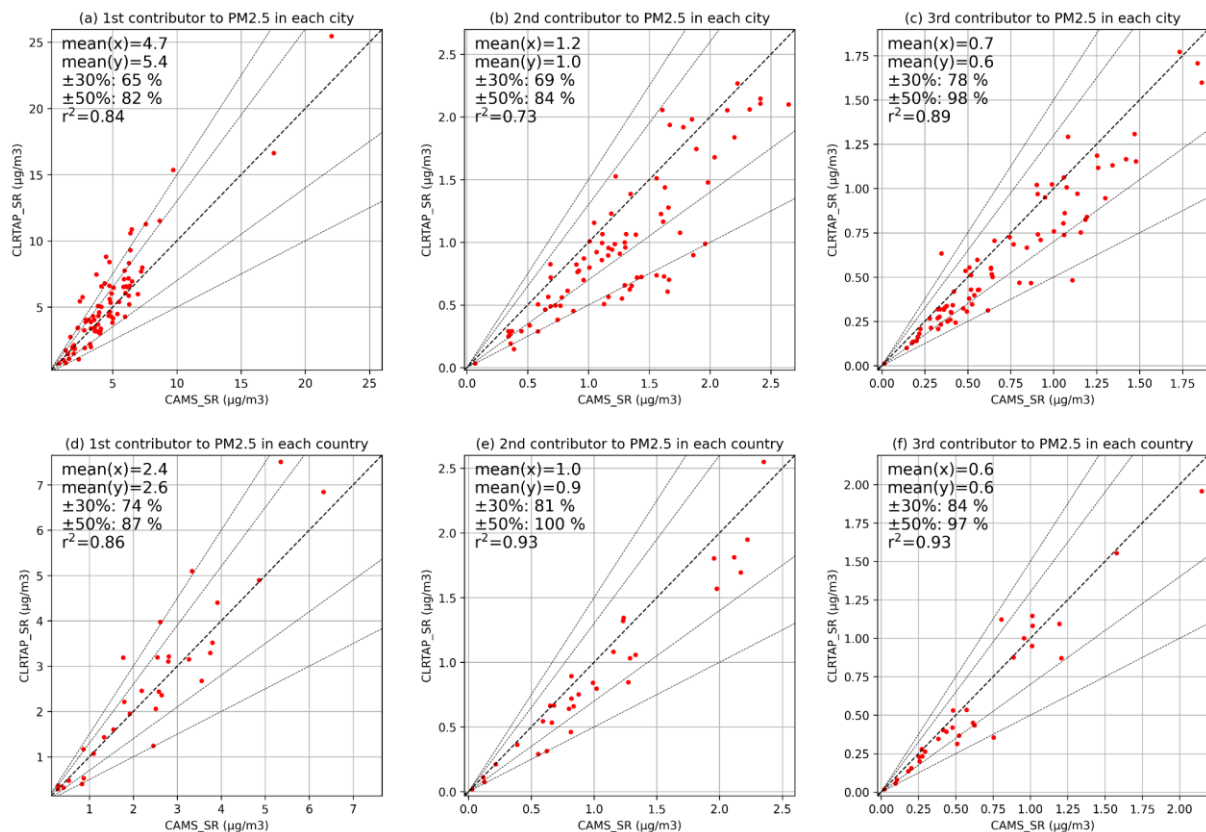


Figure 4.1: Scatterplots of annual mean PM_{2.5} concentrations due to emissions from top-three contributors calculated by CAMS_SR (x-axis) vs CLRTAP_SR (y-axis) for cities (upper panels) and countries (lower panels): the 1st, 2nd and 3rd contributors are shown on plots (a) and (d), (b) and (e), and (c) and (f) respectively. The thick dashed lines indicate perfect agreement between CAMS_SR and CLRTAP_SR, while the thin dashed lines mark deviations of 30% and 50% between CLTRAP_SR and CAMS_SR. N.B.: the top-three contributors are as identified by CAMS_SR. In the upper-right corner of each plot, the mean over all cities/countries are indicated, as well as the percentage of cities/countries where relative deviation is less than $\pm 30\%$ and $\pm 50\%$, and the squared correlation coefficient (r^2).

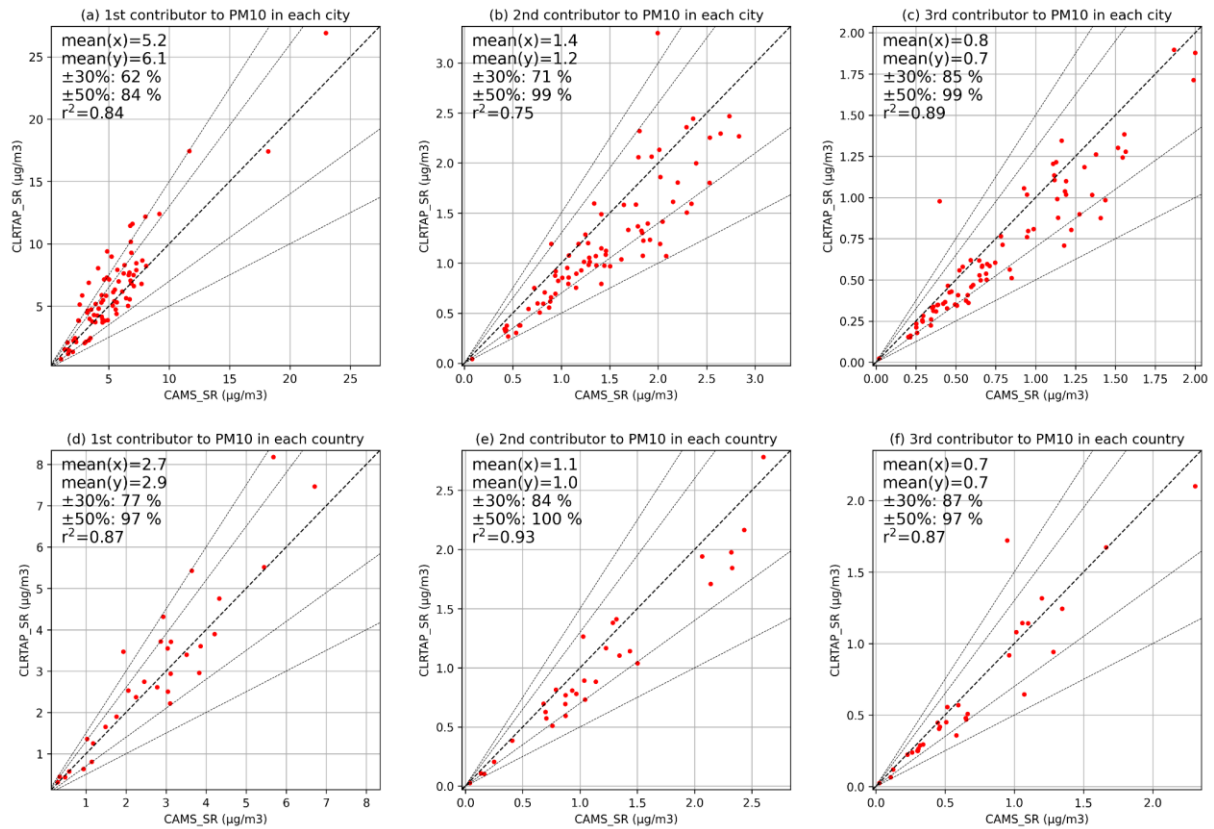


Figure 4.2: Same as Figure 4.1, but for the annual mean PM₁₀ concentration.

On the scatterplots, each red dot represents either one of the 80 cities, or one of the 31 countries, included in the SR calculations. In addition to the 1:1 line, representing equal concentrations from the same contributor obtained from CLTRAP_SR than CAMS_SR, the two other pairs of lines delineate the receptors (cities or countries) where contributions from CLTRAP_SR and CAMS_SR agree within 30 and 50%, respectively. In the upper right corner, several statistics are shown: mean concentrations from the respective contributors in CLTRAP_SR and CAMS_SR, the squared correlation coefficient (r^2) between CAMS_SR and CLTRAP_SR in the variability in top-three contributor magnitude between cities/countries, and the fractions of points (cities/countries) for which CLTRAP_SR and CAMS_SR results agree within 30 and 50%.

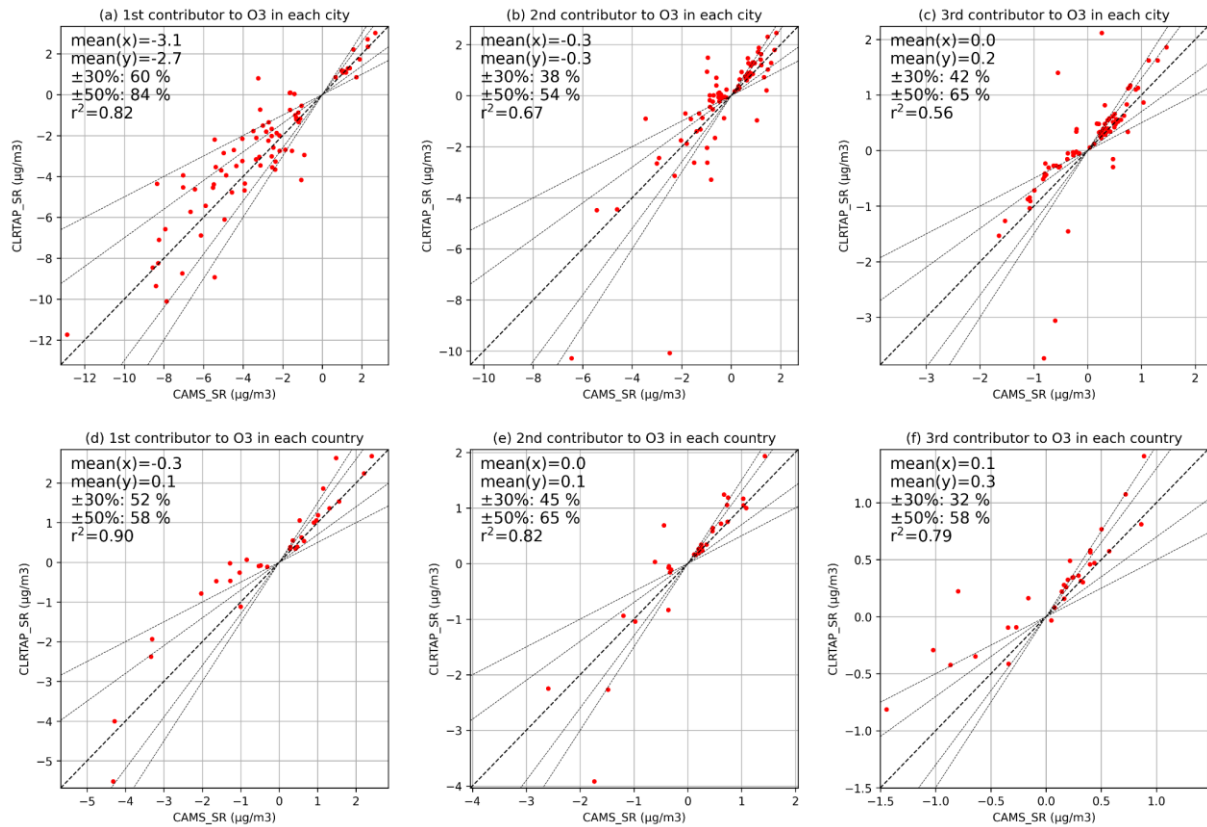


Figure 4.3: Same as Figure 4.1, but for the annual mean ozone concentration. N.B.: When identifying the largest contributors, absolute values are used, so negative contributions can also be selected.

Table 4.2 summarises the statistics of comparison between CLRTAP_SR and CAMS_SR results for the top-three contributors to PM₁₀, PM_{2.5} and ozone concentrations in the considered cities and countries. In addition to the correlation coefficients already shown on the scatterplots (Figures 4.1-4.3), Table 4.2 shows the percentage of cities and countries receptors, for which the top-three contributors in CLRTAP_SR match those indicated by CAMS_SR calculations (i.e. that they both point to the same source-country). In addition, shown is the percentage of cities and countries receptors, for which contributions from CLTRAP_SR and CAMS_SR agree within 30% (named “Agreement within 30%”).

Overall, the correlation is quite high, especially for country-to-country SRs. Somewhat lower correlations for country-to-city SR can partly be due to differences in the spatial distribution of EMEP and CAMS emissions, but also due to interpolations involved in the calculations of city average concentrations for CLRTAP_SR (see Section 4.1.3). Below, we discuss the comparison results individually for PM and ozone.

Table 4.2: Statistical comparison between CLRTAP_SR and CAMS_SR for top-three contributors (ranked as in CAMS_SR) to annual mean concentrations in countries and cities. Here: R^2 is the squared correlation coefficient; “Agreement within 30%” is the percentage of cities/countries for which contributions from CLTRAP_SR and CAMS_SR agree within 30%; “Match” gives the percentage of cities/countries for which CLTRAP_SR and CAMS_SR indicate the same country-source as the 1st/2nd/3rd largest contributor.

		Country to country SR			Country to city SR		
		1st	2nd	3rd	1st	2nd	3rd
	Contributors						
PM_{2.5}	R^2	0.86	0.93	0.93	0.84	0.73	0.89
	Agreement within 30% (%)	74	81	84	65	69	78
	Match (%)	97	81	68	100	85	71
PM₁₀	R^2	0.87	0.93	0.87	0.84	0.75	0.89
	Agreement within 30% (%)	77	84	87	62	71	85
	Match (%)	97	84	68	96	83	73
Ozone	R^2	0.90	0.82	0.79	0.82	0.67	0.56
	Agreement within 30% (%)	52	45	32	60	38	42
	Match (%)	52	32	29	76	43	38

4.4.1 PM_{2.5} and PM₁₀

The correlation coefficients, agreement in contribution values, and source match between CLRTAP_SR and CAMS_SR are fairly similar for PM_{2.5} and PM₁₀.

The correlation remains high for all top-three contributors, while we see some degradation of the correspondence regarding source-countries, i.e. decreasing Match from the 1st to 2nd, to 3rd contributor. For PM_{2.5} and PM₁₀, both CLRTAP_SR and CAMS_SR point to the same main (1st) contributor for almost all cities and countries (with the exception of Cyprus, and Nice and Palermo for PM₁₀). The main contributor is practically always (90-95% of cases) domestic (national) emissions. The correspondence is also good for the 2nd contributor (80-85 % matches) and somewhat lower for the 3rd contributor (ca. 70 % matches), for both country and city receptors (Table 4.2). Typically, the mis-matches are found for either smaller country-receptors surrounded by larger country-sources (e.g. Belgium, Slovenia) or for countries, remote to the major emission sources and impacted by the long-range pollution (e.g. Norway, Iceland). Also for coastal countries, CLRTAP_SR and CAMS_SR sometimes disagree about the importance of international shipping emissions (which is probably to be expected given differences in EMEP and CAMS emission data). The latter also applies to the country-to-city SRs for coastal cities.

PM_{2.5} contributions from the 1st contributor are systematically somewhat higher in CLRTAP_SR than CAMS_SR. This is particularly pronounced for cities (Figure 4.1a) and to a lesser extent for countries (Figure 4.1d). In contrast, the 2nd and 3rd contributors are lower in

CLTRAP_SR compared to CAMS_SR, with this difference again being more pronounced for cities (Figure 4.1b-c) than countries (Figure 4.1e-f). The main reason for these systematic differences is probably the thinner lowest layer used in CLTRAP_SR compared to CAMS_SR (50 m vs. 92 m), which means that the ground-level emissions are distributed within a thinner layer and closer to the surface. Therefore in CLTRAP_SR, the pollutant levels will be higher close to the sources (as in the case of the 1st contributor, which is typically domestic emissions). On the other hand, they will be more efficiently removed by dry deposition and thus contribute less to the long-range (transboundary) transport. Similar results are also found for PM₁₀ SRs (Figure 4.2). Nevertheless, PM concentrations due to the top-three contributors from CLTRAP_SR and CAMS_SR calculations deviate by less than 30 % in between 62 and 85% of city-receptors and in between 74 and 84 % of country-receptors, with somewhat better agreement found for the 2nd and 3rd contributors. The disagreements greater than 50% are found for less than 10-15% contributors (see Fig. 4.1 and 4.2).

4.4.2 Ozone

For ozone, we see a larger degradation of correlation between CLTRAP_SR and CAMS_SR results from the 1st to the 2nd contributor, and further to the 3rd (Table 4.2), which indicates larger uncertainties in the estimation of transboundary contributors due to non-linearities in ozone formation.

Unlike PM, emissions from the country itself are the 1st contributor to its mean ozone concentration in less than half the countries (48 and 45% of cases in CLTRAP_SR and CAMS_SR). However, domestic emissions appear to be the largest source of precursors of ozone in cities, i.e. in 75 and 73 % of the considered cities according to CLTRAP_SR and CAMS_SR respectively. In the cities and countries where the 1st contributor is not the home country, the 1st contribution is also relatively small (Figure 4.4c,f).

The 1st contributor to ozone often has a negative contribution, i.e. causes destruction (titration) of ozone, in both CLTRAP_SR and CAMS_SR (Figure 4.3a,d). Negative contributions to ozone concentrations indicate NO_x-saturated (or VOC-limited) regime, leading to ozone titration in many cities/countries. Negative contributions to ozone from the home country can occur when 1) ozone concentrations, formed from domestic NO_x and NMVOC emissions, decrease with increasing NO_x emissions; and/or 2) when domestic NO_x emissions titrate transboundary ozone.

For cities, the 1st contribution is negative in more than 80% cases. The 2nd and 3rd contributions are both positive and negative in both CLTRAP_SR and CAMS_SR calculations. CLTRAP_SR appears to calculate smaller ozone titration than CAMS_SR (slightly smaller negatives in Fig. 4.3). CLTRAP_SR also shows a tendency for larger positive contributions (though there are a few outliers with large negative contributions). Those results can at least partly be explained by a slightly higher general level of ozone concentrations in CLTRAP_SR using the CloudJ scheme. For cities in particular, the coarser horizontal resolution in CLTRAP_SR, as well as the area-weighting of grid cells partly within the city, could also contribute to reduced titration of ozone for the city average compared to CAMS_SR, leading to higher ozone contributions from the home country.

For ozone, there are more cases than for PM where CLTRAP_SR and CAMS_SR disagree about the top-three contributors, in particular for country-receptors. The matches in the identification of the main contributor to ozone is found for 52% of country receptors and 76% of city receptors (Table 4.2). The decrease in matches for the 2nd and 3rd contributors is large for ozone compared to that for PM. The agreement between CLTRAP_SR and CAMS_SR in the identification of the 2nd and 3rd important country-sources goes down (from 52) to 32 and

29% in the source allocation for countries, and down (from 76) to 43 and 38% in the source allocation for cities.

The correspondence between CLRTAP_SR and CAMS_SR calculated ozone contributions appears somewhat worse than that for PM (see explanations below). With respect to the value of the 1st contribution, the agreement within 30% is for 52% of countries and 60% of cities. Regarding the 2nd contribution, the agreement within 30% is for 45% countries and 32% cities, and for the 3rd contribution - for 38% countries and 42% cities.

4.4.3 Why is the match between CAMS_SR and CLRTAP_SR 1st contributor worse for ozone than PM?

One of the major differences in SR for ozone compared to PM is that boundary conditions (due to hemispheric transport) and Others (mostly due to BVOC and soils NO_x) are the dominating sources of average ozone in cities and especially countries. Thus, even the largest contributions from European countries to ozone concentrations are relatively very small, i.e. the sum of top-three contributions is mostly within $\pm 15\%$ for cities-receptors and within $\pm 10\%$ for countries-receptors, while top-three major country-sources contribute to PM with up to 75-80% for cities and up to 90-95% for countries. Obviously, even small differences in contributions calculated with CLRTAP_SR and CAMS_SR may affect their ranking of countries as sources for ozone.

The lower percentage of cities and countries where CAMS_SR and CLRTAP_SR identify the same source as the 1st contributor to ozone concentrations can to a large extent be explained by ozone having a smaller 1st contribution which is closer to the magnitude of the 2nd contribution. This is shown in Figure 4.4, where we plot the magnitude of the 1st contribution against the ratio of 1st and 2nd contribution in CAMS_SR, showing separately the cases where CAMS_SR and CLRTAP_SR match or not with regard to the 1st contributor. For ozone, most of the mismatches are cases where the 1st contribution is less than $\pm 2 \mu\text{g}/\text{m}^3$ and the 2nd contributor is more than half the magnitude of the 1st contributor (panels c and f). Since the contributions are of the same magnitude, the ranking of them is very sensitive to small changes.

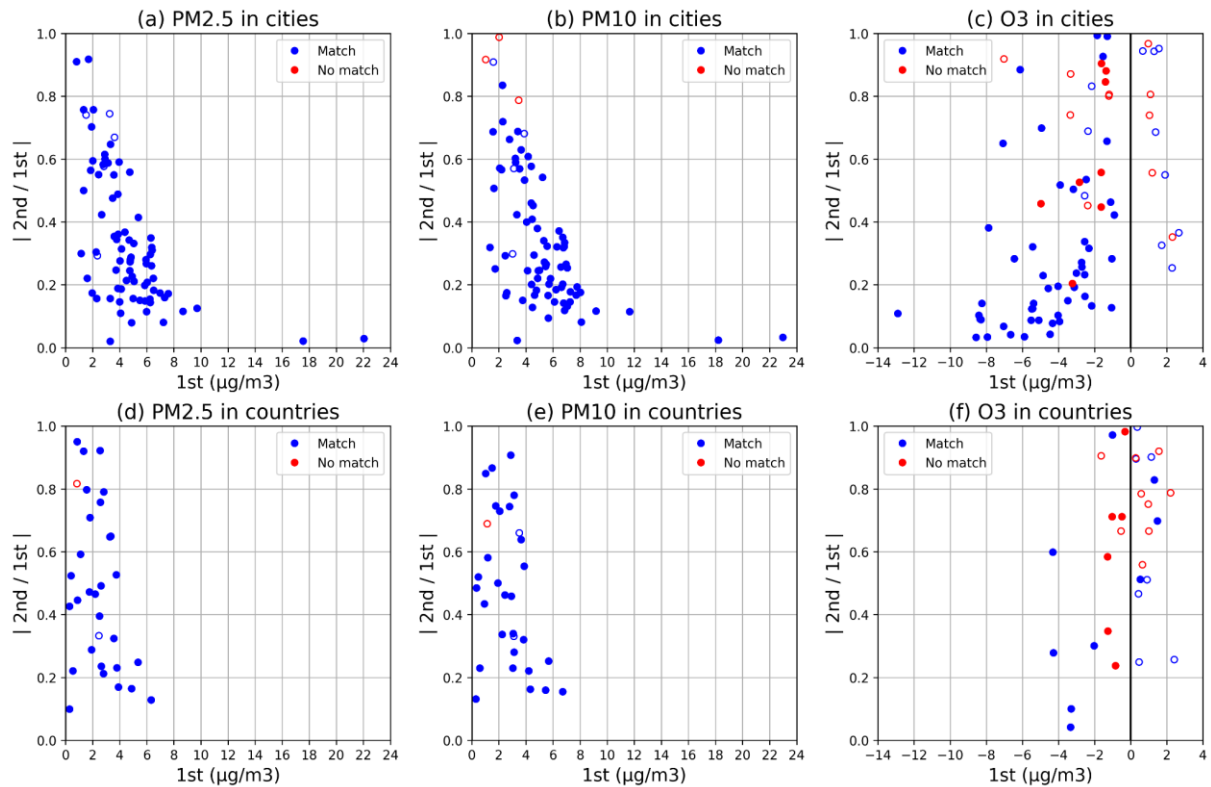


Figure 4.4: Comparison of the magnitudes of the largest (1st) vs. second-largest (2nd) contribution to PM_{2.5}, PM₁₀ and ozone in CAMS_SR, for cities (upper panels) and countries (lower panels): The x-axis gives the 1st contribution, and the y-axis gives the ratio of the 2nd and 1st contributions (absolute value). A y-value close to 1 means that the 2nd contribution is almost as important as the 1st contribution in CAMS_SR, while a y-value close to 0 means that the 1st contribution is dominating. Blue circles are cities/countries where the 1st contributor in CAMS_SR and CLRTAP_SR match, and red circles are cities/countries where they don't match. Filled circles indicate that the 1st contributor in CAMS_SR is the home country.

We also see that in nearly all cities where the 1st contribution to ozone is large and negative, the 1st contributor is the home country (indicated by filled circles) and CAMS_SR and CLRTAP_SR agree on that (panel c). For cities and countries where the 1st contributor is positive, it is usually not the home country in CAMS_SR and often points to a different source than CLRTAP_SR.

For PM_{2.5} and PM₁₀, there are also some cities and countries where the 1st and 2nd contributors are small and of similar magnitude (circles in upper-left corners of panels a,b,d,e). We see that it is here the few mismatches in 1st contributor for PM occur, and also the cases where the 1st contributor to PM is not the home country. However, CAMS_SR and CLRTAP_SR match in more of these cases compared to ozone. A probable explanation for this is that ozone as a purely secondary pollutant is more sensitive to the model setup, as opposed to PM₁₀ and PM_{2.5} which have important primary contributions. Thus, the difference in SR results calculated by reducing all precursors simultaneously vs. one at a time is probably larger for ozone than PM. Furthermore, for the home country, the annual mean contribution to ozone will be a combination of periods where it has a negative and positive contribution, which cancel out. Changes to the model setup and emissions can therefore be expected to have a larger relative impact on the annual mean ozone contributions than on annual mean PM contributions.

So in conclusion, since the mismatches between 1st contributor to ozone in CAMS_SR vs. CLRTAP_SR are mostly in cases where the contributions are small, we consider that the CAMS_SR and CLRTAP_SR are also fairly consistent for ozone.

4.5 Conclusions and recommendations

In the sections above, country-to-country and country-to-city SR results for 2022 from the EMEP model, calculated within CLRTAP and CAMS projects, were compared for PM and ozone. The underlying differences in SR run setups are believed to be responsible for much of the discrepancies in the calculation of top-three contributions to the pollution in countries and cities. Unfortunately, due to rather different model versions and input data, it was not feasible to identify the effect of different execution of emission reductions (all emissions in the same run in CAMS_SR vs individual emission reductions in separate CLRTAP_SR runs) on SR products.

In general, we find the results in terms of top-three pollution contributors from CLRTAP_SR and CAMS_SR to be reasonably consistent, given differences in the computational settings. The deviations are well within 30% for PM. Somewhat larger differences for ozone are not surprising due to rather small magnitudes of the contribution from European countries compared to ozone pollution originated beyond Europe and ozone formed from biogenic precursors.

While considering the potential co-benefits of complementary/interchangeable use of CLRTAP_SR and CAMS_SR, one should keep in mind the differences in these operational SR products with respect to the timeline of operational production, meteorological and emission year, model version, output parameters and frequency (see Table 4.3). The bottom line is that the advantage of CAMS_SR products is that they become available almost 2 years earlier than those from CLRTAP_SR, but they are based on older emissions and typically on older model versions.

Table 4.3: Differences in operational SR products

CLRTAP_SR:	CAMS_SR:
<ul style="list-style-type: none"> • SR for 2 years back • up-to-date model version • EMEP emissions for actual year • More parameters (PM_x, EC, PPM_x, • O₃ indicators, S and N deposition) 	<ul style="list-style-type: none"> • SR for current year (and earlier years) • older model version • CAMS emissions for 2 years back • Daily/seasonal/yearly • PM_{2.5}, PM₁₀, O₃, O₃ MDA8, NO₂

Thus, given a fair general consistency between CAMS_SR in CLRTAP_SR products (especially for PM), but different operational timelines, some possibilities of their complimentary usage can be suggested.

4.5.1 Potential use of CAMS_SR in CLRTAP_SR activities

Preliminary SR estimates could be provided 1.5-2 years earlier (e.g. SR for 2024 could be provided on 1 January 2025). Those would not be based on EMEP official emissions, but on CAMS emissions, typically representative for 2 years back in time, but with up-to-date meteorological conditions. Furthermore, CAMS_SR would normally use a year older model version and rely on the recreation of three-dimensional precipitation from two-dimensional precipitation and liquid cloud water from IFS forecast.

Within CAMS Policy Service, the LOTOS-EUROS model also performs forecasts of source allocation using a labelling method. In principle, SR matrices based on LOTOS-EUROS source allocation calculations could be made, provided that output of gridded concentration fields are created and stored. Thus, two complementary SR products could be provided: 1. the source attribution of air pollution based on labelling method with LOTOS-EUROS model, and 2. the potential effects of emission reductions based on emission perturbation methods (brute force or local fractions) with the EMEP model.

The comparison of country-to-country SR from LOTOS-EUROS with CLRTAP_SR was not possible at this stage of the project as the necessary output was not available from LOTOS-EUROS in the CAMS Policy Service.

4.5.2 Potential use of CLRTAP_SR in CAMS Policy Service

Due to the 1.5 year time lag in SR calculations for EMEP reporting calculations, CLRTAP_SR would not be suited for NRT application. However, it could be used in Annual Assessment Reports (AAR), providing additional information on pollution sources.

AARs describe the situation that occurred two years ago and is based on CAMS air quality model results combined with validated observations provided by the European monitoring networks. AARs aim at providing the best estimate of air pollutants concentration patterns and levels in Europe over the target year. The results for ozone, nitrogen dioxide, and particulate matter are interpreted with respect to the limit, objective and target values set in the European Air Quality Directives and air quality guidelines recommended by the World Health Organization (WHO). The assessments of air pollution are presented for annual and seasonal averages, and discussed focusing on the most polluted areas and European hotspots and the year-to-year changes in limit value exceedances since 2013.

Source-receptor results from CLRTAP_SR calculations could be used to identify the emission sources (countries) contributing to enhanced pollution in the countries of interest. SR tables could be provided for all major pollutants and indicators (i.e. PM_{2.5}, PM₁₀, O₃, O₃ MDA8, SOMO35, AOT40, and NO₂). Operationally, CLRTAP_SRs are calculated on an annual basis, but seasonal SRs could also be prepared if requested. Based on the latest version of EMEP model, meteorology from IFS reanalysis and EMEP emissions for the actual year, CLRTAP_SR results would provide quite relevant information, identifying the major sources of pollution and estimating potential impacts of emission reductions.

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Wind, P. van Caspel, W.: Generalized local fractions -- a method for the calculation of sensitivities to emissions from multiple sources for chemically active species, illustrated using the EMEP MSC-W model (rv5.5) *Geosci. Model Dev.*, Submitted 15th Nov 2024.

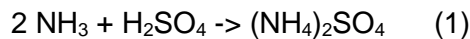
Annex A - A simplified explanation of the NH₃-(NH₄)₂SO₄-HNO₃-H₂SO₄ non-linearities

Non-linearities can be positive or negative depending on the relative concentrations of NH₃, NO₃⁻, HNO₃ and H₂SO₄, which we explain below:

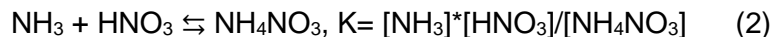
1. Simplified chemistry

A simplified version of the chemistry involving NH₃, H₂SO₄, HNO₃ and NH₄NO₃ can be presented in this way (where [x] means molar concentration of specie x):

First all ammonia and sulphuric acid available reacts to form ammonium sulfate (in reality different forms of ammonium sulfates are formed):



All H₂SO₄ is consumed by NH₃, or vice versa. If H₂SO₄ is in excess, then all NH₃ will be consumed and no NH₃ is available for further reaction. If there is any NH₃ left after reaction 1. has taken place, then we refer to NH₃ as being in excess of H₂SO₄. In that case, excess NH₃ will react with HNO₃ in an equilibrium reaction:



Since this is an equilibrium, the formation of NH₄NO₃ will be directly related both to the concentration of NH₃ and HNO₃

2. When all emission reductions (NO_x, SO_x, NH₃, NMVOC, PPM) are performed simultaneously:

Consider the case where all reductions are performed simultaneously (which is how it is done in the present CAMS Policy city SR service):

i) Typical summer situation with NH₃ in excess of H₂SO₄:

15% reduction simultaneously in NO_x, SO_x, NH₃ gives:

$$5 \quad [(\text{NH}_4)_2\text{SO}_4] = 0.85 * [\text{H}_2\text{SO}_4] \text{ (since H}_2\text{SO}_4 \text{ is the limiting factor)}$$

$$6 \quad K * [\text{NH}_4\text{NO}_3] = (0.85 * [\text{NH}_3] - 2 * [(\text{NH}_4)_2\text{SO}_4]) * 0.85 * [\text{HNO}_3] = 0.85^2 [\text{NH}_3 - 2 * \text{H}_2\text{SO}_4] * [\text{HNO}_3]$$

Concentration of NH₄NO₃ is 28% (1-0.85² = 1-0.72) lower with 15% emission reductions, i.e. substantially **more efficient reductions** in NH₄NO₃ than 15% and **negative non-linearities** in NH₄NO₃.

ii) When NH_3 concentrations are lower than H_2SO_4 , then a 15% reduction in SO_x emissions (and then roughly also H_2SO_4) will not impact $(\text{NH}_4)_2\text{SO}_4$ (since NH_3 is the limiting factor), but the 15% reduction in NH_3 would reduce $(\text{NH}_4)_2\text{SO}_4$ by ca 15%. Reducing NO_x leads to less HNO_3 , but since there is no NH_3 available (a lot of the time), no NH_4NO_3 formation can be reduced anyway, leading to less efficient reductions in NH_4NO_3 and positive non-linearities.

iii) When NH_3 concentrations are of the same magnitude as H_2SO_4 , then a 15% reduction in SO_x emissions (and then H_2SO_4) will result in H_2SO_4 consuming 15% less NH_3 , compensating the 15% NH_3 emission reduction, and the same amount of NH_3 can enter the equilibrium reaction with HNO_3 to form NH_4NO_3 . With $K = \frac{[\text{NH}_4\text{NO}_3]}{[\text{NH}_3][\text{HNO}_3]}$, one would expect NH_4NO_3 to scale roughly with the 15% NO_x emission reductions (HNO_3), and only smaller non-linearities are expected.

Another perspective to ii) and iii) is that when you reduce NH_3 and the concentration is close to that of H_2SO_4 , then reducing NH_3 will sometimes reduce NH_4NO_3 and sometimes reduce NH_4SO_4 , leading to less efficient reductions in NH_4NO_3 .

3. When emission reductions (NO_x , SO_x , NH_3 , NMVOC , PPM) are performed individually:

i) Typical summer situation with NH_3 in excess of H_2SO_4 :

the reduction will be (approximately) additive, i.e. it does not matter (much) if you reduce NO_x , SO_x and NH_3 individually in different model runs or together. Therefore, the non-linearities here are similar to the non-linearities in sector SR, when the different sector runs are added together, as discussed in Section 3.5.

This can be seen from:

15% reductions in NH_3 gives (ca 15% reduction in NH_4NO_3):

$[(\text{NH}_4)_2\text{SO}_4] = \text{H}_2\text{SO}_4$ (since H_2SO_4 is the limiting factor) Assuming NH_3 is still in excess after 15% reduction

$K * [\text{NH}_4\text{NO}_3] = [0.85 * \text{NH}_3 - (\text{NH}_4)_2\text{SO}_4] * [\text{HNO}_3] \sim 0.85 [\text{NH}_3] * [\text{HNO}_3]$ when $\text{NH}_3 \gg \text{H}_2\text{SO}_4$

15% reductions in SO_x emissions (and H_2SO_4) gives (ca 15% reduction in NH_4NO_3):

$[(\text{NH}_4)_2\text{SO}_4] = 0.85 * \text{H}_2\text{SO}_4$ (since H_2SO_4 is the limiting factor) Assuming NH_3 is still in excess after 15% reduction

$K * [\text{NH}_4\text{NO}_3] = [\text{NH}_3 - 0.85 * \text{H}_2\text{SO}_4] * [\text{HNO}_3] \sim [\text{NH}_3] * [\text{HNO}_3]$ when $\text{NH}_3 \gg \text{H}_2\text{SO}_4$

CAMEO

15% reductions in NO_x emissions (and HNO₃) gives (ca no effect on NH₄NO₃):

$[(\text{NH}_4)_2\text{SO}_4] = \text{H}_2\text{SO}_4$ (since H₂SO₄ is the limiting factor)

$K^*[\text{NH}_4\text{NO}_3] = [\text{NH}_3 - \text{H}_2\text{SO}_4] * 0.85[\text{HNO}_3] \sim 0.85[\text{NH}_3] * [\text{HNO}_3]$ when NH₃ >> H₂SO₄

Assuming NH₃ >> H₂SO₄, the emission reductions are additive.

Note that it is more efficient to reduce NH₃ to curb PM_{2.5} /PM₁₀ when NH₃ is NOT in excess of H₂SO₄, as NH₃ forms (NH₄)₂SO₄ directly (not in equilibrium), but that should not be mixed with the linearity issue discussed here.

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