

Thank you for your constructive suggestions which have been very helpful in improving the manuscript. We have been keen to follow them up. Please find below a point-to-point reply to the comments.

Reviewer 1

General Comments:

The authors conduct year-long model simulations over Europe with the EMEP CTM to intercompare four emission inventories in order to capture the uncertainty in FAIRMODE air quality metrics for PM and Ozone, focusing on emission control scenarios on specific urban centres and conglomerates. This is timely given the recent update of WHO guideline values, as well as the EU policy targets for improving air quality, and relevant within the scope of ACP (Methods for assessment of models).

The language throughout the manuscript should be improved to be made more fluent and precise, and avoid repetition.

We went through the manuscript carefully, removed repetitions and improved the text where possible.

In particular the abstract should be re-written to more clearly state the objectives and outcomes, avoid repetitions and include quantitative as well as qualitative comparisons.

We replaced the abstract with the following text.

Despite the application of an increasingly strict EU air quality legislation, air quality remains problematic in large parts of Europe. To support the abatement of these remaining problems, a better understanding of the potential impacts of emission abatement measures on air quality is required and air chemistry transport models (CTMs) are the main instrument to perform emission reduction scenarios. In this study, we study the robustness of the model responses to emission reductions when emission input is changed. We investigate how inconsistencies in emissions impact the modelling responses in the case of emission reduction scenarios. Based on EMEP simulations over Europe fed by four emission inventories: EDGAR 5.0, EMEP-GNFR, CAMS 2.2.1 and CAMS version 4.2 (incl. condensables), we reduce anthropogenic emissions in six cities (Brussels, Madrid, Rome, Bucharest, Berlin and Stockholm) and 2 regions (Po Valley Italy and Malopolska Poland) and study the variability of the concentration reductions obtained with these four emission inventories. Our study reveals that the impact of reducing aerosol precursors on PM₁₀ concentrations result in different potentials and potencies, differences that are mainly explained by differences in emission quantities, differences in their spatial distributions as well as in their sector allocation. In general, the variability among models is larger for concentration changes (potentials) than for absolute concentrations. Similar total precursor emissions can however hide large variations in sectorial allocation that can lead to large impacts on potency given their different vertical distribution. PPM appears to be the precursor leading to the major differences in terms of potentials. From an emission inventory viewpoint, this work indicates that the most efficient actions to improve the robustness of the modelling responses to emission changes would be to better assess the sectorial share and total quantities of PPM emissions. From a modelling point of view, NO_x responses are the more challenging and require caution because of their non-linearity. For O₃, we find the relationship between emission reduction and O₃ concentration change shows the largest non-linearity for NO_x (concentration increase) and a quasi-linear behaviour for VOC (concentration decrease).

We also emphasize the importance of accurate ratios of emitted precursors since these lead to changes of chemical regimes, directly affecting the responses of O3 or PM10 concentrations to emission reductions.

The authors should make clear how this study is related and complementary to Thunis et al. (2022) where emission inventories for 150 cities are investigated?

In Thunis et al. (2022), inconsistencies were only analysed at the level of the emission inventory. In this work we go one step further by assessing how these emission inconsistencies impact the model responses to emission reduction scenarios. We check whether differences in model results arise mainly from inconsistencies in emission (input data) or from the model itself. We also introduced the concept of the ensemble (median) to facilitate the comparison.

We added this to the manuscript in section 2.

Specific Comments:

In Sec. 2.2 it would be good for the reader if a table is added summarising the species present in each inventory and highlighting the differences, e.g. resolutions, bottom-up/country-totals methodology, compilation year etc.

We added the following to the manuscript:

Table 1. Overview of the four emission inventories used in this study.

Name inventory	Resolution (lon x lat) in degrees	Method	Release date	Sector classification	Condensables included	Total NOx emissions*	Total SOx emissions*	Total PM25 emissions*	Total NH3 emissions*
Edgar_v5.0	0.1 x 0.1	Bottom-up	2020	13 GNFR	No	6360	4074	1278	5116
EMEP	0.1 x 0.1	Country report	2018	13 GNFR	No	7445	2591	1229	3663
CAMS 2.2.1	0.1 x 0.05	Country report	2018	13 GNFR	No	6410	2513	1272	3708
CAMS4.2C	0.1 x 0.05	Country report	2022	12 GNFR	Yes	6419	2519	1688	3640

*Total emissions for Austria, Belgium, Bulgaria, Denmark, Finland, France, Greece, Hungary, Ireland, Italy, Luxembourg, Netherlands, Poland, Portugal, Romania, Spain, Sweden, Estonia, Latvia, Lithuania, Czech Republic, Slovakia, Slovenia, Croatia, Cyprus, Malta and Germany in Ktons/year.

The anthropogenic emissions in the four inventories are: CO, NOx, SOx, NH3, VOC, PM25, PM10. Edgar uses a bottom-up approach for all emission source sectors, based on estimates of activity data and emission factors whereas CAMS is mainly based on countries reported emissions. The differences between the same years between the CAMS inventories stems from the recalculations of the pollutants for each country.

In 2.2.4 could you please make more clear in the text how the "condensables" are different to previously reported PM2.5/10 and to quantify the expected differences, also for the examples of Poland and Turkey?

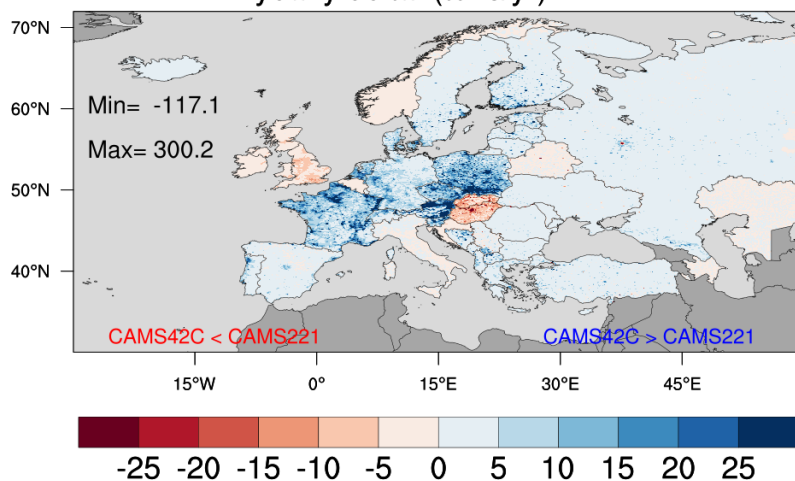
We changed the text of section 2.2.4 to emphasize the impact of condensables with respect to previously reported emissions. The text now reads as:

This inventory (Kuenen et al., 2021, 2022) is an update of the previous CAMS versions for PM emissions for the residential sector, also known as REF1, in which PM2.5 and PM10 emissions have

been updated with information on the condensable part (personal communication J. Kuenen, TNO, 2021). This inventory, also known as REF2, is hereafter denoted as CAMS42C. Condensables replace country reported PM2.5 and PM10, with a bottom-up estimate for small combustion for all fuels (not only wood but also for fossil fuels). Since 2016, more and more countries gradually included condensable emissions of small combustion devices, leading to significant differences as shown by Kuenen et al. (2022). For example, in countries such as Poland and Turkey where coal combustion in households is still an important contributor to PM, large emissions of fine and coarse condensables (118kTons/year for PM25) still take place. For Turkey the difference in PM2.5 emissions for GNFR Sector 03 is around 20% (higher in CAMS42C). For Hungary, Slovakia, Ireland, UK, Belgium and Norway the PM2.5 emissions for GNFR Sector 03 are in general lower than in CAMS42C.

The figure below (not added to the manuscript) shows the differences in PM2.5 emissions for Sector 3 (Domestic heating) between CAMS42C and CAMSv211. Over Poland, France, Slovenia and parts of the Netherlands we see large differences between the two inventories. For example, for Poland CAMS42C has about three times more PM2.5 emissions (~118kTons/year) than CAMS221. For Turkey the difference in PM2.5 emissions for GNFR Sector 03 is around 20% (higher in CAMS42C). For Hungary, Slovakia, Ireland, UK, Belgium and Norway the PM2.5 emissions for sector 3 are in general lower in CAMS42C, as illustrated in the figure below.

DIFF PM25_sec03 CAMS42C vs CAMS221 yearly delta (tons/yr)



PM25 Sec03 (tons/year)	CAMS 2.2.1	CAMS42+C
Turkey	120893	145168
Poland	65421	184363

In Sec. 2.4 the screening method should be better motivated. With which criteria were the user-defined thresholds decided? The first 3 paragraphs read as a user guide for the FAIRMODE output diagram in Fig. 3 rather than a discussion on statistical screening - please consider rephrasing.

The thresholds are arbitrary but should ideally be chosen in such a way to distinguish uncertainties from inconsistencies. Too small differences will not allow to distinguish between inconsistency and uncertainty whereas large enough differences must be considered as inconsistencies. It must be noted that these thresholds can be lowered as the analysis proceeds and inconsistencies are

progressively solved or explained. We added some sentences in the manuscript to explain these points. Regarding the 3 first paragraphs of Section 2.4, we believe they are necessary to understand the basic principles underlying our approach.

Sec. 3.1: What were the selection criteria for the cities, regions included in the study? Would it be possible to include additional locations (even to expand to all major European cities) to make the study even more encompassing and robust? (For example in Thunis et al. (2022) emission inventories for 150 cities are investigated - can this be done here or in a subsequent study to statistically assess the effectiveness and impacts of EU-wide measures?)

The impact of emission reductions on concentrations is calculated in one single simulation for all cities/regions. The areas where emission reductions are applied are therefore selected in such a way that they are far away from each other to avoid that reductions applied over one area influence the background concentration levels in another, which would hamper our analysis. In this context, extending the analysis to 150 cities is not possible. We added this to the text in section 2.

Sec. 3.2: Are the aerosol processes (secondary production) reported included and captured by the CTM model used? Are natural aerosols such as dust included in the modelled PM10?

Secondary aerosol formation is included in the EMEP model, together with biogenic VOC and Dimethyl Sulphide (DMS) emissions, together with natural dust (e.g. windblown dust from deserts, semis-arid areas, agricultural and bare lands, and Saharan; Simpson et al. 2012).

We added to Section 2.1 that secondary aerosol formation in the EMEP model is included. We removed the first two paragraphs of section 3.2 "Variability of PM10 .." as suggested by Reviewer 2.

Sec. 3.5 can be merged with Sec. 3.6: the ratio of NO_x/VOC is important - are the urban centres studied here NO_x/VOC limited in terms of O₃ production/sink? Is there a different seasonal dependence in the model results from city to city that would be important for air quality plans?

We disagree with the reviewer regarding Section 3.5 and 3.6 that deal with different aspects. On the other hand, we agree that seasonal temporal variations in the emission inventories are important on gas and aerosol calculations as shown in De Meij et al. 2009. Clappier et al. (2021) analysed the seasonal variation of the chemical regimes all over Europe. We've added the following to the text "Clappier et al. (2021) showed which chemical regimes are responsible to the secondary inorganic PM formation over Europe, and how these chemical regimes can help in designing efficient PM abatement strategies. They showed that during wintertime, PM₂₅ concentrations are predominantly NH₃-sensitive in the major part Europe. During summertime, PM₂₅ are predominantly SO₂-sensitive in most of Europe."

Regarding the reviewer's question on the dependence from city to city, in general, city centres are VOC-limited due to the abundance of NO_x emissions caused by road transport. Clappier et al., (2021) showed that for VOC-limited O₃ formation regime areas, where NO_x emission reductions of 50% lead to substantial increases in O₃ in wintertime due to a decreased titration of O₃ by NO Clappier et al. (2021).

Furthermore, the seasonal and geographical dependency on O₃ formation/depletion is addressed in a joint paper in FAIRMODE (accepted in Journal Air Quality, Atmosphere & Health), that describes

the impact of short-term emission reductions on the calculated O₃ concentrations for different cities in Europe.

Sec. 3.7: Given the difference in behaviour in specific regions how are the results of this study to be interpreted? Can the EMEP model provide a map of the different chemical regimes across European cities/regions for each inventory? What is the non-linear behaviour regarding reduction of both NO_x and VOC?

Thanks for pointing out this issue. We agree with the reviewer that chemical regimes can greatly vary between seasons and between different locations in Europe. The non-linear behaviour on O₃ formation in Europe is studied by Beekmann and Vautard (2010), who provide a comprehensive study on O₃ formation chemical regimes over Europe. They showed that during summer time, VOC-limited regimes are present especially over urbanized areas while NO_x sensitive chemical regimes occur over southern Europe.

Reducing NO_x and VOC emissions together also shows the non-linear behaviour when NO_x and VOC emissions are reduced together by different quantities, see Table 11. The formation of O₃ is less sensitive to the reduction of NO_x emissions when simultaneously also VOC emissions are reduced. This corroborates the findings of Xiao et al., 2010, Xing et al., 2017. We have made it clearer in the text.

In addition to O₃, Clappier et al. (2021) showed which chemical regimes are responsible to the secondary inorganic PM formation over Europe, and how these chemical regimes can help in designing efficient PM abatement strategies. They showed that during wintertime, PM_{2.5} concentrations are predominantly NH₃-sensitive in the major part Europe. During summertime, PM_{2.5} are predominantly SO₂-sensitive in most of Europe.

Thunis et. al (2021) showed that the peculiarity of secondary PM_{2.5} formation in the Po basin, which is characterised by contrasting chemical regimes within distances of a few (hundred) kilometres, as well as non-linear responses to emission reductions during wintertime.

Beekmann, M. and Vautard, R.: A modelling study of photochemical regimes over Europe: robustness and variability, *Atmos. Chem. Phys.*, 10, <https://doi.org/10067-10084>, 2010.

A. Clappier, P. Thunis, M. Beekmann, J.P. Putaud, A. de Meij, Impact of SO_x, NO_x and NH₃ emission reductions on PM_{2.5} concentrations across Europe: Hints for future measure development, *Environment International*, Volume 156, 2021, ISSN 0160-4120, <https://doi.org/10.1016/j.envint.2021.106699>.

Thunis, P., Clappier, A., Beekmann, M., Putaud, J. P., Cuvelier, C., Madrazo, J., and de Meij, A.: Non-linear response of PM_{2.5} to changes in NO_x and NH₃ emissions in the Po basin (Italy): consequences for air quality plans, *Atmos. Chem. Phys.*, 21, 9309–9327, <https://doi.org/10.5194/acp-21-9309-2021>, 2021.

Sec.4 Please consider using paragraphs rather than bullet points to present the findings.

We decided to keep bullet points to improve readability of the main findings but added a final concluding paragraph. Note that the bullet points were appreciated by Reviewer 2. We however added a final concluding paragraph after the bullet points.

Please clearly explain early in the text how is PMcoarse different to PM10? What is PPM and how is PPM10 and PPM2.5 different to PM10 and PM2.5? Please make sure all acronyms are properly defined and used.

Particles between 2.5 and 10 micrometers in diameter are referred to as PM coarse particles. PPM10 and PPM2.5 contain the primary component of the PM10 or PM2.5 fractions respectively. This is different from PM10, which contains also the secondary part (inorganic and organic aerosols such as sulphates, nitrates, ammonium and biogenic organic aerosols). We added this in Section 2 as suggested by the reviewer. We also checked that all acronyms are defined and consistent.

P95 should be defined when first used.

Done.

Technical Corrections: (page, line number):

p3153 newer, better -> more elaborate

Done. We added to the sentence: "contributing to smaller biases when compared to observations".

p3156 uncertainties associated to certain processes -> associated with

Corrected.

p3163 One of FAIRMODE's goal -> goals

Corrected.

p3164 explain -> investigate

Corrected.

p41121 What does it mean "only differ in terms of version"? Please clarify.

Corrected. It now reads as "release date and emission updates".

p131434 Start a new subsection so it's clear the discussion is not about VOC.

Corrected. We did the same in the section for O3 (now 3.6.3)

p131440: Can you explain why NOx has to compete with NH3 to form PM?

We rephrased the sentence and now read as:

The reduction in NO₂ concentrations leads to a reduction in HNO₃ while the increase in oxidant concentrations increases the formation of HNO₃. These two competing mechanisms effect the production of nitrate aerosol via HNO₃ + NH₃.

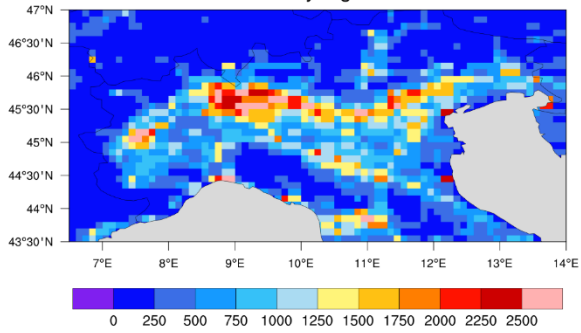
p141486: Please consider the use of a different word than dilution which has a specific meaning in chemistry that might confuse the reader (ie. rephrase "diluted by other emitted species")

Corrected and replaced by "weakened".

For the plots in the supplement pages 3-10 it would be advisable to use raster graphics showing the output on the native grid, rather than interpolated values on the map.

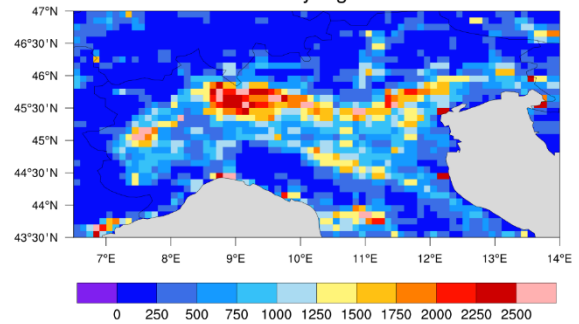
As suggested by the reviewer we corrected all the maps in Fig. S1. Below we show for illustration purposes only the new Figures S1 ac, ad, ae, and af.

PM25 total emissions CAMS221
Po Valley mg/m2



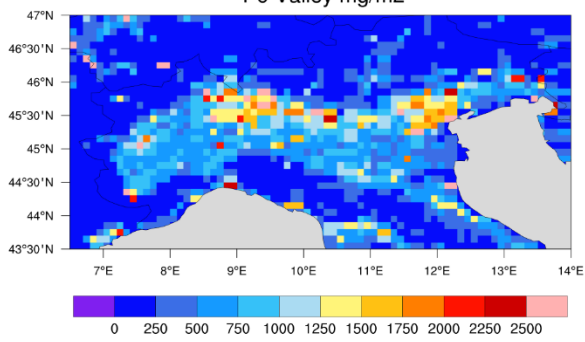
(ac)

PM25 total emissions CAMS42C
Po Valley mg/m2



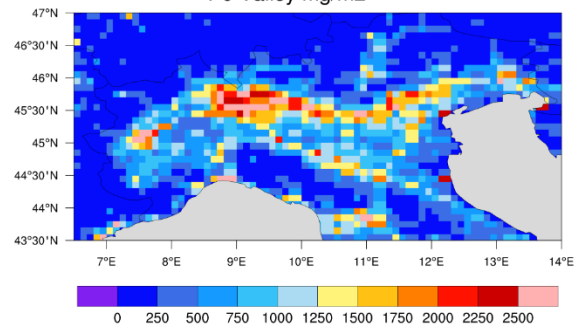
(ad)

PM25 total emissions EDGAR
Po Valley mg/m2



(ae)

PM25 total emissions EMEP-GNFR
Po Valley mg/m2



(af)