Authors' response to Reviewer 1

We sincerely thank the Reviewer for taking the time to thoroughly review our manuscript and provide relevant feedback to improve it further. Below we address their concerns and suggestions point by point.

Reviewer 1

General comment:

This article aims at proposing two methodologies for assessing impact of policy scenarios on monthly and annual pollutant concentrations ($PM_{2.5}$ and O_3) at country level on a global scale. The first approach is based on Elastic Net models and the second one on machine learning models (XGboost). Those methodologies used several datasets as inputs such as pollutant emissions, weather data, and concentrations data that are harmonized to a common a grid of $0.5^{\circ} \times 0.5^{\circ}$ from 2003 to 2021. Results show that EN models are performing well for annual total exposure, but ML models are better for evaluating the contribution of individual sector of emissions. I would like to congratulate the authors of the paper for their interesting work. The article is very well written and clearly structured. The scientific topic is of a great interest and the methodologies proposed in the paper are quite innovative considering the published approaches. However, some clarifications still need to be made in the text as highlighted in the specific comments concerning the consideration of secondary inorganic aerosols in the models and the composition of the "Other" sector of emissions for $PM_{2.5}$ estimations. Some sensitivity tests could also be performed to address the impact of these features on the final estimation as well as the effect of the proportion of the train and test sets when applying the models.

Specific comments:

Reviewer 1 Point 1 — Page 1, line 33: The following change should be made: "fine particulate matter (particles with a diameter less than $2.5 \ \mu m$, PM2.5)"

Reply: We thank the Reviewer for this comment and have added the suggested part in the text.

Reviewer 1 Point 2 — Page 2, line 4: The following change should be made: "chemistry-transport models (CTMs) are tools for calculating the impact of emissions on pollutant concentration levels"

Reply: Thank you, we agree that it would be better to specify that concentration levels refer to pollutants. We have changed the sentence accordingly.

Reviewer 1 Point 3 — Page 2, lines 7 to 10: ACT tool (Air Control Toolbox, Colette et al., 2022) should be mentioned here. ACT is a surrogate model to explore mitigation scenarios in air quality forecasts. This is designed for estimation at the European level on a daily basis.

Colette, A., Rouil, L., Meleux, F., Lemaire, V., and Raux, B.: Air Control Toolbox (ACT_v1.0): a flexible surrogate model to explore mitigation scenarios in air quality forecasts, Geosci. Model Dev., 15, 1441–1465, https://doi.org10.5194.gmd-15-1441-2022, 2022.

Reply: We thank the Reviewer for suggesting adding the ACT tool. Indeed, it is a relevant tool to discuss; we have mentioned it in the 1.0 "Introduction" Section: "The most commonly used models to evaluate air pollution policies, such as the Greenhouse Gas - Air Pollution Interactions and Synergies (GAINS) model (Amann et al., 2011; Kiesewetter et al., 2015), the SHERPA tool (Thunis et al., 2016), the TM5-FAst Scenario Screening Tool (TM5-FASST) model (Dingenen et al., 2018), and the Air Control Toolbox (ACT) tool (Colette et al., 2022), rely on a variety of methods."

Reviewer 1 Point 4 — Page 2, line 19: "The latter one is the most detailed, up-to-date reduced form air pollution model", the following clarification should be made: "on a global scale".

Reply: We agree that we should have specified the global scale reference, so we have changed the text accordingly: "The latter one is the most detailed, up-to-date reduced-form air pollution model on a global scale."

Reviewer 1 Point 5 — Page 3, line 9: Please explain further what you mean by "factors" and the impact of the monitoring of pollutant concentrations in ambient air. Perhaps, it should be mentioned that monitoring stations are used for air quality assessment and the lack of measurement points in an area is a strong constraint in that objective.

Reply: We thank the Reviewer for this comment and suggestion that we have clarified in the text. We changed the paragraph as follows: "Monitoring stations are used for air quality assessment. However, the lack of a spatially consistent large ground monitoring network in a given area is a strong constraint to achieving this objective. Despite the recent harmonization and open-access advancements in air pollution data, most publicly available global ground-level monitoring databases (e.g., OpenAQ (2024)) provide reasonable territorial coverage of the population only in developed countries, in particular in the United States and Europe. The network of ground-level monitors is growing in emerging economies such as China and India, yet urban and rural areas are largely unmonitored in middle- and low-income countries. The uneven ground-level monitoring geographical coverage is problematic, as factors driving the emissions-concentrations relationship differ between monitored and unmonitored areas (e.g., population density, distance from industrial sources, GDP per capita)."

Reviewer 1 Point 6 — Page 3, line 13: "Global, gridded reanalysis data combine and harmonize satellite air pollution measurements with ground-level monitors." The following clarification should be made: CTM estimates are also used.

Reply: We thank the Reviewer for highlighting this imprecision. We have included a reference to chemistry-transport models and changed the text to: "Global, gridded reanalysis data combine and harmonize satellite air pollution measurements and CTM output with ground-level monitors."

Reviewer 1 Point 7 — Page 3, lines 24 and 25: The following clarification should be made: "the need to homogenize different grids in terms of spatial resolution"

Reply: We agree with this suggestion and have updated the text accordingly: "Among the disadvantages, the need to homogenize different grids in terms of spatial resolution may lead to approximations during the data manipulation process."

Reviewer 1 Point 8 — Page 4, line 24: Is the odd-road transportation corresponds to shipping and aviation? Could you please clarify.

Reply: We thank the Reviewer for the opportunity to clarify this point. The "Off-road transportation" sector we employ in CLAQC includes railways and other types of non-road transports not typically used on public roads, such as agricultural machinery, construction equipment, and certain types of off-road vehicles used in industrial operations (e.g., tractors, telehandlers, excavators). It corresponds to the "Non-road transportation" (TNR) sector in the CAMS-GLOB-ANT data, to the IPCC Sector Name "Non-road ground transportation" (IPCC Sectors "Rail transport", 1A3c, and "Other transport", 1A3e), and to "SNAP 8, non-shipping and non-aviation transportation" (Granier et al., 2019, 2021; Krey et al., 2014). Therefore, it does not correspond to shipping and aviation. In general, pollutant emissions from the shipping and aviation sectors are not taken into account in CLAQC because they are beyond the scope of our analyses. We made this clearer in the text under Subsection 2.1.4. "Sectoral aggregation": "We do not include biogenic and sectoral emissions from shipping and aviation. We build the following 7 sectors from CAMS-GLOB-ANT data: Agriculture, Industry, Other (including the emissions not considered in the other sectors), Off-road transportation, Energy power generation, Road transportation, and Residential (including buildings, commercial and services). Note that the Off-road transportation sector includes railways and other types of non-road transports not typically used on public roads, such as agricultural machinery, construction equipment, and certain types of off-road vehicles used in industrial operations (e.g., tractors, telehandlers, excavators)."

Reviewer 1 Point 9 — Page 5, line 4: Are the natural emissions included in the "Other" sector? Could you please clarify.

Reply: No, the CLAQC "Other" sector does not contain biogenic emissions. CLAQC is based on anthropogenic emissions derived from the CAMS-GLOB-ANT sectors, meaning that in terms of emissions, only anthropogenic sources are included in the models. In particular, the "Other" sector contains anthropogenic emissions from the following sectors: "Solvents application and production" (SLV), and "Solid waste and wastewater handling" (SWD). Such anthropogenic emissions are summed up within

pollutants' sectoral emissions, into this residual sector, that we call "Other". We have not included biogenic emissions as CLAQC was built aiming to support policy impact. Policies generally are not likely to target non-anthropogenic sources. However, you are right in stating that those emissions in some regions of the globe are major contributors to exposure. We have clarified this in the manuscript and integrated it as a limitation of our tool subject to future improvement. In the "Limitation" Section, we have stated: "Fifth, as CLAQC was built to support policy impact evaluation, the approaches presented here do not explicitly model transboundary movements of pollution and biogenic emissions such as desert dust and sea salt". Given that earlier in the text this aspect was not discussed, we have clarified this under Subsection 2.1.4. "Sectoral aggregation" as well: "We do not include biogenic and sectoral emissions from shipping and aviation. Furthermore, natural emissions, such as desert dust and sea salt, are not taken into account since they are less likely to be subject to policy interventions".

Reviewer 1 Point 10 — Page 5, line 19: How did you manage to downscale the concentrations data to 0.5° ? Please explain further.

Reply: Thank you for asking for this clarification. We have further explained the rescaling process updating Subsection 2.4. "Grid definition" as follows: "All gridded data sources are rescaled to the same $0.5^{\circ} \times 0.5^{\circ}$ coordinate grid through linear interpolation, based on the population grid, and merged into a single data set. For instance, concentration data originally at $0.75^{\circ} \times 0.75^{\circ}$ spatial resolution are downscaled to $0.5^{\circ} \times 0.5^{\circ}$, generating intermediate values that align with the reference grid. The interpolation is implemented using the interp_like function from the xarray Python package."

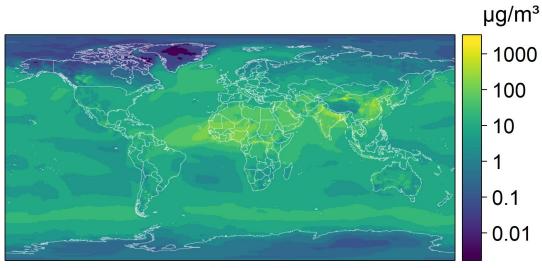
Reviewer 1 Point 11 — Page 5, lines 20 and 21: I don't understand why you must change the unit here. Aren't ECMWF data already in $\mu g/m^3$? Please clarify.

Reply: Regarding ECMWF data, the original unit of measure for GEOM Ozone is kg/kg, while for fine particulate matter is kg/m^3 . So, in both cases, we convert the unit to $\mu g/m^3$ to improve results' interpretability. We have clarified this in the text under Subsection 2.3. "Concentrations". The documentation on the unit of measurement of the ozone concentration data is available at: https://ads.atmosphere. copernicus.eu/datasets/cams-global-reanalysis-eac4-monthly?tab=overview

Reviewer 1 Point 12 — In addition, Figure 2.3 is called in the main text, but the figure numbering is Figure 1. In order to improve reading of the figure, the ticks should be added in the two colorbars to make the correspondence with the tick labels in panels a) and b).

Reply: Regarding Figure 2.3, we thank the Reviewers for noticing the mismatch in the figure naming, we have solved this issue. We also agree that label ticks would improve the readability of plots in Figure 1, so we have updated the color scales as follows.





PM_{2.5}

(b) O_3

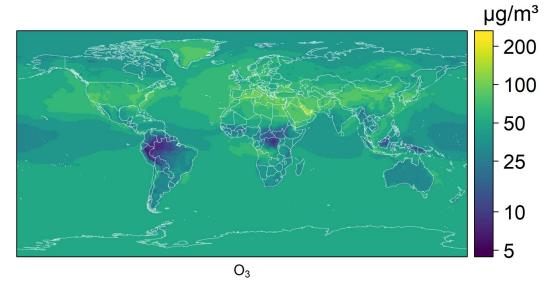


Figure 1: Level plots of EAC4 concentrations of $PM_{2.5}$ (January 2018) and O_3 (July 2018) in $\mu g/m^3$ with color bar in logarithmic scale.

Reviewer 1 Point 13 — Figure 2 title: The expression "weighted by the population" should be mentioned in the title.

Reply: To improve clarity, as suggested, we have changed the title as follows: "EAC4 concentration inputs of $PM_{2.5}$ weighted by the population and O_3 in $\mu g/m^3$ aggregated at the country level (2018)."

Reviewer 1 Point 14 — Page 7, line 25 to 30: It is a choice of simplicity to not considered secondary aerosols in the model. Have sensitivity tests carried out on the impact of this choice on the final estimation?

Reply: We apologize for not being clear on this point and have tried to explain it better in the text under Subsection 3.2. "Coefficient constraints". Secondary inorganic aerosols are not directly included in the models predicting $PM_{2.5}$ concentrations. To some extent, they can be captured through the interaction terms we had included. We had included the interactions between total pollutant emissions of $PM_{2.5}$ precursors as a proxy of secondary reactions among them. In particular, interactions between NH₃, NO_x and SO₂ enter the models (see Eq. 8, pages 9-10). We had constrained such relationships to be positive, i.e., an increase in emissions cannot lead to a decrease in PM_{2.5} concentrations. This is to avoid the models, that are designed to optimize the best predictor variables, to choose a predictor that will predict a policy to increase precursor emissions, or to avoid decreasing emissions because of small increase in secondary Imaginic aerosols, these are very local effects that tend to not occur at the country-level scale (Thunis et al., 2019; Granella et al., 2024). However, if they were to occur, the models would foresee a zero decrease instead of an increase in predicted concentrations. This means that the models would still predict the best policy path through the reduction of other precursors. We have discussed this in the 5. "Limitations" Section as follows:

"Fifth, as CLAQC was built to support policy impact evaluation, the approaches presented here do not explicitly model transboundary movements of pollution and biogenic emissions such as desert dust and sea salt. Only averages enter the models through the time and place identifiers (month-fixed effects in EN and grid identifiers in ML). Secondary inorganic aerosols (SIA) are also not directly modeled. Instead, their effects are approximated through interactions among emissions' predictors. In addition, we have not performed sensitivity analyses to assess the impact of excluding SIA on the final estimates."

Reviewer 1 Point 15 — Page 9, line 13: Have sensitivity tests been carried out on the splitting of the training and test data sets? If not, these tests should be considered.

Reply: The splitting of the training and test data sets is different across models as they have different data samples. While EN models are trained and tested on country-level aggregated data, ML models use the country-level gridded data, with a bigger sample available. Due to these data differences, we did not harmonize the data splitting across methods. Instead, we prioritized ensuring a sufficiently large training set for the EN models to minimize the variance in parameter estimates. However, we acknowledge the importance of conducting a sensitivity analysis on the choice of train-test split. Therefore, we welcome this suggestion. We would like to clarify that while we initially reported a train-test split of 80-20 for the EN models, the actual splitting used was 84-16 for models based on CAMS emissions and 80-20 for models based on DACCIWA emissions.

To provide a more robust validation of our model configurations, we have performed additional

tests by varying the train-test splits to 75-25 and 70-30, alongside the original split for EN. These sensitivity analyses confirm that our model predictions remain stable across different train-test splits. For conciseness, we present the results for the 10 countries originally provided in the manuscript. In most cases, the dominant sectors remain consistent across splits, with slight variations in the range of prediction changes. The results show that certain countries, such as Germany (DEU), Turkey (TUR), and the United States (USA), exhibit particularly robust predictions across splitting ratios for PM_{2.5}. Figures 2 and 3 illustrate these scenarios for EN models, comparing train-test splits of 84-16, 75-25, and 70-30 ratios. While there are minor variations, the overall findings remain consistent. Sectors such as Industry, Agriculture, and Residential remain stable across splits, while less dominant sectors like Off-road or Other display higher variability.

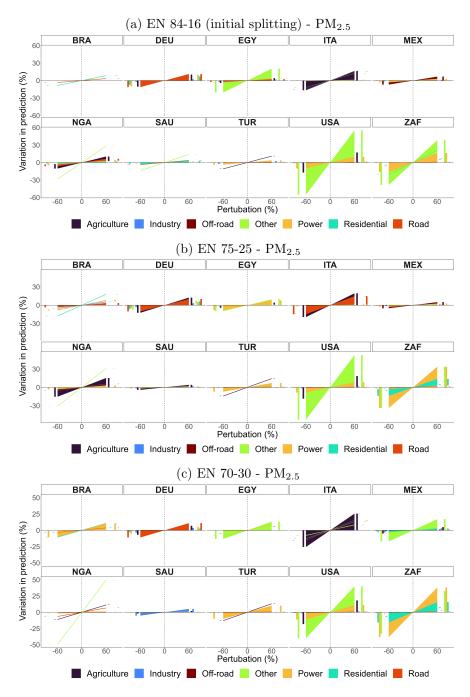


Figure 2: Percentage variation in predicted concentrations of EN models for population-weighted $PM_{2.5}$ obtained from CAMS emissions, by sector and perturbation for selected countries: 84-16 (initial splitting), 75-25, and 70-30 train-test splitting. Bar charts on the sides of each subplot help visualize overlapping variations.

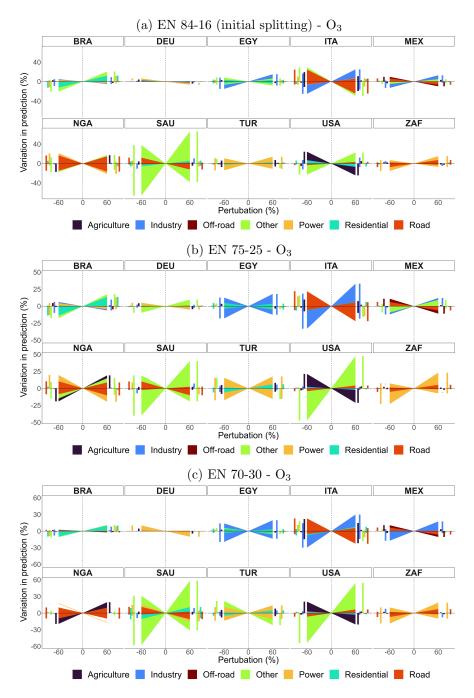


Figure 3: Percentage variation in predicted concentrations of EN models for O_3 obtained from CAMS emissions, by sector and perturbation for selected countries: 84-16 (initial splitting), 75-25, and 70-30 train-test splitting. Bar charts on the sides of each subplot help visualize overlapping variations.

Reviewer 1 Point 16 — Page 9, line 13: It is not clear if the whole period of the dataset (from

2003 to 2021) is used to train de model? Please clarify in the main text what is the periods of the train set and the test set. It is mentioned that the perturbations of emissions are applied to the last 5 years of data (page 9 line 27), thus is the model trained from 2017 to 2021?

Reply: We confirm that the whole period of the dataset (2003-2021) is used both to train and test the models for both methods. While, the last 5 years of data (2017-2021) are applied for calculating emission scenarios. We have changed the Subsection 3.3. "Elastic net models" to make this clearer: "We randomly split the 2003-2021 data into training (80% of observations) and test set (20%) stratifying by month."

Reviewer 1 Point 17 — Page 10, line 23: If the Other sector includes Natural emissions, it could have a significant impact of $PM_{2.5}$ concentrations (from desert dust and sea salt). That may bias the estimate of the machine learning model. This sector should be considered.

Reply: As specified in a previous reply, the Other sector does not consider biogenic emissions. It instead includes anthropogenic emissions derived from the following CAMS-GLOB-ANT sectors: Solvents application and production (SLV), and Solid waste and wastewater handling (SWD). We have not included biogenic emissions as CLAQC was built to support policy impact evaluation. In general, policies are unlikely to target non-anthropogenic sources. We recognize that natural emissions such as desert dust and sea salt may have a significant impact on $PM_{2.5}$ concentrations in some regions, though such emissions indirectly enter the model through the time and place identifiers. In the 5. "Limitations" Section, we have stated the following: "Fifth, as CLAQC was built to support policy impact evaluation, the approaches presented here do not explicitly model transboundary movements of pollution and biogenic emissions such as desert dust and sea salt. Only averages enter the models through the time and place identifiers (month-fixed effects in EN and grid identifiers in ML)."

Reviewer 1 Point 18 — Page 11, line 7: I'm a bit confused with the consideration of the secondary inorganic aerosol's formation in the model. It is explained in Subsection 3.2: "It is crucial to understand that in situations where secondary reactions substantially affect the overall mass of $PM_{2.5}$ within a country, our models are designed to omit these precursors from the list of predictors, thereby not reflecting a decrease in $PM_{2.5}$ levels." Please clarify if the secondary inorganic aerosols are excluded or not.

Reply: We apologize for not being clear on this point. We have addressed this concern in a previous reply to your comment, and have tried to clarify it in the text under Subsection 3.2. "Coefficient constraints". We have also added this to the 5. "Limitations" Section, where we make explicit that the model should be used under the 'fit-for-purpose' principle, i.e., for country-level policy roll-out purposes. Secondary inorganic aerosols are not directly included in the models predicting PM_{2.5} concentrations. Instead, we include interactions between specific total pollutant emissions of PM precursors as proxies

for secondary reactions. In particular, interactions between NH_3 , NO_x and SO_2 enter the models (see Eq. 8).

We have integrated the text as follows:

"While at the local scale, reducing certain precursors of secondary inorganic aerosols might not always lead to a decrease in $PM_{2.5}$ levels — due to nonlinear atmospheric reactions noted by Thunis et al. (2019); Ding et al. (2021) — our national-scale models focus on broader trends. To avoid giving undue importance to cases where local emissions reductions might result in increased levels of inorganic $PM_{2.5}$, we apply monotonic constraints between emissions and concentrations.

Rather than directly including secondary inorganic aerosols, the models incorporate interactions between PM precursors — specifically NH_3 , NO_x , and SO_2 — as proxies for secondary reactions.

It is crucial to understand that in situations where secondary reactions substantially affect the overall mass of $PM_{2.5}$ within a country, our models are designed to omit these precursors from the list of predictors, thereby not reflecting a decrease in $PM_{2.5}$ levels."

Reviewer 1 Point 19 — Page 11, line 29: Please clarify what you mean by: "We randomly split a gridded data set stratifying by grid cell. Hence, randomization occurs over the temporal dimension."

Reply: We apologize for not being clear on this point. We have changed the text as follows: "For each country-pollutant pair, the input data for ML are a grid panel dataset, composed of N grid cells and observed over T time periods. For each grid cell, we randomly assign the T observations (a time series) to the train or test set. Hence, we stratify by grid cell and randomization occurs over the temporal dimension. This stratified randomization ensures equal spatial representation in both data sets. Given unobservable but time-constant characteristics of cells (such as topography) and the desire for equal spatial representation, we prefer this method to simple randomized allocation, which might allocate the entire time series for a cell to either set. We use three-fourths of the data as the training set and the remaining fourth as the test set."

Reviewer 1 Point 20 — Page 11, line 31: Why the splitting of the training and the test data set is different from the EN model? Same question as for the EN model, were sensitivity tests on the choice of the training / test sats carried out?

Reply: We have tried to address this concern in a previous reply. To provide a more robust validation of our model configurations, we are performing additional tests by varying the train-test splits to the 80-20 and 70-30 ratios, alongside the original 75-25 split for ML as well. As for EN scenario runs, results are robust to different train-test splittings for both $PM_{2.5}$ and O_3 , with small variations. See figures 4 and 5 for variations in ML scenarios for $PM_{2.5}$ and O_3 , respectively.

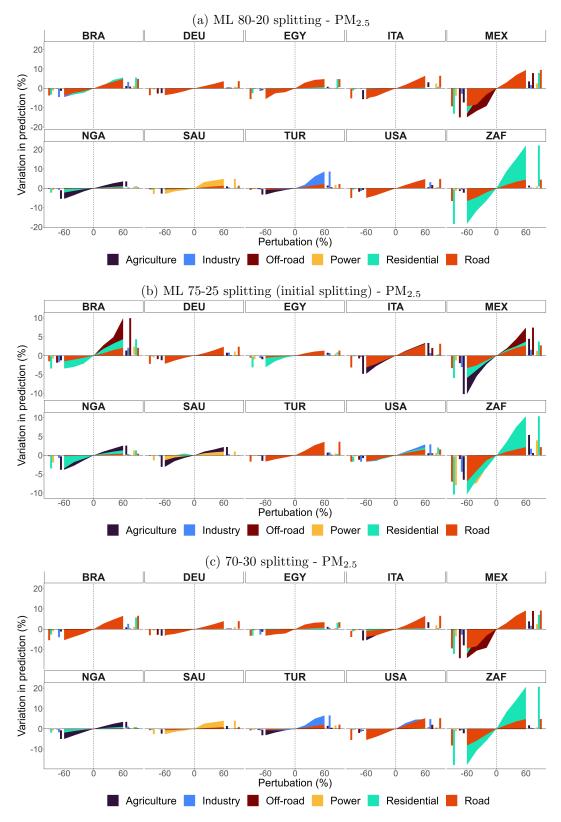


Figure 4: Percentage variation in predicted concentrations of ML models for weighted $PM_{2.5}$ obtained from CAMS emissions, by sector, pollutant, and perturbation for selected countries: 80-20, 75-25 (initial splitting), and 70-30 train-test splitting. 12

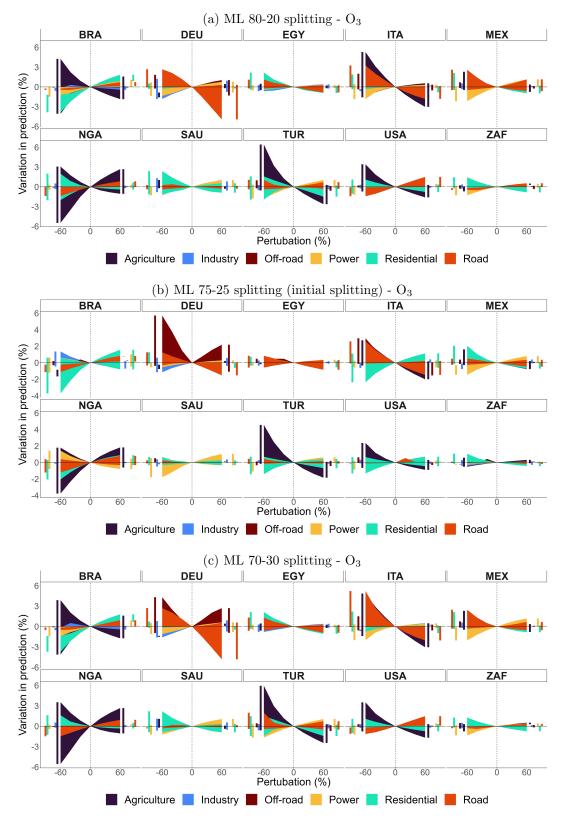


Figure 5: Percentage variation in predicted concentrations of ML models for O_3 obtained from CAMS emissions, by sector, pollutant, and perturbation for selected countries: 80-20, 75-25 (initial splitting), and 70-30 train-test splitting. 13

Reviewer 1 Point 21 — Page 12, line 2: Why not to say "Emission scenarios" instead of "Stylized scenarios" in section 4.1 title?

Reply: We welcome the Review's suggestion to simplify the Subsection 4.1. title to "Emission scenarios".

Reviewer 1 Point 22 — Page 12, line 5: Why are emission perturbations ranging to +60%, when we would expect policy scenarios to necessarily seek to reduce precursor emissions?

Reply: We thank the Reviewer for asking about this point. We have added the following paragraph under Subsection 4.1. "Model results — Stylized scenarios":

"While policies generally aim to reduce emissions, including emission-increase scenarios is crucial for a comprehensive understanding of potential air quality outcomes of a wide range of possible future conditions. For example, the persistent investments in coal in India, or the investment in gas fracking. It is important to showcase that these policy interventions may lead to exposure increases. For these reasons, we have looked at decreases and increases in emissions."

Reviewer 1 Point 23 — Page 13, line 4: Move Figure 6 in the main text. The panels of this figure are very small, and it is very difficult to read the figure correctly. It would be preferable to prepare one figure per model and per pollutant to be mor readable.

Reply: We agree on moving Figure 6 to the main text. We also welcome the suggestion of plotting one figure per pollutant and model, increasing their size to improve readability (see Figures 6 and 7). We have also corrected the x-axis plot related to EN models for predicting $PM_{2.5}$: the subplot was mistakenly displaying the range -50 to +50 instead of -60 to +60.

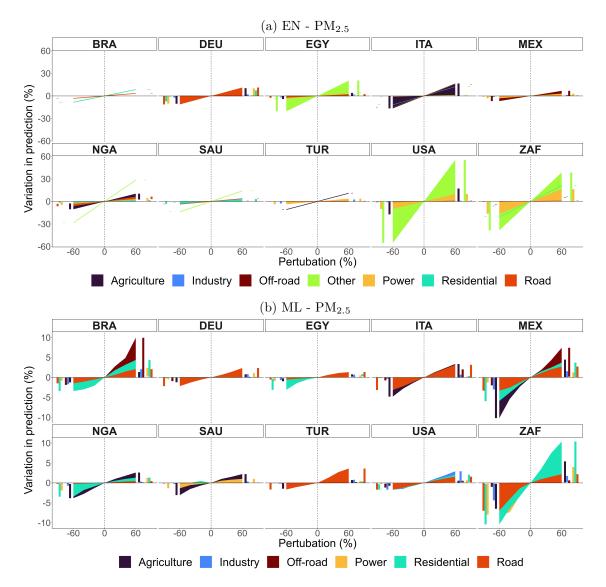


Figure 6: Percentage variation in predicted concentrations by sector and perturbation for selected countries in EN and ML models for weighted $PM_{2.5}$. Bar charts on the sides of each subplot help visualize overlapping variations.

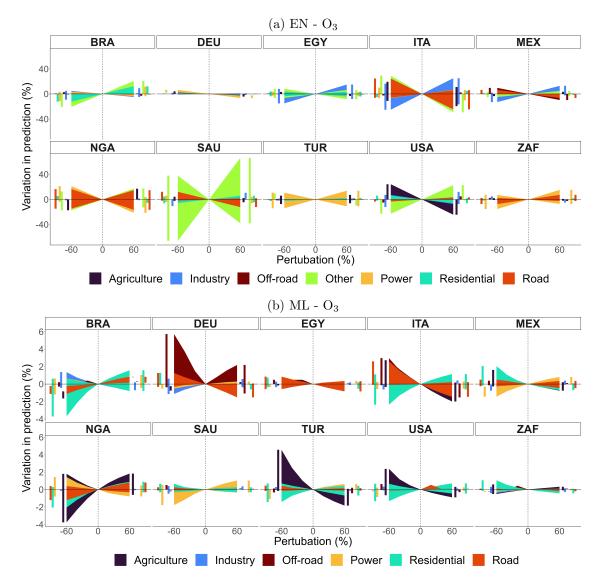


Figure 7: Percentage variation in predicted concentrations by sector and perturbation for selected countries in EN and ML models for O_3 . Bar charts on the sides of each subplot help visualize overlapping variations.

Reviewer 1 Point 24 — Page 13, line 26: Move figures 7 and 8 to the main text.

Reply: We welcome this suggestion and have changed the manuscript accordingly.

Reviewer 1 Point 25 — Page 13, line 28: Could you explain why models work better for O3 than for PM2.5?

Reply: The authors thank the Reviewer for allowing this point to be discussed. We have updated the text: "Both elastic net and machine learning models are generally better at predicting O_3 than $PM_{2.5}$ as

the former is highly correlated with incoming radiation or temperature, while the secondary chemistry of $PM_{2.5}$ is harder to grasp. Chemistry transport models predict better O_3 than PM as well, due to the more complex mixture of particles and local effects from more sources of the latter (Guérette et al., 2020)."

Reviewer 1 Point 26 — Page 13, line 42: The following clarification should be made: DACCIWA is preferred to CAMS over Africa only. (same page 14, lines 1 and 2).

Reply: We thank the Reviewer for this useful comment, on which we fully agree. We have specified in both lines that we refer to Africa only.

Reviewer 1 Point 27 — Page 13, lines 3 and 4: Move figures 9 and 10 in the main text.

Reply: We welcome this suggestion and move the figures accordingly.

Reviewer 1 Point 28 — Page 14, line 10: What you mean by "measurement error of unknown distribution"? Could you please clarify.

Reply: When harmonizing grids with different spatial resolutions, the interpolation process itself can introduce errors. Such errors can arise because data values are estimated in locations that were not originally measured in the raw data, therefore we have no means of knowing the type of distribution of these errors. We have clarified as follows: "First, results are highly dependent on input data availability and quality. Using gridded data at different resolutions requires harmonization on a common global grid. As data values are estimated in locations that were not originally measured in the raw data, the interpolation process can introduce measurement errors of unknown distribution."

Reviewer 1 Point 29 — Page 14, line 39: The evolution of the approach by the consideration of an ensemble of the models is a quite good perspective of work to improve the final estimate.

Reply: We thank the Reviewer for this comment.

Moreover, we have integrated the acknowledgement of financial support as follows:

"This work was supported by the World Bank. This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement No 101069880 - AdJUST, and from the European Union - Next Generation EU, in the framework of the project GRINS - Growing Resilient, INclusive and Sustainable project (GRINS PE00000018 – CUP C93C22005270001)."

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