RESPONSES TO REVIEWERS

Response to Reviewer #1

We would like to thank the reviewer for the positive feedback. Below, we reply to the comments from the reviewer:

Comment 1: Section 3.1: Further discussions are desirable describing (and attributing) the model biases in terms of setup, errors in model meteorology, and processes included/excluded in different simulations.
Response: We have now included more detailed model descriptions regarding the chemistry and aerosol modules in the Materials and Methods section (Lines 152-170), along with an updated and extended Table 1 providing references to chemical mechanisms used in the models. In addition, we have also provided more discussion on possible model biases in section 3.1 (Lines 280-292), including some discussion on the meteorological biases. However, this paper does not aim to make a full evaluation and error attribution of the models. It does however build on Solazzo et al. (2017) in the same special issue that makes a deep evaluation of the models.

Comment: Page 10, l. 393-395: Why does SO2 enhancement in case of reduced domestic emissions in North America are pronounced in a small belt over Europe? Is it possible to substantiate the statements with model simulated OH fields?
Response: We thank the reviewer for the careful review and we have identified a problem during the plotting. We have now corrected this plot. However, there is still a slight increase of SO2 over the Alps that is simulated by the majority of the models. The AQMEII database unfortunately does not include OH fields so we cannot further evaluate this increase in this paper.

Comment: Page 4, l.162 – “where embedded” to “were embedded”
Response: Corrected (Line 184).

Comment: Page 7, l.276: “SO2:.:.by 35% 5”. Pl. check this sentence.
Response: Corrected (Line 305).

Comment: Page 7, l.279 – “effect” to “affects”
Response: Corrected (Line 308).
Response to Reviewer #2

We would like to thank the reviewer for the careful read of the manuscript positive feedback. Below, we reply to the comments from the reviewer:

Comment: - Table 1. You are ordering runs according to groups, not according to models. After a more thorough look many of the groups use the same model, sometimes even on the same resolution. What is the use of an ensemble of groups running the same model? Ideally this should give exactly the same results unless someone makes an error or the model version is different.

- Linked to the previous bullet: you end the conclusions with raising the issue of the impact of different model parameterization. However, you do not include such information. You should include a description of important model facts and add a discussion on these linking them to your results.

- Based on this information, perhaps some model runs should be removed from the ensemble (too many of the same model? Too simple parameterizations for some species?).

Response: We have now updated and extended Table 1, providing more information on the mode specific spatial and vertical resolutions as well different chemistry and aerosol mechanisms. We have also added more information on the differences between the versions of the same models (e.g. CMAQ and WRF-Chem) by each group (Lines 152-170). The models or the versions of the same models differ from each and therefore, we think model removal is not necessary.

Comment: In the abstract you describe daily maximum 8h mean ozone. Is this what you show and evaluate in the tables and figures? Or is it monthly/annual means? You need to clarify this (in all figures/table legends as well as in the methods) or (and) only include results in the abstract which you are actually showing as results in figures/tables.

Response: The model evaluation is based on monthly means, as described in the beginning of section 3.1. and the cation of Table 3 and the captions of Fig. 1 and 2. We have calculated the impact on daily maximum 8hr O3 in order to show a policy impact of these reductions.

Comment: The RERER value analysis is interesting. It would be of great value if you describe the ozone RERER value based on monthly values (daily max 8h mean or mean), since ozone formation capacity/local contribution is seasonally dependent. Perhaps you can come up with a smart way of illustrating these rather than just adding more table values.

- I don’t see the point of showing figures 11 (GLONAM-BASENAM) to 14 (EUREURBASEEUEUR). I would much rather see geographically resolved RERER values as a complement to the other figures.

Response: We thank the reviewer for his interest in the RERER analyses and we agree that is can be more emphasized in the paper. Therefore, we have now, as suggested by the reviewer, produced spatial distribution maps for O3 and PM2.5 (Fig. 17) as well as monthly time series of the response for these pollutants (Fig 18) and added discussions on these results (Lines 578-606). On the other hand, we would like to keep Figs 11 and 14 to be consistent in the flow.
Comment: Line 221-224. The method of first taking difference then calculating mean is only valid if you are working with means. How do you treat the daily maximum 8h mean? Is the method valid for this metric (if that is what you are showing in the figures for ozone).

Response: The figures and tables only show the differences in monthly and annual means of the pollutants. Daily maximum 8hr ozone is only presented in the text as an additional information. As written in the text, we look at the difference in the mean of daily maximum 8hr ozone, but these are not presented in tables or figures.

Comment: Table 1. The number of simulations (scenarios) is different when comparing the table to the method text (for Europe). An x is missing in the table (grey area for north America-region).

Response: We thank the reviewer for the careful read. We have now corrected these.

Comment: Table 3. You state unit: % for NMB and NMGE, but the values in the table are clearly without unit. You should not have different units for North America and Europe (for RMSE in this case).

Response: We agree with the reviewer and we have now corrected the units in Table 3 caption.

Comment: You have a supplement but you do not refer to it in your manuscript.

Response: We thank the reviewer for pointing out this missing part. We have now referred to the supplement in various parts of the manuscript (Lines 194, 418-419, 520-521).

Comment: The figure legend of S3 is incorrect.

Response: We have now corrected the figure caption.

Comment: Section 2, first paragraph is messy and repetitive.

Response: We have now reorganized this paragraph (Lines 171-190).
Influence of anthropogenic emissions and boundary conditions on multi-model simulations of major air pollutants over Europe and North America in the framework of AQMEII3

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Abstract

In the framework of the third phase of the Air Quality Model Evaluation International Initiative (AQMEII3), and as contribution to the second phase of the Hemispheric Transport of Air Pollution (HTAP2) activities for Europe and North America, the impacts of a 20% decrease of global and regional anthropogenic emissions on surface air pollutant levels in 2010 are simulated by an international community of regional scale air quality modeling.
groups, using different state-of-the-art chemistry and transport models (CTM). The emission
perturbations at the global level, as well as over the HTAP2-defined regions of Europe, North
America and East Asia are first simulated by the global Composition Integrated Forecasting
System (C-IFS) model from European Centre for Medium-Range Weather Forecasts
(ECMWF), which provides boundary conditions to the various regional CTMs participating
in AQMEII3. On top of the perturbed boundary conditions, the regional CTMs used the same
set of perturbed emissions within the regional domain for the different perturbation scenarios
that introduce a 20% reduction of anthropogenic emissions globally as well as over the
HTAP2-defined regions of Europe, North America and East Asia.

Results show that the largest impacts over both domains are simulated in response to the
global emission perturbation, mainly due to the impact of domestic emissions reductions. The
responses of NO$_2$, SO$_2$ and PM concentrations to a 20% percent anthropogenic emission
reductions are almost linear (~20% decrease) within the global perturbation scenario with
however, large differences in the geographical distribution of the effect. NO$_2$, CO and SO$_2$
levels are strongly affected over the emission hot spots. O$_3$ levels generally decrease in all
scenarios by up to ~1% over Europe, with increases over the hot spot regions, in particular in
the Benelux region, by an increase up to ~6% due to the reduced effect of NOx-titration. O$_3$
daily maximum of 8-hour running average decreases in all scenarios over Europe, by up to
~1%. Over the North American domain, the central-to-eastern part and the western coast of
the U.S experience the largest response to emission perturbations. Similar but slightly smaller
responses are found when domestic emissions are reduced. The impact of inter-continental
transport is relatively small overall both domains, however, still noticeable particularly close to
the boundaries. The impact is noticeable up to a few percent, for the western parts of the
North American domain in response to the emission reductions over East Asia. O$_3$ daily
maximum of 8-hour running average decreases in all scenarios over North Europe by up to
~5%. Much larger reductions are calculated over North America compared to Europe.

In addition, values of the Response to Extra-Regional Emission Reductions (RERER) metric
have been calculated in order to quantify the differences in the strengths of non-local source
ccontributions to different species among the different models. We found large RERER values
for O$_3$ (~0.8) over both Europe and North America, indicating a large contribution from non-
local sources, while for other pollutants including particles, low RERER values reflect a
predominant control by local sources. A distinct seasonal variation in the local vs. non-local
contributions has been found for both O$_3$ and PM$_{2.5}$, particularly reflecting the spring-time
long-range transport to both continents.

1. Introduction

Regional air quality modeling has considerably developed during recent decades, driven by
increased concern regarding the impact of air pollution on human health and ecosystems.
Numerous air quality models have been developed by research groups worldwide and are
being widely used for developing and testing emission control policies. Regional atmospheric
chemistry and transport models (CTMs) are widely used to assess the past, present and future
levels of air pollutants from continental to regional scales. There are different sources of
uncertainties in models such as emissions, meteorology, boundary conditions and chemical
schemes that should be taken into account when analyzing results. These uncertainties
become more critical when these models are used for regulatory applications such as impacts
of emission reductions. Multi-model ensembles can help in reducing this uncertainty and
provide a better estimate of impacts under different scenarios (Solazzo et al., 2013; Galmarini
et al., 2013; Kioutsoukis et al., 2017).

Numerous observational and modeling studies show that long-range transport of pollutants
degrade air quality over remote continents (e.g., Wilkening et al., 2000; Holloway et al.,
2003; Akimoto, 2003; Fiore et al., 2009). Although the influence of foreign emissions on
continental scales is seen most frequently in the free troposphere, surface levels can also be
affected, in particular over locations that generally receive clean air masses (e.g. Li et al.,
2002). For example, dust storms and biomass burning can influence the tropospheric
composition on a hemispheric scale (e.g., Husar et al., 2001; Jaffe et al., 2004). Reducing air
pollution levels in surface air would improve public health as exposure to these atmospheric
constituents aggravates respiratory illness and leads to premature mortality (World Health
Organization, 2013; Im et al., 2017; Liang et al., 2017). However, attributing pollution to
specific source regions is complicated due to the different processes influencing
intercontinental transport and by a large hemispheric background and the dominance of local
emissions in contributing to high levels of particular pollutants, such as ozone (O₃) (e.g. Fiore
et al., 2009). Given these difficulties, estimates of source-receptor relationships rely heavily
on models.

Sjöström et al. (2016), using ten models participating in the second Hemispheric Transport of
Air Pollution (HTAP2) activity, showed that a 20% reduction of global anthropogenic
emissions, leads to significant changes regionally. They found that for North America (NA),
black carbon emissions controls in East Asia are more important than domestic mitigation. In
the framework of the HTAP2 activity, UN (2007) showed that a 20% reduction of North
American NOx emissions leads to a 0.22 ppb decrease in O₃ levels over Europe (EU), while a
20% decrease in East Asian NOx emissions leads to a decrease of North American surface O₃
levels by 0.12 ppb. The impacts of these emissions changes on the O₃ levels in the source
regions are much higher. The impact of lateral boundary conditions (LBC) on concentration
fields simulated by regional-scale air quality models can also be quite significant (Jimenez et
al., 2007; Mathur, 2008; Rudich et al., 2008; Song et al., 2008; Anderrson et al., 2015;
Giordano et al., 2015, Hogrefe et al., 2017; Solazzo et al., 2017a). Recently, Giordano et al.
(2015) showed that the regional models can be very sensitive to the boundary conditions
provided by the global models. Tang et al. (2007) showed that the simulated surface levels
over polluted areas are usually not as sensitive to the variation of LBCs, but are more
sensitive to the magnitude of their background concentrations. Jonson et al. (2017), in the
framework of the HTAP2 activity, showed that for ozone the contributions from the rest of
the world is larger than the effects from European emissions alone, with the largest
contributions from North America and East Asia. The majority of these studies that address
impact of emissions on regional and inter-continental transport employ global models on
coarse spatial resolution or focus on just a few species, such as O₃ or carbon monoxide (CO).
On the other hand, studies using regional chemistry and transport models at finer spatial resolutions mostly focus on sub-regional scales (e.g. Im and Kanakidou, 2012; Huszar et al., 2016). Therefore, studies addressing multi-pollutant, source-receptor relationships on inter-continental and regional scales can provide valuable information on the impact of domestic and foreign emissions on regional air pollution levels. Multi-model ensembles operating on fine spatial resolutions can increase accuracy and provide an estimate of uncertainty.

The Air Quality Model Evaluation International Initiative (AQMEII), coordinated jointly by European Commission, Joint Research Centre (EC-JRC) and the U.S. Environmental Protection Agency (EPA) has brought together regional chemistry and transport modelling groups from Europe and North America since 2008 (Rao et al., 2012; Solazzo et al., 2012a,b; Im et al., 2015 a,b). AQMEII is now running its third phase as a regional sub-project of the larger Hemispheric Transport of Air Pollution (HTAP), which in turn is a taskforce of Long Range Transport of Air Pollution program (LTRAP) of United Nations Economic Commission for Europe (UNECE) (Galmarini et al., 2017). The aim of the study is to assess the impact of global and HTAP2-defined regional anthropogenic emission reductions of 20% in Europe, North America and East Asia on major air pollutant levels over Europe and North America using a multi-model ensemble approach. The study will also investigate the local vs. non-local contributions to different air pollutant levels, adopting the Response to Extra-Regional Emission Reductions (RERER) metric developed by the HTAP2 community (Galmarini et al., 2017).

2. Materials and Methods

In the framework of the AQMEII3 project, fourteen groups contributed to the simulation of the air pollution levels for 2010 in Europe (EU) and three groups for North America (NA) in the year 2010 (Table 1 and Solazzo et al., 2017b). As seen in Table 1, different groups used same CTM models, such as the CMAQ and WRF-Chem model. The main differences among these models reside in the number of vertical levels, horizontal spacing, biogenic emissions, gas/aerosol modules in the models and the model releases (Table 1). For example, regarding groups that used the CMAQ model, UK1, DE1 and US3 calculated biogenic emissions using the BEIS (Biogenic Emission Inventory System version 3) model, while TR1, UK1 and UK2 calculated biogenic emissions through the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2012). Moreover, DE1 does not include the dust module, while the other CMAQ instances use the inline calculation (Appel et al., 2013), and TR1 uses the dust calculation previously calculated for AQMEII phase 2. Finally, all runs were carried out using CMAQ version 5.0.2, except for TR1, which is based on the 4.7.1 version. The gas-phase mechanisms and the aerosol models used by each group are also presented in Table 1. IT1 used the WRF-Chem model version 3.6, with a new chemistry that includes a better representation of the secondary organic aerosol mass in the simulation of direct and indirect aerosol effects (Tuccella et al., 2015). In addition, only direct effects were included in the IT1 simulation. ES1 model also used WRF-Chem, with different gas phase chemistry. More details of the model system are provided in the supplementary material in Im et al. (2018).
The emission inventories that are used in the second phase of AQMEII for Europe and North America (Im et al., 2015a,b) and extensively described in Pouliot et al. (2015) are also used in AQMEII3. For the EU, the 2009 anthropogenic emission inventory from the Monitoring Atmospheric Composition & Climate (MACC) anthropogenic emissions was used. In regions not covered by the Monitoring Atmospheric Composition & Climate (MACC) inventory, such as North Africa, five modelling systems have complemented the standard inventory with the HTAPv2.2 datasets (Janssens-Maenhout et al., 2015). For the NA domain, the 2008 National Emissions Inventory was used as the basis for the 2010 emissions with 2010-specific adjustments for major point sources, mobile sources and wildfires (Pouliot et al., 2015). The emissions were then treated with the SMOKE emissions processing system (Mason et al., 2012). For both continents, the regional scale emission inventories were embedded in the global scale inventory (Janssens-Maenhout et al., 2015) to guarantee coherence and harmonization of the information used by the regional and global scale modelling communities (Galmarini et al., 2017). The majority of the European groups used MACC emissions over Europe, while FI1 and FRES1 supplemented the MACC emissions with HTAP emissions over North Africa (Table 1). For NA, the temporal and vertical allocation of emissions vary between the groups that used the "SMOKE" files (DE1, US1, US3) and the gridded HTAP files (DK1), however the annual total mass are exactly the same. In order to guarantee consistency between the groups using the regional scale MACC or SMOKE emissions, and the groups using the HTAPv2.2 emissions, the regional scale emission inventories were embedded in the HTAPv2.2 inventory (Janssens-Maenhout et al., 2015; ) to Galmarini et al., 2017). Overall, there was a high level of harmonization of emission inputs even if there were some differences in how they were adapted by each modeling group for their system. Chemical boundary conditions for both domains were provided by the European Center for Medium Range Weather Forecasts (ECMWF) Composition – Integrated Forecast System (C-IFS) model (Flemming et al., 2015)

2.1. Emission perturbations

The perturbation scenarios feature a reduction of 20% of the anthropogenic emissions globally and in HTAP-defined regions of Europe, North America and East Asia (Table 2 and Fig. S1). The choice of 20% was motivated by the consideration that the perturbation would be large enough to produce a sizeable impact (i.e. more than numerical noise) even at long distances while small enough to be in the near-linear atmospheric chemistry regime (Galmarini et al., 2017). The emission reductions are implemented in both the global C-IFS model that provides the boundary conditions to the participating regional models, as well as in the regional models. The regional models use the corresponding set of boundary conditions extracted from the C-IFS model. Among the fourteen groups that participated to the AQMEII3 base case simulations, twelve groups from Europe and two groups from North America simulated at least one of the three emission perturbation scenarios, shown in Table 1. Two of the European groups (DE1 and DK1) also simulated the base and the three perturbation scenarios for the North American domain.

- The global perturbation scenario (GLO) reduces the global anthropogenic emissions by 20%. This change has been implemented in the C-IFS global model that provides
the boundary conditions to the regional models participating in the AQMEII ensemble. Therefore, the GLO scenario introduces a change in the boundary conditions as well as a 20% decrease in the anthropogenic emissions used by the regional models. Nine groups over the EU domain and four groups over the NA domain have simulated the GLO scenario.

- The North American perturbation scenario (NAM) reduces the anthropogenic emissions in North America by 20%. This change has been implemented in the C-IFS global model that provides the boundary conditions to the regional models used in the AQMEII ensemble. Therefore, the NAM scenario introduces a change in the boundary conditions while anthropogenic emissions remain unchanged for Europe, showing the impact of long-range transport of North American pollutants to Europe while for North America, the scenario introduces a 20% reduction of anthropogenic emissions in the HTAP-defined North American region, showing the contribution from the domestic anthropogenic emissions. Seven groups over the EU domain and three groups over the NA domain have simulated the NAM scenario.

- The European perturbation scenario (EUR) reduces the anthropogenic emissions in the HTAP-defined Europe domain by 20%. The EUR scenario introduces a change in the anthropogenic emissions over the EUR region in the CTMs, showing the contribution from the domestic anthropogenic emissions. Six groups have simulated the EUR scenario over the EU domain.

- The East Asian perturbation scenario (EAS) reduces the anthropogenic emissions in East Asia by 20%. Similar to the NAM scenario for the EU domain, the EAS scenario introduces a change in the boundary conditions while anthropogenic emissions remain unchanged in the regional models, showing the impact of long-range transport from East Asia on the NA concentrations. Four groups have simulated the EAS scenario over the NA domain.

In AQMEII, all participating groups were required to upload modelled hourly surface concentrations to the ENSEMBLE system at EC-JRC, at specified monitoring stations in EU and NA, as well as surface gridded data (Galmarini et al., 2012; Im et al., 2015a, b; Solazzo et al., 2017b). This study investigates the impacts of emission perturbations and boundary conditions on O3, NO2, CO, SO2, PM10 and PM2.5 levels over Europe and North America. Differences between each perturbation scenario and the base case (C-IFS global and regional models run with baseline emissions) are calculated from the gridded hourly pollutant fields, which are then monthly and annually averaged in order to estimate the impact of the perturbation of the corresponding emission or boundary condition.

To estimate the contribution of foreign emission perturbations relative to the GLO perturbation, we have also calculated the RERER metric (Galmarini et al., 2017; Huang et al., 2017; Jason et al., 2017). For Europe, RERER is calculated using the differences between the GLO vs BASE as well as the differences between EUR vs. BASE simulations for Europe (Eq. 1) while for North America; RERER is calculated using the differences between the GLO vs BASE and NAM vs. BASE simulations (Eq. 2).
\[
RERER_{EUR} = \frac{R_{GLO} - R_{EUR}}{R_{GLO}} \quad \text{Eq. 1}
\]

\[
RERER_{NAM} = \frac{R_{GLO} - R_{NAM}}{R_{GLO}} \quad \text{Eq. 2}
\]

where \( R_{GLO} \) is the response of the concentration of a given species to global emission reduction, \( R_{EUR} \) is the response of a concentration of a species to the EUR perturbation for the European domain, and \( R_{NAM} \) is the response of a concentration of a species to the NAM perturbation for the North American domain. Therefore, a subset of modelling groups that have conducted the three simulations (BASE, GLO and EUR/NAM for Europe and North America, respectively) have been used in the metric calculations (see Table 1). The higher the local response is, the smaller the RERER metric is. The RERER value can exceed the value 1 when emission reductions lead to increasing concentrations (e.g., \( O_3 \) titration by nitrogen monoxide, NO).

3. Results

3.1. Model Evaluation

The base case simulation of each model has been evaluated on a monthly-mean basis using available surface observations from Europe and North America. The observational data used in this study are the same as the dataset used in the second phase of AQMEII (Im et al., 2015a,b). The data were provided from the surface air quality monitoring stations operating in EU and NA. In EU, surface data were provided by the European Monitoring and Evaluation Programme (EMEP, 2003; http://www.emep.int/) and the European Air Quality Database (AirBase; http://acm.eionet.europa.eu/databases/airbase/). NA observational data were obtained from the NAtChem (Canadian National Atmospheric Chemistry) database and from the Analysis Facility operated by Environment Canada (http://www.ec.gc.ca/natchem/).

The model evaluation results for each model are presented in Fig. 1 and 2, and in Table 3, along with the results for the multi model (MM) mean and median values. The results show that the monthly variations of gaseous pollutants are well captured by all models with correlation coefficients (\( r \)) generally higher than 0.70. The biases in simulated \( O_3 \) levels are generally less than 10% with a few exceptions of up to -35%. The temporal variations of \( NO_2 \) levels are also well simulated (\( r>0.7 \)), but exhibit much higher biases, with underestimations up to 75%. \( CO \) levels are underestimated by up to 45% while a majority of the models underestimated \( SO_2 \) levels by up to 68%. Few models overestimated \( SO_2 \) by up to 49%. \( PM_{10} \) and \( PM_{2.5} \) levels are underestimated by 20% to 70%. Slightly higher biases are calculated for the \( PM_{10} \) levels.

The model biases can be attributed to meteorology, in particular wind speed and planetary boundary layer (PBL) height, as well as the aerosol mechanisms used in different models that can underestimate either the inorganic aerosols (e.g., IT2) or the secondary organic aerosols (e.g., DK1), leading to underestimations in simulated PM mass. As discussed in Solazzo et al. (2017), EU3 region that covers the central Europe including the Alps has the largest errors in terms of wind speed, mainly attributed to the diurnal component of the error, with some
models having also large errors in the synoptic component. This region also represents the
lowest correlation coefficients for all models. They further conclude that emissions and their
vertical distribution are the main source of model biases; in particular for the primary species
such as CO and PM. Regarding O3, they found that the models have highest biases in the
large scale synoptic component while the diurnal variations are well-captured in general. A
more comprehensive evaluation of the models is presented in Solazzo et al. (2017b),
Galmarini et al. (2017) and Im et al. (2018).

C-IFS base case results have also been evaluated along with the regional CTMs, as presented
in Fig. 1 and 2 and in Table 3. The seasonal variations for O3, NO2, CO and SO2 are well
captured with high correlation values of ~0.9. PM10 and PM2.5 showed a different seasonal
cycle than the observation by not reproducing the wintertime maxima ($r=-0.7$). C-IFS model
underestimates O3 and CO by ~20% over Europe while NO2 is slightly overestimated
($NMB=7\%$). SO2 is overestimated by ~10% over Europe, while PM10 and PM2.5 levels are
largely underestimated by ~60%, which can be attributed to the lack of secondary aerosol
mechanism in the bulk C-IFS model. Over the North American domain, C-IFS well captures
the seasonal variations of O3, NO2 and CO with correlation coefficients larger than 0.7, while
the seasonal variation of SO2 is not captured by the model ($r=0.04$). The seasonal variations
of PM10 and PM2.5 are also poorly captured ($r=0.2$). North American O3 levels are slightly
underestimated ($NMB=-10\%$), while NO2 and CO are overestimated by ~40% and 20%,
respectively. SO2 is overestimated by 35% while PM10 is largely underestimated by ~80%
and PM2.5 by ~40%. Over both Europe and North America, the wintertime PM levels are
underestimated due to lack of secondary aerosols while the spring summer peaks are
attributed to long range transport of desert dust from the Sahara, which affects mainly
the South East of North America.

3.2. Perturbation Analyses

The annual mean relative differences of each perturbation scenario from the base case
scenario, averaged over all stations, are provided in Table 4 (EU) and Table 5 (NA) for each
modeling group, along with the results for the MM ensemble mean and median. The base
case monthly mean time series for the participating groups are provided in Fig.1 and Fig. 2
for each pollutant, while Fig.3 and Fig. 4 shows the annual mean spatial distribution of the
pollutants from the MM ensemble mean calculations over Europe and North America,
respectively. As seen in the time series figures, there is a large spread among different
groups, owing to the different models used and the different sets of anthropogenic emissions
(Table 1). However, the temporal variation is consistent among all models, in particular for
the gaseous species.

3.2.1. Impact of the global emission reduction scenario (GLO)

3.2.1.1. Europe

The monthly time series of the differences between the GLO and the BASE simulations for
each pollutant are presented in Fig. 5. The annual differences are reported in Table 4.
Regarding the primary gaseous pollutants, all models simulate the smallest differences during
the summer months while the differences are largest in winter. For O₃, the simulated differences are positive in winter and negative in summer for all models except for DE1 that simulated a decrease in all months. Results suggest that wintertime O₃ over Europe is mainly controlled by anthropogenic emissions. For the other pollutants, results suggest that their levels are mainly controlled by anthropogenic emission throughout the year. The annual difference is smallest for O₃, with a reduction of -0.34±1.23 ppb (-1.04±4.00%). The annual mean value of the O₃ daily maximum of 8-hour running average decreases by -0.53±1.50 ppb (-1.62±3.99%). NO₂ levels decreased by 0.97±0.45 ppb (19.34±1.59%) over Europe while CO levels decreased by 17.35±4.03 ppb (11.22±1.17%), SO₂ levels by 0.18±0.05 ppb (20.87±0.93%), PM₁₀ by 2.38±0.68 μg m⁻³ (15.84±2.12%) and PM₁.₅ by 2.02±0.52 μg m⁻³ (18.30±1.75%). Vivanco et al. (2017) found similar reductions regarding the deposition of sulfur and nitrogen species over Europe. Almost all models simulate an overall decrease of annual mean O₃ levels over EU (-0.94% to -4.65%), with the exception of TR1 that simulated an increase of 9.31%. Regarding other pollutants, all models simulate a decrease during the simulation period. In general, DE1 and TR1 model groups stand out for introducing the smallest and largest differences, particularly for O₃, NO₂, and PM.

The geographical distribution of the change in annual mean concentrations in the GLO scenario as simulated by the MM mean is presented in Fig. 6. Regarding O₃, most of Europe is characterized by decreased concentrations (Fig.6a). Over central Europe, where most of the primary emissions are located (e.g. NOₓ), O₃ levels slightly increase by ~2%. Emission hotspots, in particular the Benelux area stands out with largest increases (~6%) due to decreased NOₓ-titration effect, which can also be seen in Fig. 6b. In addition, O₃ levels over the northern parts of Germany and France, and southern UK are increasing in response to emission reductions. There is also a clear decrease in CO levels (Fig.6c), in particular over central Europe by up to ~16%. All primary species decrease over the whole domain, especially over the industrial hot spots such as in Poland, Po Valley and the Benelux area (Fig.6d). PM levels decrease throughout the domain by up to ~20% (Fig.6e and f).

3.2.1.2. North America

The seasonal variation of the impact of 20%-decreased global emissions on the North American pollutant levels are presented in Fig.7. All models simulated a small decrease of 3% to 5% (Table 5) in O₃ levels with the largest differences in spring to summer (Fig.7a). The mean response to the emission perturbation is estimated to be -1.39 ± 0.27 ppb (-3.52 ± 0.80%). The annual mean value of the O₃ daily maximum of 8-hour running average decreases by -1.93±0.14 ppb (4.51±0.45%). All models simulated a largest NO₂ response in winter. Most models simulated a decrease of NO₂ levels while DK1 estimated an increase (Fig.7b). As shown is Table 5, the models simulated a NO₂ response of ~0.4 – 1.2 ppb (-17.8 ± 0.78%). Regarding CO, all models simulated very clear seasonal profile of the response to emission reductions, with maximum change in late winter/early spring and the minimum change in summer. Most models simulated a change around -15 to -25 ppb (-11%); with the exception of the DE1 model simulating a decrease of ~9 ppb (~7.9%). The MM mean response is calculated to be 19.2 ± 6.9 ppb (-11 ± 2.3%). The impact of the emission reduction on SO₂ levels was calculated to be -0.25 ppb to -0.48 ppb (-20.3 ± 0.2%).
The response of PM_{10} levels to the global emission reduction was calculated to be -2.4 ± 1.8 µg m^{-3} (-32.1 ± 26.6%) (Table 5). The largest relative change was calculated for DE1 (-63%). DK1 has a flat response around -1 µg m^{-3}, while DE1, which is overlapped with the Median line, and US3 have maximum responses in early spring and mid-autumn, while they simulate a minimum response in winter and late spring. Regarding PM_{2.5}, the multi-model mean response was calculated to be -1.5 ± 0.9 µg m^{-3} (-17.2 ± 1.8%). DK1 (overlapped with the Median) and US3 simulated the minimum response in May (Fig.7f), while US3 has a slightly higher second minimum in September. This minimum is also simulated by DE1 as the minimum response. DE1 simulates the lowest response among the three models.

The spatial distributions of response of different pollutants to the GLO scenario are presented in Fig.8. O_{3} levels are reduced over most of the domain (Fig.8a), with slight increases over the emission hotspots due to reduced effect of NOx-titration, as seen in Fig.8b, as well as decreased CO levels over the whole domain (Fig.8c). SO_{2} levels are also decreased throughout the domain (Fig.8d), with the largest reductions over the Atlantic (attributable to reduction in shipping emissions). The western part of the continent is characterized by the lowest reductions. PM levels are reduced throughout the domain by up to 25% (Fig.8e and f), with the largest reductions over the eastern and central parts of the domain. A large decrease, more pronounced in the PM_{2.5} response, can also be seen over California in the western coastal United States.

3.2.2. Impact of the North American emission reduction scenario (NAM)

3.2.2.1. Europe

NA emission reductions account for a reduction of European O_{3} levels of -0.22±0.07 ppb (-0.75±0.14%), with all models simulating a decrease of -0.51% to 0.86%, except for the ES1 model that simulated an increase of 1.31% (Table 4). This decrease is in agreement with previous studies, such as the HTAP2 study (UN, 2017) that calculated an O_{3} reduction over Europe of 0.22 ppb in response to a 20% decrease in the North American NOx emissions, and Fiore et al. (2009) that simulated a MM mean response of -0.4 ppb in response to a 20% reduction of anthropogenic emissions in North America. NO_{2} levels increase slightly by 0.16±0.01%. The annual mean value of the O_{3} daily maximum of 8-hour running average decreases by -0.15±0.27 ppb (-0.45±0.77%). CO levels also decreased over the EU domain by -1.39±0.27 ppb (-0.96±0.22%), much higher than ~0.1 ppb calculated by Fiore et al. (2009). PM_{10} and PM_{2.5} levels also decreased slightly by -0.02±0.03 µg m^{-3} (-0.21±0.7%) and -0.02±0.02 µg m^{-3} (-0.18±0.25%), respectively. The models had different SO_{2} responses to the NA emissions. Overall, DE1, ES1 and FRES1 simulated almost no change in the surface SO_{2} levels while DK1, ES1 and TR1 simulated an increase (0.10%, 5.75% and 0.01%, respectively) and FI1 and UK1 simulated a decrease (-0.02% and -0.03%, respectively).

Different responses can be due to different model setups including aqueous chemistry, vertical resolutions and aerosol modules (Solazzo et al., 2017).

All models were consistent in simulating the largest impact on O_{3} during spring and a second lower peak in autumn (Fig.9a). Surface mean NO_{2} concentrations (Fig.9b) increased in most
models except for FRES1 that simulated a small decrease except for winter. FI1 also
simulated a decrease during the winter period extending to the transition periods. All models,
except for ES1, simulated a similar response of CO concentrations to perturbation to NA
emissions, with a distinct seasonality (Fig. 9c). The SO2 response in models is also consistent
except for the winter period where there is a large spread in magnitude and the sign of the
response (Fig. 9d).

O3 levels decreased slightly over the entire European domain by up to 3% (Fig. 10a). The
largest impact is simulated over the western boundary and gradually decreases eastwards.
The response of O3 levels to NAM emissions is more evident during spring where there is a
clear transport from Atlantic to the western/northwestern parts of Europe such as the U.K,
northern France and Scandinavia (Fig. S2a). The transport of Atlantic air masses is also
shown for the springtime CO levels over Europe (Fig. S2ba). The ensemble mean simulates a
slight increase of up to 3% in NO2 levels over Europe (Fig. 10b). Along with the O3 levels,
CO levels show the largest decrease over northwestern Europe by up to ~2%. SO2 levels
increased over the whole domain, in particular over Eastern Europe and the Alpine region
(Fig. 10d), due to a decrease in the oxidative capacity of the atmosphere (see Fig. 10a for O3),
leading to a decrease in the SO2 to SO4 conversion. This results in an increase of the SO2
levels and a decrease in the PM2.5 levels (Fig. 10e and f).

3.2.2.2. North America

The response of North American pollutant levels to a 20% reduction of North American
anthropogenic emissions (implemented in both C-IFS and the regional CTMs) are presented
in Table 5. The NAM scenario led to a decrease of annual mean O3 levels over North
America by -0.36 ppb (US3) to -0.92 ppb (DE1), with MM ensemble mean calculated to be -
0.65±0.28 ppb (-1.45±0.88%), in agreement with Fiore et al. (2009) that calculated a decrease
of ~1 ppb. The annual mean value of the O3 daily maximum of 8-hour running average
decreases by -1.11±0.11 ppb (-2.60±0.36%), very similar to the change over Europe.
Consequently, the largest change in NO2 levels were simulated by US3 (-1.17 ppb) and
smallest by DE1 (-0.36 ppb). The MM mean response of NO2 is calculated to be -0.71±0.41
ppb (-17.24±0.58%). Similar to NO2, the largest response in CO levels were simulated by
US3 (-19.87 ppb) and the smallest by DE1 (-3.84 ppb), leading to a MM mean response of -
12.35±8.06 ppb (-7.01±3.60%). As seen in Table 5, DE1 simulated a much lower absolute
and relative change in CO response compared to DK1 and US3. SO2 levels decreased by -
0.32 ppb to -0.48 ppb, leading to a MM mean response of -0.37±0.09 ppb (-20±0.12%). PM10
levels decreased -1.78±2.08 μgm⁻³ (-15.78±3.26%). As seen in Table 5, DK1, simulated a
very low response to the NAM scenario, by ~0.60 μgm⁻³, compared to the DE1 and the US3
groups that simulated a PM10 response of ~2.02 μgm⁻³ and ~4.19 μgm⁻³, respectively.
However, the relative responses are not very different between the different groups (~16%).

The response of O3 to the NAM scenario is largest in summer (Fig. 11a): June for DK1 and
US3 and August for DE1. The O3 response clearly shows a difference from the GLO
response in spring, suggesting the impact of long-range transport in spring that does not
appear in the perturbation of the local emissions only. The largest NO2 response (Fig. 11b) is
simulated by US3, similar to the response to the GLO scenario. The response of CO to the
reductions in local emissions (Fig.11c) is different from the response to the global reduction,
where DK1 and US3 has the minimum response in spring and DE1 has the minimum
response in autumn. The response of SO2 and PM to GLO and NAM are similar, suggesting
the main drivers of SO2 and PM levels are local emissions.

Annual mean O3 levels show large reductions (~20%) over the eastern parts of the domain,
while there are slight increases or less pronounced decreases over the western parts of the
domain (Fig.12a), associated with larger NOx reductions (Fig.12b). CO and SO2 levels are
mostly reduced over the central to eastern parts of the domain (Fig.12c and d, respectively),
with shipping impacts over the Atlantic being more pronounced on SO2 levels. The western
parts of the U.S. experiences smaller SO2 reductions (~5-10%) and slight increases over the
southwestern U.S. The response of PM to the NAM scenario (Fig.12e and f) is very similar to
the response to the GLO scenario (Fig.8e and f).

3.2.3. Impact of the European emission reduction scenario (EUR)
O3 levels increase slightly by 0.01±0.40 ppb (0.25±1.35%) in response to the 20% reduction
of the anthropogenic emissions from Europe (Table 4). This response is much lower than
Fiore et al. (2009) that calculated a MM mean response of 0.8 ppb. However, as seen in
Fig.13a, the positive mean response together with the large standard deviation is due to the
DE1 model that simulated a decrease (-2.33%), while other groups simulated an increase
(0.39% to 1.72%). There is a distinct seasonality in the response with winter levels increasing
with reduced emissions and summer levels decreasing, following the emission temporal
variability. The annual mean value of the O3 daily maximum of 8-hour running average
decreases by -0.21±0.10 ppb (-0.62 0.24%). NO2 concentrations decreased by -0.75±0.26
(17.68±0.90%), with a similar seasonal response of SO2 levels (-17.52±1.70%) and CO levels
(-6.26±1.07%), consistent with the findings of Vivanco et al. (2017). An opposite seasonal
variation is calculated for the O3 response (Fig. 13.b-d)., The DE1 model also stands out in
the NO2 response together with the FRES1 model in the magnitude of the response (Fig.13b).
PM10 and PM2.5 levels have similar responses to the emissions reduction (-14.43±2.84% and -
15.67±2.12%, respectively) with similar seasonality.
The MM mean geographical distribution of the O3 response is very similar with that of the
GLO perturbation (Fig.14a), with relatively smaller decreases by up to ~3%. O3 levels
increase over the central and in particular over northwestern Europe by up to ~6%. NO2
levels decrease uniformly over the entire domain by up to ~20% (Fig.14b). CO levels
decrease over the emission sources, mainly over central and Eastern Europe (Fig.14c). PM
levels also decrease over the entire domain, especially over central and Eastern Europe
(Fig.14e and f).

3.2.4. Impact of the East Asian emission reduction scenario (EAS)
As seen in Table 5, the impacts of East Asian emissions on North American O3 levels are
much lower than the impacts from the reductions in global and local emissions. The largest
impact is simulated by DE1 as -0.99 ppb (-0.35%), while other models give similar responses
The O₃ response as calculated by the MM mean ensemble is -0.25±0.07 ppb, in agreement with the HTAP2 findings and Fiore et al. (2009). The annual mean value of the O₃ daily maximum of 8-hour running average decreases by -0.28±0.07 ppb (-0.65±0.20%). NO₂ and SO₂ response to reductions in EAS emissions were simulated to be very small (-0.04±0.08% and 0.01±0.02%, respectively). The CO response to EAS was simulated to be -2.60 ppb (DE1) to -4.16 ppb (DK1), with the MM mean response of -3.37±0.68 ppb (DE1) to -4.16 ppb (DK1), with the MM mean response of -0.25±0.07 ppb (-0.60±0.20%). The annual mean value of the O₃ daily maximum of 8-hour running average decreases by -0.28±0.07 ppb (-0.65±0.20%). NO₂ and SO₂ response to reductions in EAS emissions were simulated to be very small (-0.04±0.08% and 0.01±0.02%, respectively). The CO response to EAS was simulated to be -2.60 ppb (DE1) to -4.16 ppb (DK1), with the MM mean response of -3.37±0.68 ppb (-2±0.29%). Regarding PM₁₀, DE1 simulated a very large response (~-0.56 µgm⁻³) compared to DK1 and US3 (~-0.05 µgm⁻³), leading to a MM mean response of -0.21±0.30 µgm⁻³ (-5.63±8.50%). However, the PM₂.₅ response was much lower (~-0.02±0.03 µgm⁻³; -0.20±0.35%), suggesting that the PM₂.₅ levels are largely driven by local emissions.

The O₃ response to EAS emission reductions was highest in spring and autumn, suggesting that long-range transport is important in these seasons (Fig.15a). The NO₂ response was negative, being maximum in winter and minimum in summer, except for DK1 showing an increase in NO₂ levels in all seasons (Fig.15b). The impact of EAS emissions on North American CO levels showed a distinct seasonality (Fig.15c), similar to the impact of the global emission reductions (Fig.5c), suggesting that regional CO levels over North America are driven by both local emissions and long-range transport. The response of SO₂ to East Asian emission reductions varied largely from model to model with US3 showing an overall reduction while DE1 and DK1 simulated increases in winter, spring, and autumn, and decreases in summer (Fig.15d). The PM₁₀ response simulated by DK1 (overlapped with the median) and US3 were simulated to be small, being largest in spring (Fig.15e). However, DE1 simulated a large and opposite response, with spring having the smallest response and winter with the largest response. DE1 also simulated a different PM₂.₅ response in terms of the sign of the change and thus, seasonality in response to DK1 and US3 (Fig.15f). Largest differences were simulated in spring, similar to PM₁₀ by DK1 and US3, while DE1 simulated the largest response in winter and summer and the spring response was minimum.

The impact of the East Asian emissions over the western parts of North America is clearly seen for all pollutants in Fig.16. The impacts are low for all pollutants, being up to 5%. The impacts are particularly pronounced for CO (Fig.16c), SO₂ (Fig.16d) and PM (Fig.16e and f). The largest O₃ response was simulated over the northwestern parts of North America (Fig.16a). The springtime transport of O₃ from East Asia is more evident compared to the annual average of the perturbation response (Fig. S3a), where the western NA O₃ levels decrease by up to ~1.5%. The springtime CO levels also decrease by up to 6% (Fig. S3b), showing the importance of long-range transport from East Asia.

3.2.5. RERER analyses

As discussed in Section 2, the RERER metric (Galmarini et al., 2017; Hang et al., 2017; Jason et al., 2017) is designed to quantify the relative impact of local vs. non-local emission sources on pollutant levels in the receptor regions EU and NA. Using gridded hourly pollutant concentrations from the base case, GLO and EUR simulations, the RERER metrics for the EU have been calculated using gridded annual mean pollutant concentrations from the BASE, GLO and EUR simulations for the annual mean concentrations response for
the individual groups as well as for the ensemble mean. For the NA domain. The RERER
metrics have been calculated using the annual mean concentrations from the base case BASE.
GLO and NAM simulations. Table 6 presents the RERER metric calculated for the European
domain. The table shows differences in the strengths of non-local source contributions to
different species among the different models. Regarding the RERER metric for O3 in Europe,
most values calculated are below one, except for the IT1 model, which shows a significant
increase of O3 levels in Europe in response to emission reductions compared with the other
models. A RERER value of 0.8-0.9 is calculated for the majority of models, implying the
dominance of non-local sources in Europe, except for the DE1 model, where the RERER
value is lower (~0.5), giving an equal contribution of local vs. non-local sources in Europe.
The MM mean RERER value for O3 is ~0.8, showing a much larger contribution of non-local
sources compared to local sources in Europe. This result is in agreement with, however
slightly smaller, Jonson et al. (2017) that calculated a MM mean RERER value of 0.89.
Regarding NO2, the RERER metrics (~0.4) show that NO2 is controlled by local sources. In
addition, the RERER metrics calculated for DE1 and FI1 are slightly negative, implying that
the signal is not sensitive to non-local emissions. RERER calculated for the ensemble mean
for NO2 (~0.2) also shows the high sensitivity of NO2 concentrations to local sources. The
RERER metric calculations for CO shows similar contributions from local vs. non-local
sources, with RERER values of 0.4-0.6, except for IT1. IT1 has a RERER metric value of
~0.9 suggesting a large contribution of non-local sources, leading to the higher sensitivity of
CO to non-local sources compared to other model groups. The RERER values calculated for
the ensemble mean (~0.6) shows a slightly larger contribution of non-local sources compared
to local sources. The MM mean RERER value of 0.55 for CO from this study is in very good
agreement with Jonson et al. (2017) that calculated a MM mean RERER of 0.51. RERER
metrics calculated for SO2 are also in the low range (0-0.4). While DE1 and FI1 show almost
no signal for the non-local contribution, DK1, IT1 and UK1 are in the higher end of the
range. The CO MM mean RERER value of ~0.3 shows that CO levels are largely controlled
by local emissions. Finally, the metrics calculated for PM10 and PM2.5 shows that local
sources are the main contributor to the PM levels in Europe (RERER = ~0 - 0.3), leading to
an ensemble mean contribution of local sources (RERER = ~0.2).

Regarding the local vs. non-local contributions to different pollutants over the North
American domain, three groups out of four simulated the GLO and NAM scenarios needed to
calculate the RERER metrics. RERER metrics show that O3 is largely controlled by non-local
sources. European model groups DE1 and DK1 simulate a larger influence of non-local
sources (~0.8 - ~0.9) compared to the US3 group, which simulated lower RERER metric
values of ~0.5, indicating that O3 levels are driven equally by local and non-local sources.
This lower value is also consistent with the findings of Huang et al. (2017), who simulated
the largest impacts on O3 in May and June with RERER values around ~0.5. The ensemble
mean shows that O3 responses are largely attributable to non-local sources (RERER = ~0.8),
which are similar to those found for Europe. RERER metric values calculated for NO2 by
different models (RERER = ~0 - ~0.2) and the ensemble mean (RERER = 0.05) clearly
shows that NO₂ is controlled by local sources, similar to the Europe case. The sensitivity of
CO to local and non-local sources are similar to those for O₃, with DE1 and DK1 simulating a
large contribution from non-local sources while US1 shows that CO is controlled equally by
local and non-local sources (RERER = 0.5). Similar to NO₂, all models show that SO₂ is
largely driven by local sources with RERER values between ~0.1 and ~0.2. Regarding the
particles, models simulate very similar responses to changes in the local and non-local
sources. RERER values are calculated to be ~0.08 and ~0.11 for PM₁₀ and PM₂.₅,
respectively, showing the large local contribution compared to non-local sources.

Fig. 17 shows the spatial distributions of the MMM RERER values for O₃ and PM₂.₅, as
constructed from the annual mean responses to perturbation scenarios over Europe and North
America. Fig. 17a shows that O₃ is dominantly controlled by non-local sources with RERER
values higher than 0.5 throughout the domain. Higher values are calculated over the north
western Europe, in particular over UK and the north western part of the domain covering the
Atlantic. In contrary, PM₂.₅ levels are controlled by local sources with RERER values around
0.2 (Fig. 17b). North American O₃ levels are largely controlled by non-local sources over the
western part of the domain, with RERER values above 0.5 (Fig. 17c). Local sources play a
more important role in controlling O₃ levels over the eastern part of the U.S. where much
lower RERER values are calculated. PM₂.₅ levels are dominantly controlled by the local
sources, similar to the case in Europe, with lower RERER values throughout the domain (Fig.
17d). PM₂.₅ levels over the western part of the domain has however a relatively larger
contribution from non-local sources. It is important to note that the sharp gradients in the
PM₂.₅ RERER values over both the eastern part of the Europe domain and the Mexican part
of the NA domain is due to HTAP2-definition of source regions where the perturbations are
introduced. Therefore, due to the large contribution of the local sources to PM₂.₅ levels, large
gradients are calculated across the HTAP2 borders. As O₃ is largely controlled by non-local
sources, these gradients do not exist.

In order to further analyze the impact of local vs non-local sources, the monthly variations of
RERER values for O₃ and PM₂.₅ over both domains are presented in Fig. 18. All models
simulate a larger non-local source contribution during the spring period for both domains and
pollutants. For both pollutants and domains, the local sources have relatively larger
contribution in winter periods, reflected by the lower RERER values compared to other parts
of the year. Regarding European O₃, majority of the models show a RERER value of between
0.5 and 1, while DE1 shows much lower and IT1 much higher values (see also Table 6). DE1
and FI1 simulates the lowest RERER values for PM₂.₅ (< 0.1), while other models calculate
RERER values between 0.1 and 0.5. Regarding O₃ over North America, US3 shows that in
winter months, O₃ is controlled more by local emission with RERER values much lower than
0.5, while DE1 shows the highest non-local contributions throughout the year.

CONCLUSIONS
In the framework of the third phase of the Air Quality Model Evaluation International Initiative (AQMEII3), the impacts of local vs. foreign emissions over the European and North American receptor regions are simulated by introducing a 20% decrease of global and regional emissions by research groups, using different state-of-the-art chemistry and transport models. The emission perturbations were introduced globally, as well as over the HTAP2-defined regions of Europe, North America and East Asia. Base case and the perturbation scenarios are first simulated using the global C-IFS global model, which provides the boundary conditions to the regional CTMs.

The base case simulation of each model has been evaluated against surface observations from Europe and North America. The temporal variabilities of all pollutants are well captured by all models with correlations generally higher than 0.70. O$_3$ levels are generally simulated with a MNB less than 10% with few exceptions of MNB values up to -35%. NO$_2$, CO and SO$_2$ levels are simulated with underestimations up to 75%, 45% and 68%, respectively. PM$_{10}$ and PM$_{2.5}$ levels are underestimated by 20% to 70%, with slightly higher biases in PM$_{10}$ levels.

Results from the perturbation simulations show that the largest impacts over both Europe and North American domains are simulated in response to the global emission perturbation (GLO). These responses are similar, however slightly lower, as compared to the local emission perturbation scenarios for Europe (EUR) and North America (NAM). In contrast to the GLO scenario, O$_3$ levels over Europe slightly increase by 0.13 ppb (0.02%). The annual mean value of the O$_3$ daily maximum of 8-hour running average decreases in all scenarios over Europe, highest in the GLO scenario by ~1% and lowest in the NAM scenario by ~0.3%. Over North America, the annual mean value of the O$_3$ daily maximum of 8-hour running average decreased by ~5% in the GLO scenario, 3% in the NAM scenario and 0.7% in the EAS scenario. The impact of foreign emissions simulated by the NAM scenario for Europe and EAS scenario for North America were found to be lowest, however still noticeable, particularly close to the boundaries. This impact is especially noticeable (up to only a few percent) for the western parts of the North American domain in response to the emission reductions over East Asia. The response is almost linear (~20% decrease) to the change in emissions for NO$_2$, SO$_2$ and PM in the global perturbation scenario (GLO), while O$_3$ levels decrease slightly (~1%).

Despite these small differences, there are large geographical differences. NO$_2$, CO and SO$_2$ levels are mainly affected over emission hot spots in the GLO scenario as well as in the EUR scenario for Europe and the NAM scenario for North America. O$_3$ levels increase over the hot spot regions, in particular the Benelux region in Europe, by up to ~6% due to the reduced effect of NOx-titration. Over the North American domain, the central-to-eastern part and the western coast of the U.S experience the largest response to the global emission perturbation.

For most of the pollutants, there is distinct seasonality in the responses particularly to the global and local emission perturbations. The largest responses are calculated during winter months, where anthropogenic emission are highest, except for O$_3$, where largest responses are seen during spring/summer months, suggesting photochemistry still plays an important role in O$_3$ levels.
The RERER metrics have been calculated to examine the differences in the strengths of non-local source contributions to different species among the different models. The large RERER values over Europe and North America for O₃ (~0.8), show a larger contribution of non-local sources, while for other gaseous pollutants (NO₂, CO and SO₂) and particles (PM₁₀ and PM₂.₅), low RERER values (< 0.5) indicate that these pollutants are largely controlled by local sources. Results show that the contribution of local sources on NO₂, SO₂ and PM levels are larger in North America compared to Europe, while for CO, local sources have a larger share in Europe in comparison with North America. In addition, RERER analyses shows that European O₃ is largely controlled by non-local sources (RERER > 0.5) throughout the domain. PM₂.₅ levels are largely controlled by local sources with RERER values around 0.2 throughout the domain. Local sources play a more important role in controlling O₃ levels over the eastern part of the U.S. PM₂.₅ levels over the western part of NA has a relatively larger contribution from non-local sources compared to the rest of the domain. A larger non-local source contribution during the spring period for both domains and pollutants has been calculated, suggesting long-range transport from non-local sources. For both pollutants and domains, the local sources have relatively larger contribution in winter periods, reflected by the lower RERER values compared to other parts of the year.

Overall results show that there is a large spread among the models, although the majority of the models simulate a similar seasonal variation. These differences suggest that despite the harmonization of inputs, such as emissions and boundary conditions, to regional models, there are still large differences between models, such as different gas phase and aerosol modules, deposition schemes, meteorological drivers and spatial and vertical resolutions. Therefore, the use of multi model ensembles can help to reduce the uncertainties inherent in individual models.

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Fishing and Environment. University of Murcia thanks the Spanish Ministry of Economy for the research contract CGL2014-59677-R (also partially funded by the FEDER programme).

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Table 1. Key features (meteorological/chemistry and transport models, emissions, horizontal and vertical grids) of the regional models participating to the AQMEII3 health impact study and the perturbation scenarios they performed.

<table>
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<tr>
<th>Group Code</th>
<th>Model</th>
<th>Emissions</th>
<th>Horizontal Resolution</th>
<th>Vertical Resolution</th>
<th>Gas Phase</th>
<th>Aerosol Model</th>
<th>Europe</th>
<th>North America</th>
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<tr>
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<td>COSMO-CLM/CMAQ</td>
<td>HTAP</td>
<td>24 km × 24 km</td>
<td>30 layers, 50 hPa</td>
<td>CB5-TUCL</td>
<td>3 modes</td>
<td>BASE</td>
<td>GLO NAM EUR</td>
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<tr>
<td>DK1</td>
<td>WRF/DEHM</td>
<td>HTAP</td>
<td>50 km × 50 km</td>
<td>20 layers, 100 hPa</td>
<td>Brandt et al.</td>
<td>2 modes</td>
<td>BASE</td>
<td>GLO NAM EUR</td>
</tr>
<tr>
<td>ES1</td>
<td>WRF/CHIMERE</td>
<td>MACC</td>
<td>23 km × 23 km</td>
<td>22 layers, 50 hPa</td>
<td>RADMC2</td>
<td>3 modes</td>
<td>BASE</td>
<td>GLO NAM EUR</td>
</tr>
<tr>
<td>FI1</td>
<td>ECMWF/SILAM</td>
<td>MACC+HTAP</td>
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<td>12 layers, 13 km</td>
<td>CB4</td>
<td>1.5 bins, VBS</td>
<td>BASE</td>
<td>GLO NAM EUR</td>
</tr>
<tr>
<td>FRES1</td>
<td>ECMWF/CHIMERE</td>
<td>HTAP+HTAP</td>
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<td>9 layers, 50 hPa</td>
<td>MELCHIOR2</td>
<td>8 bins</td>
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<tr>
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<td>WRF/CHIMERE</td>
<td>MACC</td>
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<td>15 layers, 50 hPa</td>
<td>RACM-ESHR</td>
<td>1 modes, MADE/VBS</td>
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<td>GLO NAM EUR</td>
</tr>
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<td>WRF/CAMx</td>
<td>MACC</td>
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<td>CB5</td>
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<td>BASE</td>
<td>GLO NAM EUR</td>
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<tr>
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<td>MACC</td>
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<td>22 layers, 100 hPa</td>
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<td>3 modes</td>
<td>BASE</td>
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<td>CB5-TUCL</td>
<td>3 modes</td>
<td>BASE</td>
<td>GLO NAM EUR</td>
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<td>WRF/CMAQ</td>
<td>HTAP</td>
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<td>21 layers, 100 hPa</td>
<td>CB5-TUCL</td>
<td>3 modes</td>
<td>BASE</td>
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</tr>
<tr>
<td>US3</td>
<td>WRF/CMAQ</td>
<td>SMOKE</td>
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<td>15 layers, 50 hPa</td>
<td>CB5-TUCL</td>
<td>3 modes</td>
<td>BASE</td>
<td>GLO NAM EUR</td>
</tr>
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</table>

1 MACC: Modelling group used only the MACC emissions, MACC+HTAP: Modelling group used MACC emissions for Europe and HTAP emissions over North Africa.
Table 2. Perturbations of global/regional anthropogenic emissions and boundary conditions in the perturbation scenarios.

<table>
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<th>North America</th>
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<td>EUR</td>
</tr>
<tr>
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<td>-</td>
<td>-20%</td>
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<tr>
<td>Boundary conditions (Emissions in the IFS model)</td>
<td>-20%</td>
<td>-20%</td>
<td>-20%</td>
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### Table 3. Monthly statistics of Pearson’s Correlation (r), Normalized Mean Bias (NMB), Normalized Mean Gross Error (NMGE) and Root Mean Square Error (RMSE: µg m⁻³ for Europe, while ppb for gases and µg m⁻³ for particles for North America) calculated for each model group.

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<td>O₃</td>
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<tr>
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<td>0.75</td>
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<tr>
<td>RMSE</td>
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<td>NO₂</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>r</td>
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<tr>
<td>RMSE</td>
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<td>SO₂</td>
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<td>r</td>
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<td>PM₁₀</td>
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<tr>
<td>PM₂₅</td>
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<tr>
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Table 4. Annual mean absolute differences (ppb for gases and µg m\(^{-3}\) for particles) between the base case and the different emission perturbation scenarios as calculated by the different model groups over the European domain.

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<th>FI1</th>
<th>IT1</th>
<th>IT2</th>
<th>TR1</th>
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Table 5. Annual mean absolute differences (ppb for gases and µg m⁻³ for particles) between the base case and the different emission perturbation scenarios as calculated by the different model groups over the North American domain.

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Table 6. Annual mean RERER values calculated for the multi-model mean ensembles over Europe and North America.

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Fig. 1. Observed and simulated monthly mean air pollutant levels, averaged over the monitoring stations over Europe.
Fig. 2. Observed and simulated monthly mean air pollutant levels, averaged over the monitoring stations over North America.
Fig. 3. Multi-model mean air pollutant levels over Europe as simulated in the base case.
Fig. 4. Multi-model mean air pollutant levels over North America as simulated in the base case.
Fig. 5. Absolute impact of the 20% reduction of the global anthropogenic emissions over Europe (GLOeur-BASEeur).
Fig. 6. Spatial distribution of the annual mean relative differences between the global perturbation scenario and the base case over Europe as simulated by the multi-model mean ensemble.
Fig. 7. Absolute impact of the 20% reduction of the global anthropogenic emissions over North America (GLO\textsubscript{NAM}-BASE\textsubscript{NAM}).
Fig. 8. Spatial distribution of the annual mean relative differences between the global perturbation scenario and the base case over North America as simulated by the multi-model mean ensemble.
Fig. 9. Absolute impact of the 20% reduction of the North American anthropogenic emissions over Europe (NAMEUR-BASEEUR).
Fig. 10. Spatial distribution of the annual mean relative differences between the North American emissions perturbation scenario and the base case over Europe as simulated by the multi-model mean ensemble.
Fig. 11. Absolute impact of the 20% reduction of the North American anthropogenic emissions over North America (GLO\textsubscript{NAM}-BASE\textsubscript{NAM}).
Fig. 12. Spatial distribution of the annual mean relative differences between the North American emissions perturbation scenario and the base case over North America as simulated by the multi-model mean ensemble.
Fig. 13. Absolute impact of the 20% reduction of the European anthropogenic emissions over Europe (EUR$_{EUR}$-BASE$_{EUR}$).
Fig. 14. Spatial distribution of the annual mean relative differences between the European emissions perturbation scenario and the base case over Europe as simulated by the multi-model mean ensemble.
Fig. 15. Absolute impact of the 20% reduction of the East Asian anthropogenic emissions over North America (GLO\textsubscript{NAM}-BASE\textsubscript{NAM}).
Fig. 16. Spatial distribution of the annual mean relative differences between the East Asian emissions perturbation scenario and the base case over North America as simulated by the multi-model mean ensemble.
Fig. 17. Spatial distribution of RERER values constructed from the annual mean responses of O₃ and PM₂·₅ over Europe and North America.
Fig. 18. Seasonal variations of RERER values of O₃ and PM₂.₅ over Europe and North America.