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The effects of intercontinental emission sources on European air pollution levels

Jan Eiof Jonson[1], Michael Schulz[1], Louisa Emmons[2], Johannes Flemming[3], Daven Henze[4], Kengo Sudo[5], Marianne Tronstad Lund[6], Meiyun Lin[7], Anna Benedictow[1], Brigitte Koffi[8], Frank Dentener[8], Terry Keating[9], and Rigel Kivi[10]

[1]Norwegian Meteorological Institute, Oslo, Norway
[2]National Center for Atmospheric Research Boulder, Colorado, USA
[3]ECMWF (European Centre for Medium Range Forecast)
[4]University of Colorado Boulder, Colorado, USA
[5]NAGOYA-U,JAMSTEC,NIES, Japan
[6]Center for International Climate and Environmental Research (CICERO) - Oslo, Norway
[7]Program in Atmospheric and Oceanic Sciences of Princeton Univeristy and NOAA Geophysical Fluid Dynamics Laboratory, Princeton, New Jersey, USA
[8]European Commission, Joint Research Centre, Ispra, Italy
[9]U.S. Environmental Protection Agency
[10]Finnish Meteorological Institute, Helsinki, Finland

Correspondence to: Jan Eiof Jonson (j.e.jonson@met.no)

Abstract. This study is based on model results from TF HTAP (Task Force on Hemispheric Transport of Air Pollution) phase II where a set of source receptor model experiments have been defined, reducing global (and regional) anthropogenic emissions by 20% in different source regions throughout the globe, with main focus on year 2010. All the participating models use the same set of emissions. Comparisons of model results to measurements are shown for selected European surface sites and for ozone sondes, but the main focus here is on the contributions to European ozone levels from different world regions, and how and why these contributions differ depending on model. We investigate the origins by use of a novel stepwise approach combining simple tracer calculations and calculations of CO and O_3. To highlight differences, we analyse the vertical transects of the mid latitude effects from the 20% emission reductions.

Based on the relative emission changes from different world regions the models agree 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 contribution will however depend on the choice of ozone metric. There are also considerable contributions from other nearby regions to the east and from international shipping, Whereas ozone from European sources peaks in the summer months, the largest contributions from non European sources are mostly calculated for the spring months when ozone production over the polluted continents starts to increase, while at the same time the lifetime of ozone in the free troposphere is relatively long. At the surface contributions

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from non European sources are of similar magnitude for all European sub regions considered, defined as TF HTAP receptor regions (north west, south west, east and south east Europe).

1 Introduction

This paper is based on the HTAP model experiment phase 2 (HTAP2), where chemical tracer models perform model sensitivity studies, perturbing the emissions in different world regions. TF HTAP (http://www.htap.org/ is organized under the auspices of the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP Convention) The HTAP2 experiment is described in more detail in Galmarini et al. (2017) and in the HTAP2 work plan, posted on the HTAP2 web site www.htap.org. All models should use the same set of emissions, see Janssens-Maenhout et al. (2015).

In particular the experiments is set up to:

- Examine the transport of air pollution, including ozone and its precursors and particulate matter and its components (including black carbon), across the Northern Hemisphere.
- Assess potential emission mitigation options available inside and outside the UNECE region.
- Assess their impacts on regional and global air quality, public health, ecosystems, and near-term climate change.
- Promote collaboration both inside and outside the Convention.

HTAP2 is a follow up of the HTAP phase 1 model experiment (HTAP1). Results from HTAP1 has been described in a series of peer review papers, including (Casper-Anenberg et al., 2009; Fiore et al., 2009; Reidmiller et al., 2009; Jonson et al., 2010; Sanderson et al., 2008; Shindell et al., 2008), and in the the HTAP1 main report (TF HTAP, 2010). The HTAP1 model experiment showed that intercontinental transport of ozone and ozone precursors could explain a large portion of the ozone over Europe, but results differed substantially between the models.

A large number of CTMs and GCMs have uploaded their results to the HTAP2 database. This study is limited to those models that, in addition to the base run, as a minimum have uploaded their source receptor calculations for ozone reducing all anthropogenic global emissions and European emissions by 20%. Seven of the models fulfil these criteria.

Several papers from HTAP2 have been published. Janssens-Maenhout et al. (2015) describes the common set of emissions to be used by the models. Stjern et al. (2016) study the effects of changes in the atmospheric load of black carbon (BC), organic aerosols (AE) and sulphate on radiative forcing. Using a regional model with lateral boundaries from a global model, Huang et al. (2017) study the effects of intercontinental ozone on North America. With a regional model citeKaramchandan2017 calculates the contributions from several emission sectors, including model boundary conditions, to tropospheric O_3 and $PM_{2.5}$. Turnock et al. (2018) use the HTAP2 source receptor relationships to

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parametrize future changes in ozone. Several additional HTAP2 papers have also been published or have been submitted for review.

In this paper we aim to enhance our understanding of the contributions to European ozone levels from European and non-European sources. In order to better understand the transport patterns between the continents we use a novel stepwise approach, starting with a simple CO like tracer using the CO anthropogenic emissions and a fixed decay rate of 50 days. As all models use the same emissions, differences in model results can be ascribed to differences in transport (advection, including also convection and diffusion) only. Unfortunately only two models have provided such tracer data for at least the BASE, GLOALL and EURALL model run, while a third model has provided tracer data for BASE and GLOALL. Secondly we look at CO. The main sink for CO is the reaction with OH, and thus differences in OH is one of the main factors affecting CO. Finally we look at ozone. The causes of the differences in calculated ozone are hard to identify, but some clues can be identified based on the calculations of the CO like tracer and CO.

In this paper we first briefly discuss the model comparison to measurements in section 3. In section 4 we go on to describe the source receptor relationships for Europe, including a discussion on how and why the model results differ. Finally we discuss how this information on how models compare to measurements, and in what way the model results differ could be used to harmonize and improve future model calculations in section 5.

2 The HTAP2 model setup

The HTAP2 model model experiment was set by the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) A project work plan, a description of the model experiment etc can be found on the TF HTAP web page (http://www.htap.org/). A more detailed description of the requested model runs, emissions, requested model output and formats etc is also included in (Galmarini et al., 2017) and references therein. A detailed description of the emissions can be found in Janssens-Maenhout et al. (2015). More documentation about the models can also be found in the supplementary material.

In this paper we focus on the effects on Europe. Even though a substantial number of models have uploaded their results to the database, model results for ozone (and CO) are only available from 7 of the models for the BASE model runs and for at least the two scenario runs reducing all anthropogenic emissions by 20% globally (GLOALL) and in Europe (EURALL). These models have different resolutions, chemical mechanisms etc. Additional model runs reducing all anthropogenic emissions in North America (NAALL), East Asia (EAALL), South Asia (SAALL), Middle East (MDALL), Russia, Belarus, Ukraine (RBUALL) and ship emissions (OCNALL) are also discussed here. The definition of these regions is given in Koffi et al. (2016). The models are a subset of the HTAP2 models listed and described in Stjern et al. (2016). Since then additional model result have also been provided for theGFDL_AM3 model (but not uploaded to the HTAP2 database), raising the number

of models to 8. Additional information on the models are also listed in the supplementary material. Access to model data are available upon registration from http://aerocom.met.no.

3 Models vs measurements

In this section we briefly discuss the performance of the models compared to measurements. For ozone a comprehensive model to measurementcomparison is published in Galmarini et al. (2017), including a comparison of both global and regional model results. For surface ozone we therefore refer to this paper. Comparisons of model calculated vertical profiles to ozone soundings are included in the supplementary material. As the focus of this paper in on Europe, only European sites are shown. We have only included models with model output also for the GLOALL and the EURALL scenarios.

3.1 Surface

Monthly averaged timeseries of measured versus model calculated CO are shown in the supplementary material for a number of European GAW sites. Some statistics for these sites are listed in Table 1. At most sites CO has a clear winter maximum and a summer minimum. All models in general reproduce the seasonal cycle well at most sites, reflected in their high correlations with the measurements. The results for the two CHASER model versions with high (1.1 $\times$ 1.1 degrees) versus low (2.8 $\times$ 2.8 degrees) resolutions differ, but they are qualitatively similar. Some sites with very high concentrations (as Hegyhatsal) are clearly affected by local/regional sources not resolved by the global models.

This study also includes an evaluation of model results at several mountain sites. Results for these sites are shown, but should be interpreted with great caution. The elevation of mountain sites are poorly resolved in the models. Furthermore concentrations are likely to be affected by sub scale circulation patterns as mountain subsidence and upslope winds etc, that are not resolved by the models.

A comparison of the Base model calculations and ozone measurements from the EMEP and airbase measurement networks is presented in Galmarini et al. (2018) as part of HTAP 2 and ACMEII. Therefore we do not include an extensive comparison of measured and model calculated ozone here. Scatter plots for the BASE model runs for ozone versus measurements are shown in the supplementary material. We only show results for one of the CHASER models as the two versions again are similar.

3.2 Vertical ozone profiles

Seasonal model calculated vertical profiles of ozone are compared to ozone sonde measurements for several European sites in the supplementary material. Model calculated profiles are included in the calculations for the approximate same point in time as the ozone sondes, and then averaged seasonally. The number of soundings included in the average for any site and season is listed in the individual panels.

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The vertical profile from the GEOSCHEMADJ model relax to zero in the upper part, and this is an artefact of the ajoint method. There is a tendency for most models to underestimate ozone in the free troposphere in the summer (JJA) months.

4 Source allocation, focusing on Europe

In this section we use the models to allocate the sources of ozone from different world regions, focusing on effects on European ozone levels. In order to better understand the differences between the models, we use a step-wise approach, starting the discussion with the CO like tracer, then we compare results for CO, where the treatment of the sources should be similar in all models, and the main sink is through the reaction with OH. Finally we compare the model results for O_3.

The calculations of the anthropogenic contributions from the different source regions are based on the difference between the base model runs and HTAP2 model scenario runs reducing all anthropogenic emissions globally (GLOALL), in addition to the reductions in the specific HTAP2 regions. We first compare the model calculated effects of the GLOALL scenario for vertical trans-sections, and discuss the source allocation of domestic European anthropogenic sources versus external trans-continental anthropogenic sources expressed as "response to extra-regional emission reductions" (RERER) (Galmarini et al., 2017)

$$RERER = \frac{EURALL - GLOALL}{BASE - GLOALL}.$$

Again, BASE is the reference model run and EURALL the model runs reducing all European emissions by 20%. RERER is then a measure of the effects of external trans-continental versus domestic European emissions on the species in question. Given a fully linear chemistry, a RERER of one means that the concentrations in Europe are completely determined by sources outside Europe, whereas a RERER of 0 means that concentrations are determined by European sources alone. Unfortunately the chemistry is often far from linear. In particular for ozone, ozone titration, mainly in the winter months, can result in RERER values well above one, and in some cases even negative. In the section below annual RERER values are given for Europe as a whole and for four separate receptor regions, NW, SW, SE and GR+TU as shown in Figure 1.

For ozone we also show the source attribution of European ozone further split into separate world regions for the the different models on a seasonal basis.

4.1 CO tracer

The CO tracer is calculated with the same anthropogenic emissions as CO, and with a set rate of decay giving a lifetime of 50 days. Any differences between the individual models can then be attributed to differences in transport processes.

Table 2, lists RERER calculated by the EMEP_rv48 and the IFS_v2 models for Europe and the four European sub regions. For Europe as a whole, RERER is also shown in Figure 2. For the CO

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tracer RERER is ranging from 0.35 to 0.60, depending on model and European sub-region. There is a moderate difference in RERER between the two models. The highest RERER is calculated for the Gr+Tr region as this region is close to regions outside Europe as Russia, Belarus, the Ukraine, the Middle East and also the Mediterranean Sea.

Figure 3a,d,g shows the annual mean difference in BASE - GLOALL of longitudinal CO tracer concentrations as an average between 30 and 60 degrees north. For all 3 models (EMEP_rv48, IFS_v2 and GFDL AM3) the largest impacts of the 20% emission reduction on concentrations can be seen over the source continents in North America, Europe and in particular over east Asia. There are marked differences between the models as to what extent the CO tracer from the polluted boundary layer is lifted into the free troposphere. The EMEP model (Figure 3b), with high RERER (Table 2) has somewhat higher tracer contributions in the free troposphere than the other two models (Figure 3d,g). This may indicate that material lifted into the free troposphere, possibly through too strong convection, is transported further from its sources and subsequently contributes more to the tracer levels in distant regions before being decayed.

The seasonal cycle of the difference in BASE - GLOALL the over Europe, defined as the area bounded by 10°W to 50°E and 25 to 65 °N, is shown in Figure 4a,d,g. Differences in concentrations peak in the first part of the year when emissions are high and the exchange between the boundary layer and the free troposphere is weak. There are moderate differences between the two models, but tracer levels in the free troposphere are again highest in the EMEP model.

4.2 CO

Emissions of CO and the CO tracer are identical, and the results for CO resemble the results for the CO tracer in section 4.1. The dominant sink for CO in the atmosphere is the reaction with the OH radical, with a winter minimum and peaking in summer.

Table 2 lists RERER values for the seven models for Europe as a whole and for the four European sub regions shown in Figure 1. RERER is ranging from 0.24 to 0.71, depending on model and European sub-region. Differences between the models are now caused by transport (as for the CO tracer) and chemistry. For the EMEP_rv48 and IFS_v2 models RERER is higher than for the CO tracer. Assuming that the CO chemistry is close to linear, this indicates a longer lifetime in the atmosphere than the 50 days for the CO tracer. IPCC Working group 1: the scientific basis, https://www.ipcc.ch/ipccreports/tar/wg1/130.htm#tab41a) reports a lifetime of 0.08 to 0.25 years (about 30 to 90 days) depending on location and season, on average longer than 50 days.

As shown in Table 2 and Figure 2, the spread in RERER between the models is again moderate. For the EMEP_rv48 and IFS_v2 models the difference in RERER is slightly larger than for the CO tracer. As for the CO tracer, the highest RERER is in general calculated for the GR+TR region as this region is close to the outer border of the European domain.

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Figure 3c,e,h and 4a,b,c shows the annual mean difference in BASE - GLOALL CO concentrations as an average between 30 and 60 degrees north. For all the models large differences in concentrations can be seen over the polluted continents North America, Europe and in particular over East Asia. As for RERER, there are differences between the models, in particular in the free troposphere. The EMEP_rv48 model (Figure 3b), with high RERER, has higher CO contributions in the free troposphere than the other models. As CO is lifted into the free troposphere transport between continents is rapid, and CO can be transported further before decay, suggesting that the higher RERER is a result of rapid lifting and subsequent efficient intercontinental transport in the free troposphere.

The seasonal cycle of the difference in BASE - GLOALL over Europe is shown in Figure 4, middle panels. As for the CO tracer, differences in concentrations peak near the surface in the first part of the year when emissions are high and the exchange between the boundary layer and the free troposphere is weak. In addition the differences are magnified by the seasonal cycle in the OH sink.

We don't have access to the OH levels for all the models, but for those models providing OH (EMEP_rv4.8, CHASER_re1, OsloCTM3 and CAMchem) annually averaged tropospheric levels are shown in the supplementary material along with the difference between the average and the four individual models. OH levels in the EMEP_rv4.8 model are low compared to the average, at least in the upper and middle troposphere. This may lead us to suspect that the widening gap in RERER from CO tracer to CO between the IFS_v2 and the EMEP_rv4.8 model is caused by differences in OH (however, this can not be confirmed, as OH is not available from the IFS_v2 model). Likewise, the higher than average OH levels in the OsloCTM3 model may explain the lower than average CO RERER values for this model.

Furthermore the lifting of pollutants from the boundary level to the free troposphere is likely to affect the chemistry in the free troposphere this causing (parts of) the differences in OH. The EMEP_rv48 does not perturb aircraft emissions in the BASE-GLOALL scenario, and this could explain large parts of the differences between this model and the 3 other models. See also discussion on ozone in section 4.3 below.

4.3 O_3

Tropospheric Ozone differs from CO and the CO tracer as it is not emitted, but rather it is a secondary product involving combinations of chemical production and loss processes, exchange with the stratosphere, surface deposition and transport. Ozone in the troposphere is advected from the stratosphere mainly by stratospheric folding events, but its main sources (and sinks) are in the troposphere. Net ozone production require ample sunlight and a sufficient supply (and mix) of mainly NMVOC, CH_4 CO and NO_x.

Table 2, lists annual average RERER, for Europe and for the four European sub regions. RERER is ranging from 0.56 to 1.38, depending on model and European sub-region. As seen in Table 2 and Figure 2 O_3 RERER values are higher than for the CO tracer and for CO even though its lifetime in

the atmosphere is ranging from 0.01 to 0.05 years (about 4 to 20 days) for ozone, see IPCC Working group 1: the scientific basis, https://www.ipcc.ch/ipccreports/tar/wg1/130.htm#tab41a). The high RERER values are therefore caused by the non-linear chemistry that for some models can result in RERER values even exceeding one, and for seasonal RERER even negative values (not shown). The spread in RERER between the individual models is markedly larger than for CO and the CO tracer. Differences in transport, depositions and in particular a nonlinear chemistry, give substantial room for variability in ozone levels between the models. In NW Europe little sunlight throughout much of the year as a result of its northerly location and high cloud fractions, in combination with high NO_x emissions, result in ozone titration and calculated RERER around 1 for a majority of the models. The lowest RERER is calculated for the Gr+Tr (Greece + Turkey) and partially SW European regions. The EMEP and the IFS are the only two models where RERER can be calculated for the CO tracer, CO and ozone. Whereas for the IFS_v2 model RERER is lower than the EMEP_rv48 model for the CO tracer and for CO, RERER jumps to well above one for ozone, well above any of the other models.

Based on the HTAP2 model calculations, Huang et al. (2017) have calculated RERER for the North American continent. In general these RERER values are markedly lower than for Europe. In addition to the effects of little sunlight discussed above, less favourable for local ozone production, Europe is also affected by nearby source regions as Russia, Belarus, Ukraine, the Middle East, North Africa and shipping. These two factors are likely to explain the higher RERER values over Europe compared to North America.

Figure 3c,f,i and 4d,e,f shows the annual longitudinal mean difference in BASE - GLOALL O_3 concentrations as an average between 30 and 60 degrees north. The differences between the models are markedly larger than for CO and the CO tracer. One notable difference stems from the interpretation of the scenario definition. The Oslo CTM2 model, CAMchem model and the CHASER models have included a 20% emission reduction also in aircraft emissions in the GLOALL scenario, whereas the EMEP_rv48 model, the IFS_v2 and the GEOSchem adjoint models have not. As a result the additional ozone from BASE - GLOALL is much higher in the middle and upper troposphere for the first three models listed. For the Oslo CTM3 model the O_3 signal from aircraft emissions is located much lower in the troposphere than for the CAMchem and CHASER models. O_3 in the lower troposphere, and in particular in the boundary layer, are not so much affected by the aircraft emissions. But also here the models differ substantially. As is the case for CO and the CO tracer, the EMEP model (Figure 3c), has higher O_3 contributions in the free troposphere than the IFS_v2 and GEOSCHEMADJ models (the two other models not perturbing aircraft emissions).

The seasonal cycle of the difference in BASE - GLOALL over Europe is shown in Figure 4 right panels. Whereas the contributions from aircraft peaks in summer and autumn, the differences in BASE - GLOALL in general peaks in spring in the lower troposphere.

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4.4 O_3 source allocations by world region

Based on the difference between the BASE model runs and the 20% perturbations of global and European emissions (not accounting for the effects of CH_4) we attribute a major portion of ozone of anthropogenic origin in Europe to sources outside Europe. As part of the HTAP2 requests, model calculations have also been made reducing anthropogenic emissions by 20% in other major world regions. In Figure 5 the contributions to European ozone levels calculated by the different models are shown with sources originating from these different world regions. None of the models have made the calculations for all the regions. For each model the contribution from other regions is calculated by subtracting the added regional contributions from the BASE - GLOALL contribution. Thus the portion related to other regions includes a varying number of region definitions depending on the model.

There are large differences between the models, but there are some common features: For all models and all seasons except for the CHASER re1 in summer, the contributions from regions outside Europe are larger than the contribution from European sources. The contributions from non European sources are largest in Spring. The largest non European contributions are from North America (NAMALL) and East Asia (EASALL). Contributions from Russia, Belarus, Ukraine (RBUALL) and the Middle East (MDEALL) are mixed, with significant calculated contributions calculated by two models (EMEP_rv48 and CHASER). There are also substantial contributions from ocean shipping (OCNALL), but this source has only been calculated by the EMEP model. For Europe substantial contributions from shipping has also been shown in other studies as Brandt et al. (2013); Jonson et al. (2015).

Compared to the HTAP1 calculations (TF HTAP, 2010) the contribution to European ozone levels from the world regions differ. Part of the reason may be that the models included in the HTAP1 and HTAP2 ensemble are not the same. From 2001 to 2010 emissions of ozone precursors have decreased in USA (see https://www.epa.gov/air-emissions-inventories/air-pollutant-emissions-trends-data and Europe (Mareckova et al., 2017), and increased in East Asia. Turnock et al. (2018) lists the percentage change in the HTAP1 world regions confirming these emission trends between 2001 (the base year for HTAP1) and 2010. This may explain the 0.37 to 0.22 ppb decrease in the contributions from North America, and the 0.17 to 0.22 ppb increase in the East Asian contribution from HTAP1 to HTAP2. In HTAP1 the EUR region was a simple latitude - longitude box, also including parts of North Africa, the Middle East, Russia, Belarus, Ukraine and large sea areas included as separate, non European regions in HTAP2. As a result emissions from the EUR region is no longer comparable. Contributions from South Asia are small in both HTAP1 and HTAP2 (0.07 versus 0.05). A combined effect of the change in the definition of the European domain and the changes in emissions is that the relative model calculated contributions to surface ozone levels from non European sources is much larger in HTAP2 compared to HTAP1.

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The calculated contributions from non European sources have also been calculated by Karamchandani et al. (2016) using a regional model. They too calculate a much smaller contribution from non European sources than in this study. In the Karamchandani et al. (2016) study non European ozone is defined as the boundary influx to the model domain. As a result shipping, and nearby non Central European regions, are included in the domain, similar to the definition of the HTAP1 European domain.

4.4.1 Effects of a 20% CH_4 perturbation

As shown in Figure 5 four of the models have also calculated the effects of a 20% increase in CH_4 concentrations. Averaged over the four models the calculated effects for Europe of 20% changes in CH_4 levels is almost three quarters of the effects of the BASE - GLOALL model runs. However, a direct comparison of a 20% change in CH_4 concentrations and the effects of the GLOALL scenario should not be made. Because of its relatively long lifetime of the order of 10 years in the atmosphere a 20% change in concentrations corresponds to an approximate 40 years of historic CH_4 trends (Meinshausen et al., 2011). The effects of CH_4 is insensitive to the location of the emissions, and there are only moderate differences in the response in ozone levels by world region (Fiore et al., 2008). The agreement between the model estimates is a lot better for the CH_4 perturbation compared to the BASE - GLOALL estimates, and not too different for the HTAP I estimate of about 1 ppb (Fiore et al., 2008). The sensitivity of ozone to CH_4 is discussed in more detail in Turnock et al. (2018).

4.5 Does the choice of ozone metric matter?

In Figure 5 the contributions to European ozone levels are shown as seasonal and annually averaged ozone. In Europe several other metrics are also used calculating the effects of ground level ozone. The two metrics listed below are designed to capture the effects of ground level ozone on human health (SOMO35) and on the environment (POD_1 forest):

- SOMO35: Sum of Ozone Means Over 35 ppb is the indicator for health impact assessment recommended by WHO. It is defined as the yearly sum of the daily maximum of the running 8-hour running average of ozone above 35 ppb.
- POD_1 forest: Phyto-toxic Ozone Dose for forests is the accumulated stomatal ozone flux over a threshold Y integrated from the start to the end of the growing season. For deciduous forests, discussed here, the critical level of 4 mmol m^{-2} is exceeded in most of Europe, indicating a risk of ozone damage to forests. See Mills et al. (2011a, b) for further description of this metric.

Unfortunately the two latter metrics have only been provided by the EMEP model. The annual effects of the 20% reductions in anthropogenic emissions from different world regions are shown for mean ozone, SOMO35 and POD_1 (deciduous) forest in Figure 6 as percentage contributions where

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100% refers to the difference betwee the BASE and GLOALL scenario. The figure clearly shows that the choice of metric matters, in particular for the effects of European Emissions.

POD_1 forest is only accumulated over the growing season, in particular excluding the winter month with frequent ozone titration from local sources. This metric is mostly accumulated in the summer, when the contributions from local European sources are high. Likewise SOMO35, with a cutoff value at 35 ppb, is accumulated mainly in the summer months when local ozone production peaks over the European continent.

Contributions to annual mean ozone are accumulated regardless of season and ambient ozone levels. In the EMEP model contributions from NAM and EAS have already been shown to be little affected by ozone titration and a major source mainly in the spring months before the local European sources gathers momentum. Contributions from RBU and OCN are a mixture of nearby and more distant sources, and effects on annual mean ozone, SOMO35 and POD_1 forest are similar. It is likely that the difference between the ozone metrics would be considerably larger if calculated with the other models, and in particular those models with substantial titration effects from European Emissions as already shown in Figure 5.

5 Discussion on individual models

As shown above there are large differences between the models. Furthermore, these differences amplify going from the simple CO tracer, via CO, to ozone. This stepwise amplification provides an opportunity to pinpoint probable causes. At the same time we also use the comparisons to measurements as a guidance. Some of the results from the individual model calculations are summed up in Table 3. Below we discuss the characteristics and the results for the individual models.

The horizontal resolution of the EMEP_rv48 model is 0.5 $\times$ 0.5 degrees, higher than any of the other models. Compared to the other models, the difference between BASE and GLOALL is among the highest compared to the other models for CO and the CO tracer. Much of this may be caused by a larger rate of exchange (possibly by convection) between the boundary layer and the free troposphere. On the other hand this model performs among the best both for CO and ozone compared to measurements. Calculated CO levels at remote sites are not high, see Table 1) and supplementary material, compared to the other models. Ozone sondes show overestimation of ozone in free troposphere in winter and spring months.

The horizontal resolution of the IFS_v2 model is 0.7 $\times$ 0.7 degrees. The RERER results for CO are close to the ensemble mean and CO levels close to observations. For ozone RERER is higher than the other models, and above 1 in all European regions except Greece and Turkey. European net Ozone production strongly affected by ozone titration resulting in net ozone loss from European sources for all seasons except summer. Calculated ozone levels in Europe are low compared to measurements.

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The horizontal resolution of the OsloCTM3 model is 2.8 × 2.8 degrees. For CO RERER is well below the model ensemble mean. A possible reason for this is that the advection is solved using the Prather scheme, giving very little numerical diffusion, possibly restraining the vertical exchange more than for the other models. The model Underestimates CO, and overestimates O_3 compared to measurements.

The two models CHASER re1 (resolution 2.8× 2.8 degrees) and CHASER t106 (resolution 1.1× 1.1 degrees) differ only in resolution, and results from the two models are very similar. RERER for CO close to ensemble mean. RERER for ozone almost 30% lower than ensemble mean.

The horizontal resolution of the GEOSCHEMADJ model is 2.0 × 2.5 degrees. CO concentrations underestimated by 20+ percent. O_3 concentrations overestimated by 14%. The vertical profiles of ozone relax to zero above the tropopause.

RERER calculated by the GFDL AM3 model is close to the ensemble mean for both CO and O_3. RERER for CO 20% below ensemble mean. RERER o3 17% higher than the ensemble mean.

The horizontal resolution of the CAMchem model is 1.9 × 2.5 degrees. CO concentrations are underestimated by 25% and O_3 concentrations overestimated by 22%. RERER close to ensemble mean for both CO and O3.

6 Conclusions

The HTAP1 experiment showed a very large spread in model results. (TF HTAP, 2010). Part of this spread may have been caused by differences in the 2001 emissions, as each modelling group used their own set of emissions. In HTAP2 all models are required to use a common set of emissions. Even so, the spread in model results remains large. The model calculated relative contributions to surface ozone levels from non European sources is much larger in HTAP2 compared to HTAP1. In parts differences could be explained by decreasing emissions in North America and Europe and increased emissions in other regions as East Asia from year 2001 to 2010. However, the results from the two HTAP phases can not easily be compared, partially because the model ensemble has changed, but mainly because the definition of the European area has changed considerably from HTAP1 to HTAP2. The HTAP2 source and receptor regions are better designed for characterising export and import of air pollution to and from the individual regions. For HTAP2 additional diagnostics were defined which allow better understanding of transport efficiencies, such as the utilisation of idealized CO tracer and more information on the vertical distribution of tracers in the output requirements.

Not surprisingly, our study reveals that the magnitude of the inter-model spread in hemispheric transport, characterised by RERER, increases with the complexity of the processes involved. We demonstrate that the spread in European RERER increases from the idealized CO tracer to fully prognostic CO and ozone. Atmospheric transport alone can not be made responsible for the large RERER difference between CO and ozone, as the residence time in the troposphere is of the order

of 0.01 to 0.05 years for ozone, and for CO considerably longer, 0.08 to 0.25 years, see IPCC Working group 1: the scientific basis, https://www.ipcc.ch/ipccreports/tar/wg1/130.htm#tab41a). For comparison the 50 days lifetime of the CO tracer translates to about 0.14 years. The increase in RERER from CO to O_3 is likely caused by more complex non-linear chemistry forming ozone and not by a longer atmospheric lifetime of O_3 compared to CO.

Model results from the two CHASER models, differing in model resolution only, are qualitatively similar when compared to measured CO and O_3 at background measurement sites. Horizontal resolution does not affect the source receptor calculations much at intercontinental sales.

The joint and consistent analysis of a CO tracer, CO and O_3 in this paper is a tool in understanding where and why (right or wrong) the models differ, however, it could probably be used more. and as a result enhancing our understanding of the result an also as a tool for model improvements, reducing the overall uncertainty in future model calculations. We believe that in order to close the gap in model results, and subsequently improving the reliability of the model output, possible future model inter-comparisons should be more process oriented (transport, depositions, chemistry etc). The largest spread in model results is clearly induced by differences in the model chemistry. Our study shows that the models differ markedly already for CO (and CO tracer with 2 models). We believe that a comparison of the chemical mechanisms used in the models, in combination with an extensive evaluation with atmospheric measurements, may be a first step improving the models.

The HTAP2 results, using state of the art global models, reflecting updated emission estimates and refined receptor region definitions, confirm the importance of hemispheric transport of air pollution. Based on seasonal and annual averaged ozone, all the models agree that the contribution from non European sources to European surface ozone levels is considerable. However, calculations with the EMEP_rv4.8 model shows that this conclusion to some extent will depend on the choice of ozone metrics. Alternative metrics, such as SOMO35 and POD_1 forest, will to a larger extent accumulate in the summer months when ozone production peaks over the European continent. As a result the potential for reducing the detrimental effects from ozone caused by European emissions alone is higher when applying these metrics.

The model results suggest that it will be difficult to achieve sizeable reductions in ozone levels with European emission reductions alone, and that reductions in the emissions of ozone precursors should be made in a combined global effort (or at least throughout the northern hemisphere). Emissions of ozone precursors have already been reduced in Europe and North America and are expected to decrease further here. However, this decrease has so far been partially counteracted by increases elsewhere. Other regions, such as East Asia, are currently facing severe air pollution problems. Part of the remedy for the elevated European ozone levels may well be local and regional air pollution control to curb air pollution in these regions.

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Table 1. Annual mean measured and model calculated CO in ppb for the European CO GAW sites downloaded from http://ds.data.jma.go.jp/gmd/wdcgg/. See also auxiliary material for figures. The comparison is based on monthly average model and measured data. Model IFS2 is IFS_v2, EMEP is EMEP rv4.8, ADJO is GEOSCHEMADJOINT, CAMC is CAMchem, OSLO is OsloCTM3_v2, GFDL is GFDL_AM3 and CHAS are the CHASER models (CHASER_t106/CHASER_re1). **Bold face**/*italic* numbers represent the model calculated concentration highest/lowest model bias/correlation.

Site:	Obs.	IFS2	EMEP	ADJO	CAMC	OSLO	GFDL	CHAS	IFS2	EMEP	ADJO	CAMc	OSLO	GFDL	CHAS
Mountain sites															
Summit	121	103	**109**	87	85	*75*	84	87/88	0.92	*0.89*	0.94	0.91	0.93	0.91	0.91/**0.96**
Zugspitze	153	172	133	**146**	134	168	*130*	133/130	0.61	0.57	0.62	0.45	**0.65**	*0.25*	0.57/0.51
Hohenpeissenb	76	200	151	146	134	**168**	*130*	133/137	**0.96**	**0.96**	0.95	0.96	0.86	*0.83*	0.97/**0.99**
Jungfraujoch	131	168	141	135	124	*185*	**130**	124/138	0.65	**0.90**	0.65	0.69	*0.33*	0.70	0.74/0.73
Rigi	181	*242*	138	135	124	**185**	130	126/138	0.76	**0.95**	0.87	0.94	*0.64*	0.88	0.86/0.93
West and central Europe															
Heimaey	123	**118**	108	90	88	*77*	84	86/89	*0.41*	**0.95**	0.92	0.82	0.92	0.88	0.88/0.94
Mace Head	120	109	**110**	93	90	*78*	88	91/92	0.90	**0.96**	0.88	0.87	0.92	*0.83*	*0.83*/0.89
Kollumerward	93	158	137	123	118	**172**	*111*	131/115	**0.96**	0.86	0.94	0.90	*0.65*	0.94	0.93/0.89
Neuglobsow	184	**151**	136	127	*118*	127	121	127/*118*	**0.98**	*0.81*	0.96	0.91	0.88	0.95	0.88/0.82
Ochsenkopf	147	*164*	142	**150**	133	131	134	144/137	0.53	**0.78**	*0.43*	0.47	0.58	0.45	0.66/0.62
Payern	216	**179**	149	135	*124*	131	130	127/127	0.91	0.85	**0.96**	0.81	0.78	*0.61*	0.90/0.83
Schauinsland	157	*212*	**156**	147	136	152	152	142/153	0.77	**0.96**	0.83	0.88	0.80	*0.75*	0.93/0.89
Northern Europe															
Pallas	131	111	**114**	99	94	*78*	86	95/87	0.93	0.91	0.94	0.89	0.95	*0.80*	0.92/**0.96**
Zeppelinfjelle	25	104	**111**	91	88	*77*	86	84/86	**0.94**	*0.87*	0.93	*0.87*	**0.94**	0.93	0.90/**0.94**
South and Eastern Europe															
Hegyhatsal	212	**164**	141	132	126	*120*	138	134/123	**0.91**	0.72	0.88	0.73	0.77	0.79	0.85/*0.71*
Krvavec	153	*218*	**148**	139	138	125	135	138/120	0.88	**0.96**	0.85	0.82	0.93	*0.80*	0.92/0.94
Lampedusa	128	**112**	108	95	104	93	*91*	101/101	0.82	**0.94**	0.86	*0.53*	0.68	0.66	0.85/0.91
Izana	104	95	**96**	80	79	*75*	79	85/85	0.89	**0.98**	0.91	0.90	0.77	0.84	*0.71*/0.83

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Table 2. Annual RERER values for the European sub-regions shown in Figure 1 for the CO tracer, CO and O_3.

Model	Europe	NW Europe	SW Europe	E Europe	SE Europe
			CO50 tracer		
EMEP_rv48	0.48	0.49	0.49	0.40	0.60
IFS v2	0.41	0.43	0.39	0.35	0.55
			CO		
EMEP_rv48	0.64	0.68	0.61	0.57	0.71
IFS v2	0.51	0.55	0.47	0.44	0.60
CHASER re1	0.52	0.53	0.53	0.45	0.64
CHASER t106	0.50	0.52	0.50	0.43	0.62
OsloCTM3_v2	0.44	0.49	0.43	0.36	0.53
CAMchem	0.54	0.57	0.55	0.46	0.62
GEOSCHEMADJ	0.41	0.43	0.24	0.35	0.56
GFDL-AM3	0.51	0.54	0.49	0.53	0.60
model mean	0.51	0.54	0.48	0.45	0.61
			Ozone		
EMEP_rv48	0.87	1.01	0.80	0.81	0.76
IFS v2	1.12	1.38	1.04	1.10	0.83
CHASER re1	0.63	0.71	0.56	0.57	0.64
CHASER t106	0.64	0.74	0.56	0.58	0.63
OsloCTM3_v2	0.89	1.06	0.80	0.91	0.71
CAMchem	1.02	1.38	0.87	1.09	0.71
GEOSCHEMADJ	1.04	1.59	0.86	1.06	0.68
GFDL-AM3	0.94	1.14	0.82	0.94	0.75
model mean	0.89	1.13	0.79	0.88	0.71

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Table 3. 18 European CO sites, 113 European ozone sites. Models to measurements bias in percent. RERER: deviation from model average in percent. Percentage deviations more than 15% preceded by large +/- signs.

		Concentration				**RERER**	
		CO		O_3			
Model	CO Tr.?	bias	Corr.	bias	corr.	CO	O_3
EMEP_rv48	yes	-16	0.87	+ 18	0.75	+ 25	-2
IFS v2	yes	1	0.82	-18	0.66	0	+26
OsloCTM3_v2	no	-19	0.82	+ 22	0.59	-14	0
CHASER re1	no	- 24	0.80	10	0.66	2	- 29
CAMchem	no	- 25	0.80	22	0.73	6	15
GEOSCHEMADJ	no	- 22	0.85	14	0.69	- 20	17
GFDL-AM3	partially	-13	0.77			0	6

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Figure 1. HTAP2 regions. NW – Western Europe north of the Alps. SW – western Europe south of the Alps. E – eastern Europe. Gr + Tu – Greece and Turkey.

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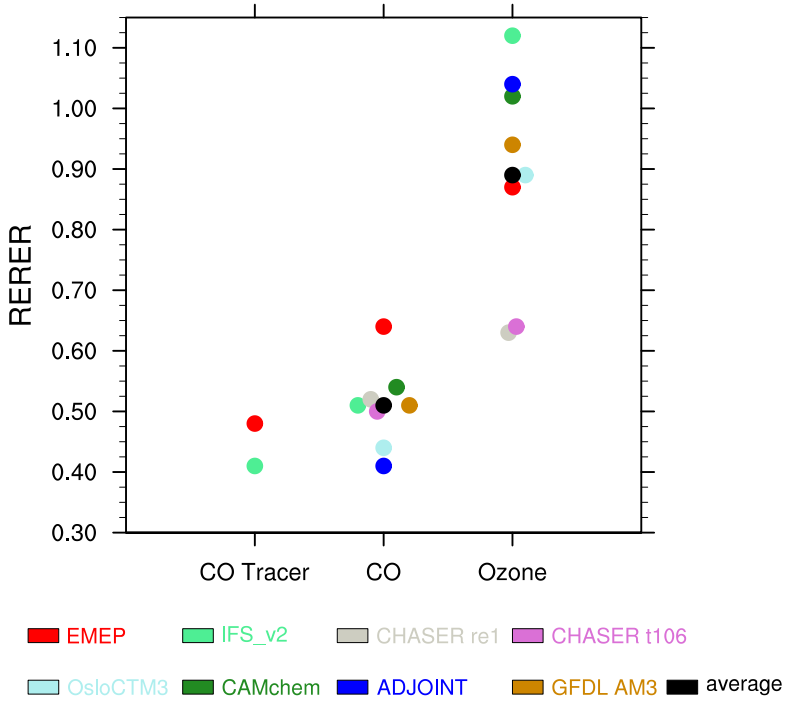

Figure 2. Model calculated annual CO tracer, CO and ozone RERER (Response to Extra-Regional Emission Reductions) values for Europe calculated by the models, see equation in section 4. Similar RERER values have been displaced horizontally.

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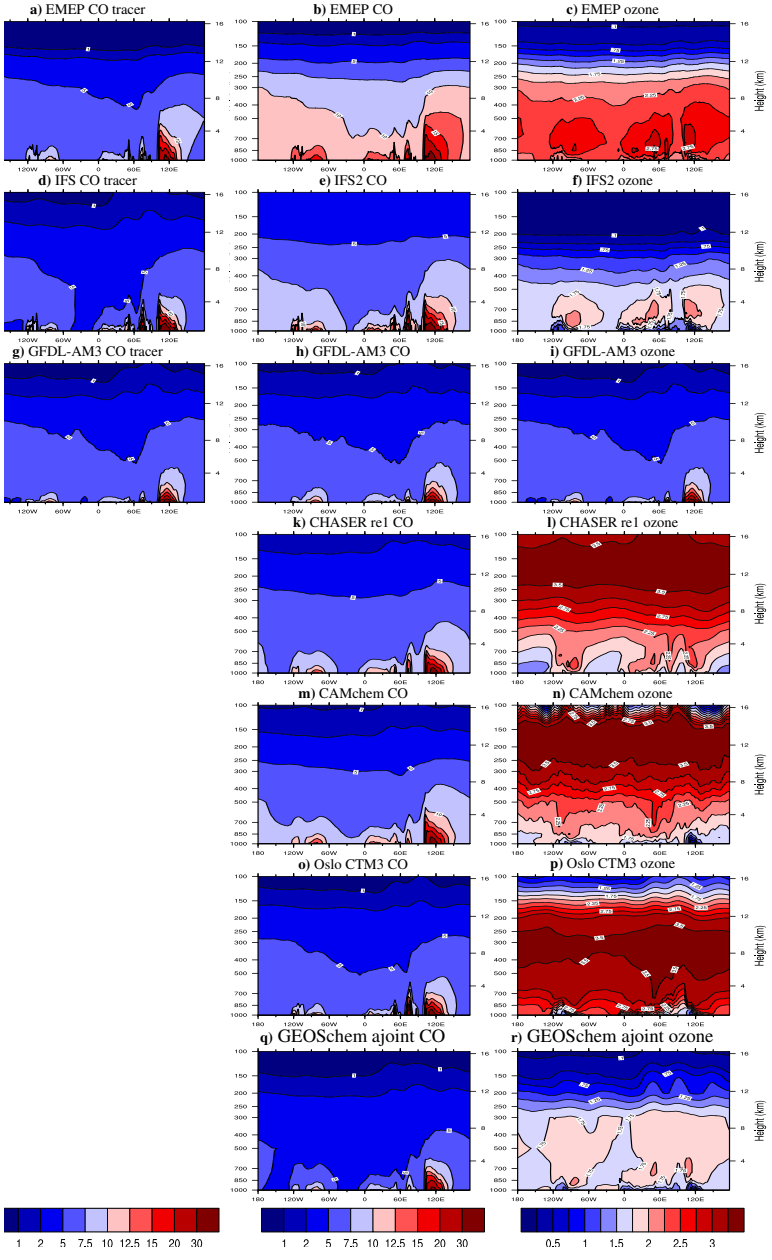

Figure 3. 20% of the anthropogenic (BASE – GLOALL) contributions to co50 tracer (a,d,g), CO (b,eh) and O_3 (c,ef) in ppb zonally averaged between 30 and 60 deg. N. The models have been interpolated to a common vertical grid.

Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-79
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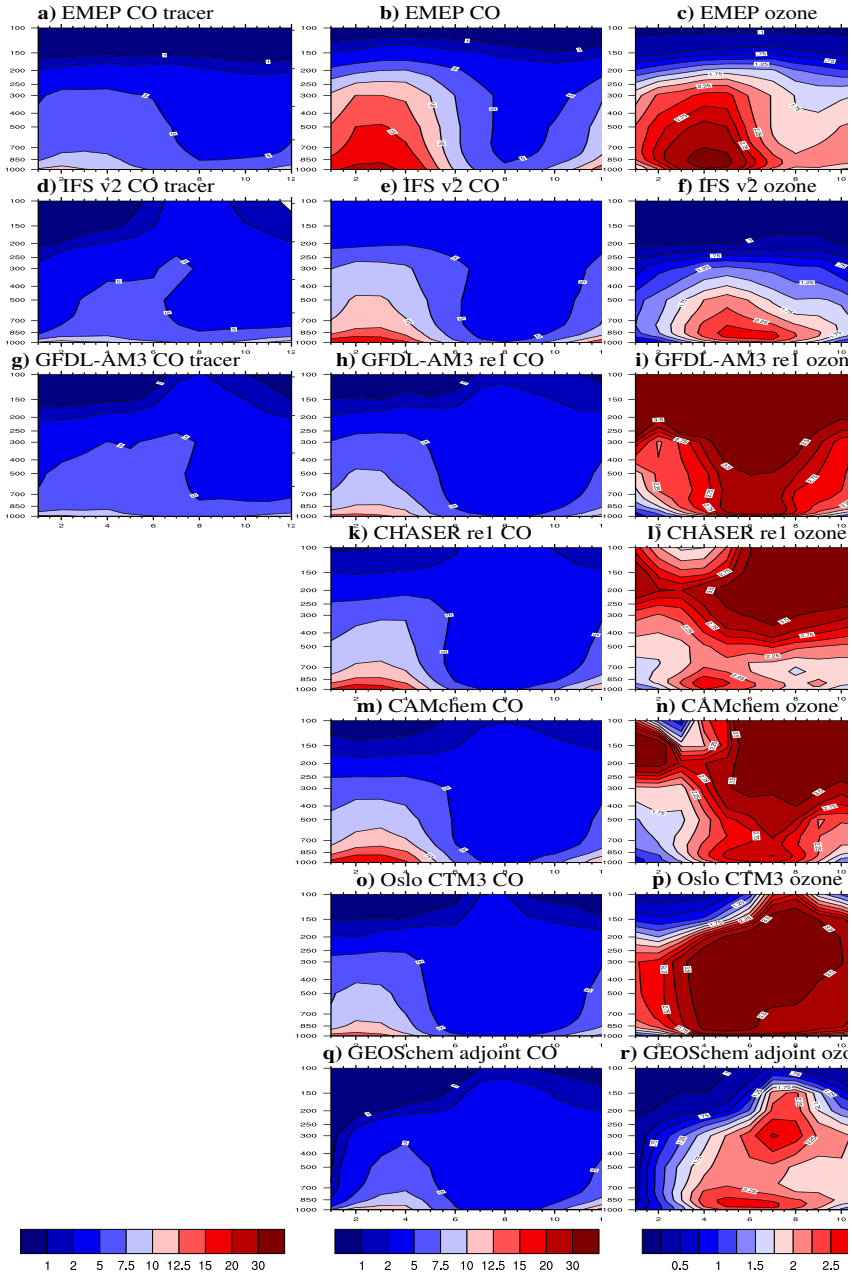

Figure 4. Monthly contributions from the 20% (BASE – GLOALL) perturbations of the anthropogenic emissions to co50 tracer (a,d,g), CO (b,eh) and O_3 (c,ef) in ppb averaged for the area bounded by 10°W to 50°E and 25 to 65 °N. The models have been interpolated to a common vertical grid.

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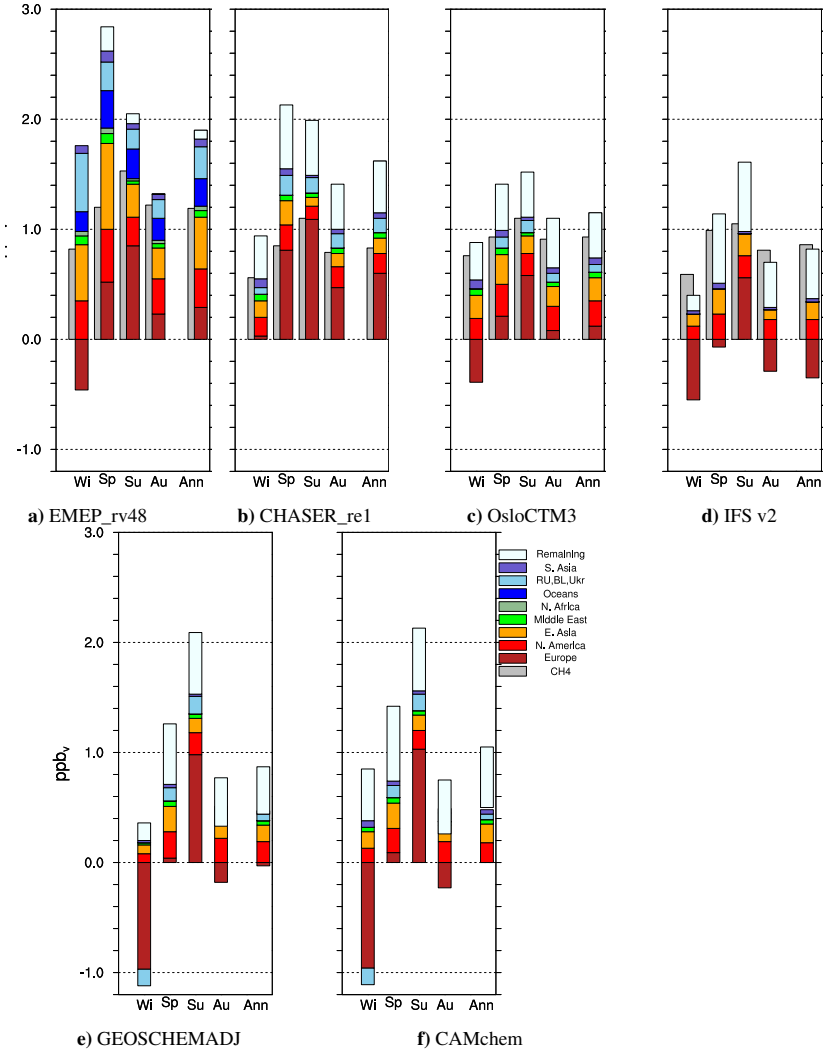

Figure 5. Contributions to European ozone levels (in ppb) from different world regions. Note that the separate contribution from North Africa (NAFALL) and ocean shipping (OCNALL) is only included in the EMEP_rv48 model calculations. The Middle East (MDEALL) and Russia, Belorussia and Ukaine (RBUALL) is not included in the IFS v2 model. For all models contributions from missing regions are included as "remaining". For the four top row models the effects of a 20% increase in CH_4 is shown as a separate bar.

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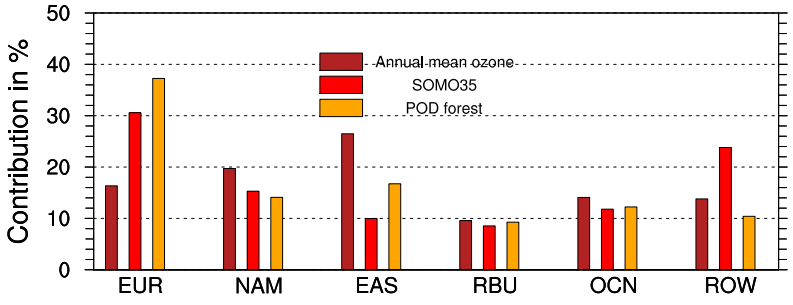

Figure 6. Contributions to ozone metrics annual mean ozone, SOMO35 and POD_1 forest in percent as calculated by the EMEP model. The metrics have been scaled so that the difference between the the BASE - GLOALL calculations is 100% (the sum of EUR, NAM, EAS, RBU, OCN and ROW adds up to 100%).