Dear editor,

thank you very much for guiding the editorial process.

According to the referee comments we thoroughly revised our manuscript. The most important changes are:

- We revised Section 2 by adding a Section 2.1 describing the source apportionment method and a Section 2.2 which describes and motivates the performed simulations in more detail.
- We added a Section 3 with a brief model evaluation focusing on ozone
- We rephrased the abstract and the conclusion to highlight important findings from our study for other modelling communities

Further, we rephrased different parts of the manuscript, including a better description of MECO(n) and a more detailed discussion about potential reasons for the differences of the results from EMAC and CM50. We have the feeling that some of the comments from referee#2 are based on a miss-understanding of the concept of MECO(n). Therefore, we tried to clarify this in the replies and the revised manuscript.

Attached are the comments to the two referees (original comments in italic, answers in normal fonts, changes in the manuscript in bold) together with the revised manuscript. In the revised manuscript all modifications are highlighted (latexdiff).

We are looking forward to your reply,

Mariano Mertens
(on behalf of all co-authors)

Dear referee#1,
thank you very much for your review of our manuscript GMD-2019-07. After our short comment we would like to reply to your review in detail. In the following, referee comments are given in italics, our replies in normal font, and text passages which we included in the text are in bold.

This paper presents an analysis of simulations at various horizontal resolutions, with emissions at various resolutions, and different emissions inventories.
Reply: To be more precise, our analysis focuses on diagnosed ozone contributions and uncertainties of these contributions, which arise due to model limitations (e.g. resolution, parametrisations), limited resolution of emission inventories, and uncertainties of the emission inventories. To make this more clear we revised the manuscript at several points (see below) and add also an addition Section (Sect. 2.1) which discuss the source apportionment in more detail.

The research is technically sound, and the application of source tagging and attribution is well illustrated. However, the paper does not seem to have any new results. The models and tagging technique used have all been published previously. The majority of their conclusions confirm previous work. Their strongest conclusion seems to be that different emissions inventories making the largest difference in ozone simulations, which I think is well known, but they do not offer any assessment about which might be more accurate. If the authors feel they have more compelling results, then they should make them much clearer.

Reply: First of all thank you very much for honouring our work. Indeed our analysis is very technical and focuses on the impact of technical limitations of models on the results of source apportionment diagnostics. However, we do not agree with referee#2 that our manuscript does not show any new results. Clearly, the dependence of simulated ozone concentrations on the resolutions of model and emissions are well known (see p1l4f, p2l8ff of our manuscript), and where appropriate we cite previous literature. The focus of our manuscript, however, is not on simulated ozone concentrations but on diagnosed contributions to ozone. We are not aware of any previous publication, which investigates the impact of these factors on the results of a source apportionment (e.g. tagging) method. Further, we are not aware of any similar model system allowing for such an analysis, as it requires a consistent global-regional model chain applying the identical source-attribution method on the global and regional scale. Previous publications applying source attribution on the regional scale (e.g. Dunker et al., 2002; Li et al., 2012; Kwok et al., 2015; Valverde et al., 2016; Karamchandani et al., 2017) considered only the contributions as simulated by the regional model and are not able to attribute ozone transported from the stratosphere or across the lateral borders of the regional model domain to specific emission categories.

In addition, we would like to remark that publications in GMD are not primarily about presenting new scientific results. Publications in GMD are mainly to document model developments, document experimental set-ups of model simula-

tions, document evaluation of model systems, present model evaluation strategies and to present technical analyses of model systems. We think that our manuscript documents the influence of model and emission inventory resolutions on source attribution results. This is clearly important to asses source apportionment results and their related uncertainties, also for other model systems.

To make the importance of our study also for other modelling communities more clear, we revised especially the conclusion (and the abstract) as discussed below in more detail.

It is not apparent why the authors thought GMD was the best journal for this work. It does not seem to have any new model development, or even quantitative evaluation of the model.

Reply: The main goal of our manuscript is to analyse the impact of technical limitations (e.g. model and emission inventory resolutions and/or the applied model) on the simulated contributions to ozone. Accordingly, our research questions are rather technical and focus on the impact of differences due to model limitations and/or differences due to input data. This does not necessarily imply in new scientific results, but it yields certainly important new insights for other researchers in the same field. Therefore, we chose GMD instead of ACP as journal and choose "development and technical paper" as manuscript type. These type of manuscripts also includes: '[...] papers relating to technical aspects of running models and the reproducibility of results' (GMD website).

The paper reads very much like a technical report for MESSy users. For example, it would help the general reader if 'ONEMIS' was defined and explained on p.5.

Reply: Of course the paper should not read as a technical report to MESSy users. Even tough the specific results we discuss are only valid for the specific model system and set-up (as it is common for most model studies) the general conclusions (see next paragraph) are also important to other researchers using source apportionment methods in a variety of models (e.g. CMAQ, WRF). As discussed in the next paragraph, we revised the manuscript in such a way that the general findings, which are important for the whole community, will become more clear.
For the revised manuscript we have carefully checked the manuscript again and describe specifics of the MESSy world, which are not defined in detail. For your specific example of ONEMIS on p5l1f we write: 'Emissions of soil-NOx and biogenic isoprene (C5H8) are calculated by the MESSy submodel ONEMIS (Kerkweg et al, 2006), which uses the parametrisations of Yienger and Levy (1995) for soil-NOx, and Guenther et al. (1995) for C5H8.'

While I see no errors in this work, I feel significant revisions are required to

make it suitable for publication. The paper should emphasize new results, not the confirmation of previous results. It would also be valuable to include comparisons to observations, and perhaps then conclusions can be drawn as to how fine does model horizontal resolution need to be to reproduce observations, and to reproduce accurately physical phenomena (e.g., vertical transport) that affect ozone distributions.

Reply: First of all we would like to thank referee#1 that she/he generally confirms that our analysis does not have any errors. As mentioned above, we think that our study offers important new results, which are also important for communities outside the MESSy community. In particular, our research offers insights into uncertainties of diagnosed ozone contributions. These new results are:

- Diagnosed contributions of anthropogenic emissions are rather robust on the continental scale. Differences due to the applied model, model and emission inventory resolutions and anthropogenic emissions are 10 % at maximum.
- Uncertainties of contributions at ground level due to downward transport of ozone are rather large. We find differences of up to 30 % on the continental scale.
- On the regional scale differences in contributions of land transport emissions are rather large and can reach up to 20 % and more, due to different reasons. Therefore fine resolved models and fine resolved emission inventories are important for regional assessments of ozone source apportionment.
- Source attribution diagnostics are a valuable tool to better understand inter-model differences.

However, the comment from referee#1 also clearly shows that we did not clearly bring up these new results. Therefore,we have highlighted the most important findings in more detail in our conclusion (and the abstract). The changed conclusion reads:

Apart from many model specific findings of this study, its results have important implications for other modelling studies and modellers applying source apportionment methods. These implications are:

- **First, our study shows that average continental contributions of anthropogenic emissions are quite robust with respect to the used model and the used model resolution. This means that global models at coarse resolution can be used to perform ozone source apportionment in this global context.**
- **Second, our results also show that on the regional scale, the differences either caused by different models, but also by model resolution are much larger. These effects arise mainly near hotspot**

regions like the Po Valley or near major shipping routes in the Mediterranean Sea. However, especially in these areas, contribution analyses of anthropogenic emissions are very important and spurious effects, such as artificially increased ozone levels and contributions caused by the coarse resolution of models and or emission inventories should be avoided. Hence, for regional analyses fine resolved models and emission inventories are required.

- **Third, our results clearly indicate how large the spread between models with respect to STE is. The importance of stratospheric ozone, both in the global and the regional model, corroborates the necessity of tracing the contributions of stratospheric ozone to ground level ozone explicitly by the source apportionment methods. However, only few currently available methods used on the regional scale account for this process.**

Further, we agree with referee#1 that a detailed comparison with observations is very valuable. However, ozone contributions cannot be measured directly. Therefore, more complex evaluation strategies involving proxies, which can be measured, are needed. This, however, is beyond the scope of this manuscript, because we here focus on the influence of technical aspects and try to estimate uncertainties, which arise only due to technical limitations. In a follow up study we work on a more detailed analysis involving detailed observations of specific measurement campaigns, which are confronted with simulated mixing ratios and diagnosed contributions to further constrain uncertainties of source attribution results. However, as also referee#2 asked for a section on model evaluation we added Sect. 3, with a basic model evaluation section focusing on ozone. This evaluation clearly indicates, that the vertical mixing of CM50 is too strong and CM50 likely overestimates the contributions of stratospheric ozone at the surface.

p.4, l.17: 'to calculate' should be 'calculation of'
Fixed. Thanks!
p.4, l.31 and elsewhere: 'lighting' should be 'lightning'
Indeed. Thanks!
p.6, l.17: See -¿ Sea
Fixed. Thanks!
p.14, l.27+: use "%" instead of "percentage points"; also 'respectively' is unnecessary.
We removed the respectively, but we stay with the percentage points. The difference in percentage points are obvious from the figure. Calculating % from the percentage-points might lead to missunderstandings.
p.15, l.1: 'effect' -¿ 'affect'
Changed
p.15, l.12: 'to quantify' -¿ 'for quantifying'

Fixed.

We are looking forward to your reply,
Mariano Mertens
(on behalf of all co-authors)

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Dear referee#2 ,
thank you very much for your review of our manuscript GMD-2019-07. Please find our replys to your comments below. In the following, referee comments are given in italics, our replies are in normal font, and text passages which we included in the text, are in bold.

This manuscript explores whether the source apportionment of surface ozone would be affected by model resolution. It performed model simulations with the different resolution of the model itself and the emission inventories. The difference in the source apportionment using a self-consistent tagging method is attributed to the model resolution and emission inventory resolution. The topic itself and the self-consistent tagging method are interesting.

Reply: We thank referee#2 for this summary and honouring our work with the self-consitent tagging method.

However, the analyses presented in the manuscript are too useful;
Reply: We do not understand this comment.

the discussion and conclusions are not insightful (or not having any new results as pointed out by Anonymous Reviewer#1). I'd suggest the following items to improve the manuscript.
Reply: First of all we thank referee#2 for the ideas on how to improve the manuscript which we comment in detail below. As already discussed in our reply the referee#1 we think that we provide new results, because, at least to our knowledge, the impact of the model resolution (and other technical factors) on the results of source apportionment methods has not been investigated in detail. Such an investigation, however, is important for two reasons:

- To investigate how robust the source apportionment results from global models are on the regional scale, and
- to estimate the range of uncertainties of source apportionment caused only by technical limitations of the models and emission inventories.

Even tough our results are only valid for a specific model, they provide new insights about possible ranges on model caused uncertainties. Such results are important for the community involved in source apportionment methods, both on the global and the regional scale.

Finally, we would like to remark that publications in GMD are not primarily about presenting new scientific results. Publications in GMD are mainly to document model developments, document experimental set-ups of model simulations, document evaluation of model systems, present model evaluation strategies and to present technical analyses of model systems. We think that our manuscript documents the influence of model and emission inventory resolutions on source attribution results. This is clearly important to asses source

apportionment results and their related uncertainties, also for other model systems.

To underline the importance of our findings to other modelling communities we largely rewrote the conclusion section. The new part reads:

Apart from many model specific findings of this study, its results have important implications for other modelling studies and modellers applying source apportionment methods. These implications are:

- **First, our study shows that average continental contributions of anthropogenic emissions are quite robust with respect to the used model and the used model resolution. This means that global models at coarse resolution can be used to perform ozone source apportionment in this global context.**
- **Second, our results also show that on the regional scale, the differences either caused by different models, but also by model resolution are much larger. These effects arise mainly near hotspot regions like the Po Valley or near major shipping routes in the Mediterranean Sea. However, especially in these areas, contribution analyses of anthropogenic emissions are very important and spurious effects, such as artificially increased ozone levels and contributions caused by the coarse resolution of models and or emission inventories should be avoided. Hence, for regional analyses fine resolved models and emission inventories are required.**
- **Third, our results clearly indicate how large the spread between models with respect to STE is. The importance of stratospheric ozone, both in the global and the regional model, corroborates the necessity of tracing the contributions of stratospheric ozone to ground level ozone explicitly by the source apportionment methods. However, only few currently available methods used on the regional scale account for this process.**

1. better defining the differences between simulations/models, be specific about what processes causing the variations in source apportionment. Here are just a few examples to improve.
Reply: As discussed in detail below we think that we are discussing a lot of processes causing these variations in detail. We agree that some explanations could be improved (see below). To better define the model and simulation differences we revised the manuscript accordingly and added the Section 2.2 in which we discuss the different simulations and the motivation for performing these simulation in more detail.

(a) Meteorological inputs such as temperature and light are different in some simulations, which would result in different biogenic emissions in methods of

'on-line calculated' and 'calculated by EMAC.'

Reply: No: As stated on p5l32ff of the original manuscript MECO(n) is applied in the so called quasi-chemistry transport model mode (QCTM-mode). In this mode the coupling between chemistry and dynamics is disconnected and each model instance simulated the same meteorology in all simulations. Of course, the dynamics differs between the different model instances due to different resolutions and/or physical parametrizations, which leads to differences in the biogenic emissions. We have discussed this issue on p3l4ff (of the original manuscript). For this reason the simulation *EBIO* is performed to investigate the impact of different biogenic emissions.

We added a note about QCTM in Sect. 2.2:
In this mode chemistry and dynamics are decoupled to increase the signal-to-noise ratio for small chemical perturbations. **This means, that even tough the emissions differ in the different simulation each model instance (EMAC, CM50 and CM12) simulated the same dynamics in all simulations. The dynamics between EMAC, CM50 and CM12, however, differs due to different resolution and physical parameterisations.**

Further, we added a longer description on the motivation of the *EBIO* simulation:
In the simulation *EBIO* the biogenic C_5H_8 and soil-NO_x emission as calculated by EMAC are transformed down and applied in CM50. By comparing the results from CM50 of *REF* and *EBIO* the effect of the different biogenic emissions can be analysed. These differences of the biogenic emissions are due to differences in the simulated meteorology between EMAC and CM50.

(b) Same anthropogenic emissions in different resolution might result in the same total emission but large regional differences. How do these emissions differ?

Reply: The coarse resolution of the emissions leads to a dilution of emissions over larger areas. Please see Fig S1 showing the MACCity land transport emissions in EMAC and in CM50. This figure is also added to the revised Supplement. Further, we added tables with the total emissions of the different simulations to the Supplement (Table S2-S10 in the new Supplement). To investigate the impact of the emission resolution onto the results the simulation *ET42* was performed.

(c) What are actually causing the differences in STE flux in the coarse vs. fine resolution model? Could it be related to on-line vs. off-line meteorology/convections and/or temporal and horizontal averaging of meteorological inputs (just some examples I am familiar with, like in Yu et al. (2018) and Hu et al. (2017); certainly, many other literature on this topic are available)? The

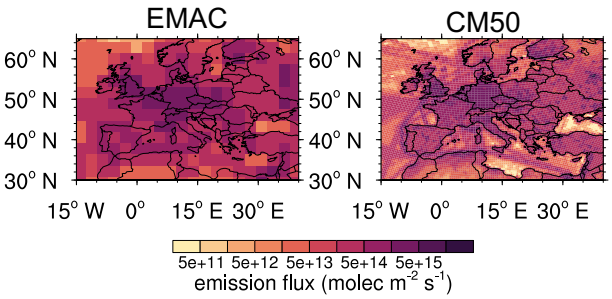

Figure S1: Annual averaged emissions flux (in molec m^{-2} s^{-1}) of NOx due to all anthropogenic emission sources (land transport, anthropogenic non-traffic, shipping; *REF* simulation) for EMAC and CM50.

contribution from downward transport seems to be the largest differences among models, and it should be quite interesting to explore.

Reply: Indeed, the largest differences between EMAC and CM50 are the differences of the STE. EMAC and COSMO/MESSy are chemistry-climate models, no chemistry transport models. Hofmann et al. (2016) already investigated in detail differences of the STE between EMAC and COSMO/MESSy, therefore we don't want to discuss this topic in detail again. Generally, the finer resolution of COSMO/MESSy leads to a better representation of the physical processes of individual STE events. However, in our manuscript we do not focus on individual events but rather on multi-year average values. For these multi-year average values the increased contribution of ozone from stratospheric origin is mainly confined to the planetary boundary layer. The reason for this is more efficient vertical mixing in COSMO/MESSy, partly caused by more vigorous convection and by an too unstable boundary layer during night. Taking the biases compared to observations into account this vertical mixing in COSMO/MESSy seems to be too strong, which indicates that the larger contribution of stratospheric ozone (and also for the categories aviation, lightning and N2O) is an artefact of this too strong vertical mixing. As discussed below we added a new Sect. 3 including a model evaluation to the manuscript. Further, we discuss the reason for the STE difference in more detail in the revised manuscript (see various changes in Sect. 4)

(d) It looks like the total lightning NOx emissions are the same across simulations, do their also have the same 3D distribution?
Reply: Yes. Over all simulations and over all model instances the same lightning-NO_x emissions are calculated. These are the emissions calculated by EMAC which are transformed during runtime from the EMAC grid onto the grid of CM50/CM12. The procedure is described in the model description section, but we rephrased the description to make it more clear. The new sentence is:

The lightning NO_x emissions are calculated only in EMAC using a

parametrization based on Price and Rind (1992), which is scaled to a global nitrogen oxide emission rate of $\approx$ 5 Tg(N) a^{-1} from flashes. In CM50 and CM12 we use the emissions from EMAC (i.e. with same geographical, vertical and temporal distribution), which are transformed on-line onto the grids of CM50 and CM12, respectively.

'Inter-model differences' should be better defined and documented and can provide insights on the calculated contributions. Specific discussion of these processes rather than vaguely saying because of the resolution would make this paper more useful.

Reply: The manuscript is about discussing the processes and other possible explanations for the differences between the different model results. Some examples are (page and line number refer to the original manuscript):

- p9l5ff [..] Due to increased vertical mixing in CM50 compared to EMAC ozone which is produced in the upper troposphere is transported downward more efficiently. [..]
- p10l12ff [..] mainly caused by a decreased net ozone production and a stronger vertical mixing in CM50 compared to EMAC. [..]
- p11l3ff [..] As the analyses of the *ET42* simulation results shows, the coarse resolution leads to an artificial increase of P_O3 which in turn leads to an increase [..]
- p11l10ff [..] On the coarse EMAC grid most parts of Southern Italy are considered as sea, affecting especially the calculation of dry deposition in EMAC, as dry deposition of ozone is lower over sea as over land.[..]
- p11l24ff [..] Especially in Southern Germany this is mainly caused by the better resolved topography and larger contributions of stratospheric ozone [..]
- p11l30ff [..] in Western Germany CM12 simulates a larger contribution of the CH_4-category to ozone compared to CM50, which is consistent with the larger tropospheric oxidation capacity in CM12 compared to CM50 (Mertens et al., 2016). [..]

We used the term 'inter-model differences' in some parts of the original manuscript to refer to the differences which we discussed before. In some parts we also referred to previous findings of Mertens et al. (2016). We rephrased these parts to be more precise. As an example we added the following note in the newly added Sect. 2.2:
Differences between the results of the EMAC and CM50 (and CM12) can be attributed to different effects: First, the dynamical core and physical parametrizations between EMAC and COSMO/MESSy differ, second the resolution of these models differs and third EMAC

and COSMO/MESSy calculate different soil-NO_x and biogenic C_5H_8 emissions. The latter due to the meteorology dependence and due to different soil types in EMAC and COSMO/MESSy.

Similarly, we added more detailed explanations in Sect. 3 and Sect. 4: An example from Sect. 3:
As already noted by Mertens et al. (2016), CM50 exhibits a larger positive ozone bias compared to EMAC. This bias is mainly caused by a more efficient vertical mixing in COSMO, as well as by a less stable boundary layer during night. The latter is a common problem of many models leading to diurnal cycles with too large ozone values during night, which results in an overall ozone bias (e.g. Travis and Jacob, 2019). The coarser resolution of the emissions (*ET42*) as well as the different biogenic emissions (*EBIO*) between EMAC and CM50 contribute only partly to the bias of CM50 compared to EMAC. The CM50 ozone bias is larger in *ET42* and *EBIO*. The pattern of the ground level ozone mixing ratio bias of CM50 compared to EMAC is similar for all simulations (see Fig. 3). Generally, CM50 has a positive ozone bias compared to EMAC over most parts of Europe.

Further, we add some more discussion about the differences of the stratospheric contribution between CM50 and CM12. These differences can mainly be attributed to stronger vertical mixing caused by stronger updraft and downdraft massfluxes in CM12 compared to CM50.

2. the terms used in the manuscript are very confusing for readers from outside the MESSy model community, particularly when referring to the specific simulation. For example, CM50 is used to compare with EMAC, while one refers to the resolution of 50km of one model; the other refers to a different model. ET42 refers to 'the MACCity emissions are transformed to the coarse grid of EMAC (T42), to investigate the impact of the resolution of the emission inventory.', but it sounds like it is done by the COSMO model only, so do all the REF, EBIO, EVEU simulations. Table 2 seems to suggest that EMAC also has those four simulations. Table 1 is not useful in the context of this manuscript but just adds confusions by adding a bunch of acronyms. This manuscript should not be 'read very much like a technical report for MESSy users' as pointed by the other reviewer. Readability should be improved.

Reply: We have th feeling that some of the confusion is caused by a missunderstanding of the concept of the MECO(n) model system. However, as the concept is explained in detail in a series of 5 different papers cited in Sect. 2 we wanted to recap only the basic concept of MECO(n). Obviously this basic recap was too short. Therefore we added a slightly longer description of MECO(n). This new part reads:
We apply the MECO(n) model system, which couples the global chemistry-climate model EMAC during runtime (i.e. on-line) with

the regional chemistry-climate model COSMO-CLM/MESSy (Kerkweg and Jöckel, 2012b). Both models, EMAC and COSMO-CLM/MESSy, calculate the physical and chemical processes in the atmosphere and their interactions with oceans, land and human influences. They use the second version of the Modular Earth Submodel System (MESSy2) to link multi-institutional computer codes (Jöckel et al., 2010). The core atmospheric model of EMAC is the 5th generation European Centre Hamburg general circulation model (ECHAM5, Roeckner et al., 2006). The core atmospheric model of COSMO-CLM/MESSy is the COSMO-CLM model (Rockel et al., 2008), a regional atmospheric climate model jointly further developed by the CLM-Community based on the COSMO model. In the model systems acronym 'n' denotes the number of COSMO-CLM/MESSy instances nested into the global model framework. The initial and boundary conditions, which are required for each of these nested regional model instances, are provided by the next coarser resolved model instance. This model instance can either be EMAC or COSMO-CLM/MESSy. Due to the on-line coupling the boundary conditions for the regional model instances can be provided at every time step of the driving model instance. This especially important to resolve short term variations of chemically active species. As EMAC and COSMO-CLM/MESSy calculate both, atmospheric dynamics and composition, the meteorological and chemical boundary conditions are as consistent as possible. In addition, the same chemical solver and kinetic mechanism is applied, leading to highly consistent chemical boundary conditions. Therefore, there is no need of lumping (i.e. treading different chemical species with similar chemical formula as one species), scaling boundary conditions for specific chemical species or taking boundary conditions from different models. More details about the MECO(n) model system are presented in a set of publications including a chemical and meteorological evaluation (Kerkweg and Jöckel, 2012a,b; Hofmann et al., 2012; Mertens et al., 2016; Kerkweg et al., 2018). The set-up of the simulation applied in the present study is very similar to that described by Mertens et al. (2016). Therefore, we present only the most important details of the model set-up. The complete namelist set-up is part of the Supplement.

It is important to understand that in every model simulation different instances of the MECO(n) model run at the **same** time and share necessary boundary and initial fields via MPI communication. For the applied MECO(2) set-up the running model instances are: EMAC, COSMO/MESSy with 50 km resolution (named COSMO(50km)/MESSy) and COSMO/MESSy with 12 km resolution (named (COSMO(12km)/MESSy)). These terms where introduced by Hofmann et al. (2012) and to ease readability the short terms CM50 and CM12 were introduced by Mertens et al. (2016). We don't want to add confusion by introducing new terms and therefore stick to these previously introduced abbre-

viations. We think that this is the best way of having a clearly defined model system. However, we are open for concrete suggestions for an improved naming.

To make clear thatin MECO(n) different model **instances** run at the same time (and we do not perform simulations with different models) we used the term troughout the revised manuscript.

Using the MECO(2) system (EMAC → CM50 → CM12) we performed different simulations (*REF*, *EVEU*, *EBIO* and *ET42*) and compare the results of all three model instances. To differentiate between model instances and simulation names, the simulation names are written in italics throughout the manuscript. As shown in Table 2, EMAC is running in all simulations, but with the same set-up including the same emissions. To make this more clear we added a new subsection called 'investigation concept' (Sect. 2.2).

With respect to Table 1 we do not agree with referee#2. We think this table is the best and shortest way of showing the model set-up, which is (to our opinion) very important in terms of reproducibility. We list the name of the individual submodels as they are published in peer reviewed literature under these names. For people not familiar with these submodels we have a short description, stating the physical/chemical process or the diagnostic provided by this submodel as well as a reference describing the individual submodel in detail.

3. this paper could benefit from a section of model evaluation by adding comparisons with observations. This way could suggest which simulations are 'in practice' better and if the model simulations are actually realistic.

Reply: We added a basic section of model evaluation and compare the performance of the individual model instances for the different simulations with observations (new Sect. 3). For this, we use ground based station measurements as well as ozone sonde measurements. This should give an impression of the overall model performance. However, from this model evaluation it is not possible to evaluate ozone contributions as these are pure model diagnostics.

4. the metrics used to quantify simulation difference: this manuscript mostly uses the average concentration of ozone and relative contribution of a specific source. These tend only to show minimal differences among simulations; even though the manuscript claims 'up to 20%' in the calculated contribution of transport emissions, the absolute amount is small. One way to improve is looking at the probability distribution of concentrations or contributions, which could be much more useful to examine differences in model chemical pathways and for specific air pollution episodes, i.e., examples like in Fiore et al. (2002) and Yu et al. (2016).

Reply: We do not fully agree with this comment. In Fig. 3 and Fig. 7 we show box-whisker plots which indicate the range of the simulated values (e.g.

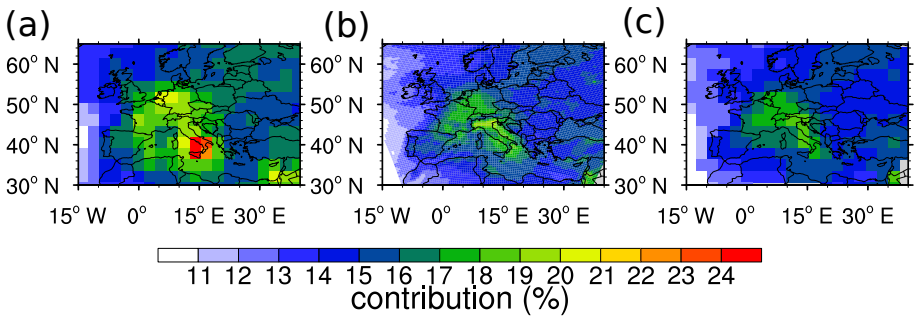

Figure S2: 95th percentile of the contribution of O_3^{tra} to ground level O_3 (for JJA between 9–18 UTC) for **(a)** EMAC, **(b)** CM50 and **(c)** CM50 transformed onto the EMAC grid ($CM50_E$).

range, 25th and 75th percentile, mean and median). Of course we could also have chosen PDFs instead, but they offer similar information. Further, we also discuss differences of the 95th percentile of the contributions of land transport. The differences of these extreme values are of course larger than the average differences.

We agree that the differences for these averaged values show only small differences, but the focus of your analysis is on time scales on which global models (e.g. multi-year averages) and not on the scale of individual pollution events. Therefore, we prefer to stick to the applied metrics. Further, we are not aware where we claim 'up to 20%' in the calculated contributions of transport emissions', as we claim a differences of up to 20 % between the simulated contribution of the different models/model set-ups. These differences are simulated around the Naples region, were the relative contributions between EMAC (17 %) and CM50 (13 %) differ. These relative contributions refer to absolute contributions of 3 to 4 nmol mol^{-1}. For the 95th percentile (see Fig. S2) of the relative contribution of O_3^{tra} these difference increase to around 6 percentage – points. We added this figure to the Supplement (Fig S6).

We are looking forward to your reply,

Mariano Mertens
(on behalf of all co-authors)

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Are contributions of emissions to ozone a matter of scale? - A study using MECO(n) (MESSy v2.50)

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~~This study investigates if ozone source apportionment results using a tagged tracer approach depend on the resolutions of the applied model and/or emission inventory. For this we apply a global to regional atmospheric chemistry model, which allows to compare the results on the global and regional scale. Our results show that differences on the continental scale (e.g. Europe) are rather small (10 %), on the regional scale, however, differences of up to 30 % were found.~~

Abstract. Anthropogenic and natural emissions influence the tropospheric ozone budget, thereby affecting air-quality and climate. To study the influence of different emission sources on the ozone budget, often source apportionment studies with a tagged tracer approach are performed. Studies investigating air quality issues usually rely on regional models with a high spatial resolution, while studies focusing on climate related questions often use coarsely resolved global models. It is well known that simulated ozone ~~concentrations~~ mixing ratios depend on the resolution of the model and the resolution of the emission inventory. Whether the contributions simulated by source apportionment approaches also depend on the model resolution, however, is still unclear. Therefore, this study ~~is a first attempt~~ firstly attempts to analyse the impact of the model, the model resolution, and the emission inventory resolution on simulated ~~ozone contributions~~ contributions to ozone diagnosed with a tagging method. The differences of the ~~ozone contributions~~ contributions to ozone caused by these factors are compared with differences which arise ~~due to~~ from the usage of different emission inventories. To do so we apply the MECO(n) model system which on-line couples a global chemistry-climate model with a regional chemistry-climate model equipped with a tagging scheme for source apportionment. The results of the global model (300 km horizontal resolution) are compared with the results of the regional model at 50 km (Europe) and 12 km (Germany) resolution. ~~Averaged over Europe the simulated contributions of land transport emissions to ground-level ozone differ by 10 % at maximum. For other anthropogenic emission sources the differences are in the same order of magnitude, while the contribution of stratospheric ozone to ground level ozone differs by up to 30 % on average. This suggests that ozone~~ Besides model specific differences and biases which are discussed in detail, our results have important implications for other modelling studies and modellers applying source apportionment methods: First, contributions of anthropogenic ~~emission sources averaged on~~ emissions averaged over the continental scale are ~~rather~~ quite robust with respect to ~~different models, model~~ the model, model resolution and emission inventory ~~resolutions. On regional scale , however, we quantified differences of the contribution of land transport emissions to ozone of up to 20 %. Depending on the region the largest differences are either caused by inter model differences, or differences of~~ resolution. Second, the

differences on the regional scale caused by different models and model resolutions can be quite large and regional models are indispensable for source apportionment studies on the sub-continental scale. Third, the difference of the contributions of ozone from the stratosphere transported to the ~~anthropogenic emission inventories. Clearly, the results strongly depend on the compared models and emission inventories and cannot necessarily be generalised, however we show how the inclusion of~~ surface strongly differs between the models, mainly caused by differences in the efficiency of the vertical mixing. As many models show a large differences in the downward transport of ozone to the surface, and this stratospheric ozone plays an important ole for ground-level ozone it is important that source apportionment methods ~~can help in analysing inter-model differences~~account for this source explicily.

1 Introduction

Emissions from land transport, industry or shipping contribute largely to global budgets of trace gases like NO_x and O_3, hereby impacting air-quality and climate (e.g., Eyring et al., 2007; Matthes et al., 2007; Hoor et al., 2009; Fiore et al., 2012; Young et al., 2013; Hendricks et al., 2017; Mertens et al., 2018). To quantify the impacts of these emissions, typically source-receptor relationships are calculated using ~~perturbations~~ perturbation or source apportionment methods (e.g., Dunker et al., 2002; Emmons et al., 2012; Stock et al., 2013; Matthias et al., 2016; Huang et al., 2017; Clappier et al., 2017; Butler et al., 2018). Many studies exist quantifying the influence of anthropogenic and natural emission sources (e.g. land transport emissions or lightning) on the ozone budget, but the uncertainties of such analyses are large. Three main sources of uncertainties exist: (1) the emission inventories, (2) model biases/errors, and (3) the resolutions of the models and/or emission inventories. The influences of the first two factors, emission inventories and model biases, have been investigated by multi-scenario and/or multi-model analyses (e.g., Eyring et al., 2007; Hoor et al., 2009; Fiore et al., 2009). ~~Although~~ Even tough, the influence of the model and emission inventory resolutions ~~onto simulated ozone~~ on simulated ozone mixing ratios is well known (e.g. Wild and Prather, 2006; Wild, 2007; Tie et al., 2010; Holmes et al., 2014; Markakis et al., 2015), the impact of the third factor - the model and emission inventory resolutions - on the simulated contributions of specific emission sources to ozone has not yet been systematically investigated in detail. ~~Such an investigation, however, is important~~ It is important to investigate this third factor, as source apportionment studies focusing on climate usually use rather coarsely resolved global climate models (e.g. Wang et al., 1998; Lelieveld and Dentener, 2000; Grewe, 2006; Matthes et al., 2007; Dahlmann et al., 2011; Emmons et al., 2012), while air quality related studies use finer resolved regional models (e.g. Dunker et al., 2002; Li et al., 2012; Kwok et al., 2015; Valverde et al., 2016; Karamchandani et al., 2017). Therefore it is unclear, ~~whether the results on the global and the regional scale~~ if the results from global and regional models are comparable and how large potential errors, caused by the coarse resolution of global models, are. The present study is a first attempt to investigate the influences of the model and of the emission inventory resolutions on the ozone contributions. In detail, we investigate the influences of four different aspects on source attribution results of ozone:

- the applied model,

- the resolution of the model,
- the resolution of the emission inventory, and
- the emission inventory.

We apply the MECO(n) (**M**ESSy-fied **E**CHAM and **CO**SMO models nested **n** times, e.g. Kerkweg and Jöckel, 2012b; Mertens et al., 2016) model system together with a detailed source apportionment ~~(tagging, Grewe et al., 2017) method~~ method (tagging, Grewe et al., 2017). This model system couples ~~on-line~~ during runtime the global chemistry-climate model EMAC (**E**CHAM5/**M**ESSy for **A**tmospheric **C**hemistry, Jöckel et al., 2006, 2010) with the regional chemistry-climate model COSMO/MESSy (Kerkweg and Jöckel, 2012a), which consists of the COSMO model (Consortium for Small-scale Modelling) equipped with the MESSy (**M**odular **E**arth **S**ubmodel **Sy**stem, Jöckel et al., 2005, 2010) infrastructure. Due to the MESSy infrastructure, we apply identical submodels for calculating the chemical processes as well as the ~~identical~~ same source apportionment method ~~(Grewe et al., 2017) at all nesting steps. Further~~in the global and regional model instances. In addition, the global model instance provides consistent boundary conditions for the source apportionment to the regional model instances, allowing a detailed intercomparison of the source apportionment results on different scales. Therefore, this model system is, to our knowledge, the first available model system allowing a seamless contribution analysis from global to regional scale. With this model chain we can directly compare the results ~~of the~~ at regional and global ~~model~~scale, which allows us to estimate uncertainties of the contribution analyses caused by the model, the model resolution and emission inventory resolution. ~~In addition, this model system is, to our knowledge, the first available model system allowing a seamless contribution analysis from global to regional scale.~~

This paper is organised as follows. First, Sect. 2 gives an overview of the model system~~followed by an analysis~~, discusses the the investigation strategy and the performed simulations. In Sect. 3 we present a brief evaluation of the model results against ground level and ozone sonde observations as well as a comparision of the ozone production rates simulated by EMAC and COSMO in Sect. 3.1. In Sect. 4 the differences in the ozone contributions caused by differences of model and emission inventory resolutions are analysed in detail. We provide a quantification of these differences caused by model and emission inventory resolutions in Sect. 5. ~~To set these numbers in to context, we compare these differences with those caused using different emission inventories.~~

2 Model description and experimental set-up

~~The~~ We apply the MECO(n) model system, which couples the global chemistry-climate model EMAC during runtime (i.e. online) with the regional chemistry-climate model COSMO-CLM/MESSy ~~(from now on COSMO/MESSy, Kerkweg and Jöckel, 2012a)~~ (Kerkweg and Jöckel, 2012b). Both models, EMAC and COSMO-CLM~~(COSMO model in Climate Mode) is the community model of the German regional climate research community~~ /MESSy, calculate the physical and chemical processes in the atmosphere and their interactions with oceans, land and human influences. They use the second version of the Modular Earth Submodel System (MESSy2) to link multi-institutional computer codes (Jöckel et al., 2010). The core atmospheric model of

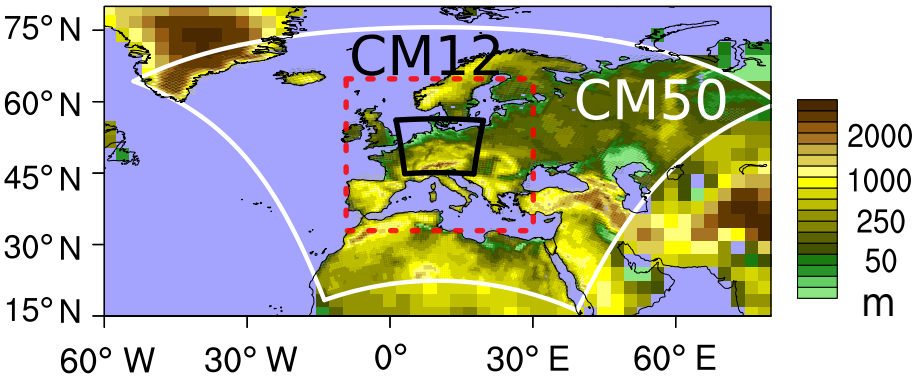

Figure 1. Domains of the CM50 (white line) and CM12 (black line) instances. Depicted is the topography of the continents (in m) at the resolution of the corresponding instance. Outside the CM50 domain the topography of EMAC is displayed. Shown is the entire computational domain including the relaxation area. The dashed red square indicates the region analysed in Sect. 4. Figure is ~~largely~~ reproduced from Mertens (2017).

EMAC is the 5th generation European Centre Hamburg general circulation model (ECHAM5, Roeckner et al., 2006). The core atmospheric model of COSMO-CLM/MESSy is the COSMO-CLM model (Rockel et al., 2008), a regional atmospheric climate model jointly further developed by the CLM-Community ~~(Rockel et al., 2008). The technical details of MECO(n), as well as meteorological and chemical evaluation~~ based on the COSMO model. In the model systems acronym 'n' denotes the number of COSMO-CLM/MESSy instances nested into the global model framework. The initial and boundary conditions, which are required for each of these nested regional model instances, are provided by the next coarser resolved model instance. This model instance can either be EMAC or COSMO-CLM/MESSy. Due to the on-line coupling the boundary conditions for the regional model instances can be provided at every time step of the driving model instance. This especially important to resolve short term variations of chemically active species. As EMAC and COSMO-CLM/MESSy calculate both, atmospheric dynamics and composition, the meteorological and chemical boundary conditions are as consistent as possible. In addition, the same chemical solver and kinetic mechanism is applied, leading to highly consistent chemical boundary conditions. Therefore, there is no need of lumping (i.e. treading different chemical species with similar chemical formula as one species), scaling boundary conditions for specific chemical species or taking boundary conditions from different models.

More details about the MECO(n) model system are presented in a set of publications ~~(Kerkweg and Jöckel, 2012a, b; Hofmann et al., 201~~ ~~. Further, the~~ including a chemical and meteorological evaluation (Kerkweg and Jöckel, 2012a, b; Hofmann et al., 2012; Mertens et al., 201 . The set-up of the simulation applied in the present study is very similar to that described by Mertens et al. (2016)~~, including an evaluation of atmospheric key constituents~~. Therefore, we present only the most important details of the model ~~system and the~~ set-up. The complete namelist set-up is part of the Supplement.

A MECO(2) set-up with one COSMO-CLM/MESSy (from now on COSMO/MESSy) instance over Europe with a resolution of 0.44° x 0.44° (~~hereafter named CM50 for COSMO(50km) /MESSy~~ around 50 km) and one instance covering Germany with

a resolution of 0.1° x 0.1° (~~hereafter named CM12 for COSMO(12km) /MESSy~~ around 12 km) was applied (see Fig. 1 for the computational domains). For simplicity, we name these two model instances hereafter CM50 and CM12. EMAC, CM50 and C12 are running simultaneously in the same way as in externally coupled earth system models the different earth compartment model run in parallel (see Fig. 2 in Mertens et al. (2016) for details of the data exchange between the nested model instances). Both COSMO/MESSy instances use 40 vertical model levels reaching up to a height of 22 km, the damping zone starts at 11 km height. The boundary conditions for CM50 ~~instance is driven~~ are provided by EMAC, which is operated at a resolution of T42L31ECMWF, i.e. with a spherical truncation of T42 (corresponding to a quadratic Gaussian grid of approx. 2.8° x 2.8° in latitude and longitude) with 31 hybrid pressure ~~level~~ levels in the vertical up to 10 hPa. The boundary conditions for ~~the~~ CM12 ~~instance~~ are provided by ~~the first COSMO/MESSy instance~~CM50. The applied MESSy version is a modified version of MESSy 2.50, including ECHAM 5.3.02 and COSMO ~~5.0.0~~5.00. All changes are included in MESSy 2.51. To facilitate a ~~one to one~~ one-to-one comparison with observations, EMAC is 'nudged' by Newtonian relaxation of temperature, divergence, vorticity and the logarithm of surface pressure (Jöckel et al., 2006) towards ERA-Interim (Dee et al., 2011) reanalysis data of the years 2007 to 2010. Sea surface temperature and sea ice coverage are prescribed as boundary conditions for the simulation set-up from this data source.

Due to the MESSy infrastructure the same ~~submodels (e.g.~~ diagnostics or chemical process descriptions ~~)~~ are applied in all model instances~~(see~~ . Following the modular structure of MESSy each diagnostic or process description is coded as a so-called submodel. The applied submodels are listed in Table 1~~for a list of the most important applied submodels)~~. Besides the name of the submodel and their reference a short description provides general information on the process or diagnostic represented by the respective submodel. Most importantly the identical ~~chemical~~ kinetic solver (MECCA, Sander et al., 2011) and the identical TAGGING submodel (Grewe et al., 2017) are applied. ~~The simulations are performed using the mechanism 'CCMI-base-01-tag.bat'. This mechanism includes~~ The chemical mechanism used by MECCA considers the chemistry of ozone, methane and odd nitrogen. While alkynes and aromatics are not considered, alkenes and alkanes are considered up to C_4. We use the Mainz Isoprene Mechanism (MIM1, Pöschl et al., 2000) for the chemistry of isoprene and some non-methane hydrocarbons (NMHCs). The mechanisms of MECCA ~~and SCAV (scavenging of traces gases by clouds and precipitation, Tost et al., 2006a,~~ as well as for the submodel calculating the scavenging of trace gases by clouds and precipitation (SCAV, Tost et al., 2006a, 2010) are part of the supplement. The TAGGING submodel ~~allows to calculate~~ calculates the contributions of different emission sources to ozone and the relevant precursors~~(e.g Mertens et al., 2018). At the lateral and top boundaries of the regional model instances, the tracers of the TAGGING submodel are treated in the same manner as all chemical tracers. Accordingly, the tagged tracers in COSMO/MESSy are relaxed towards the mixing ratios provided by EMAC (or the coarser resolved COSMO instance, respectively) at the lateral and top boundaries. In contrast to this , other tagging schemes, which are used in regional chemistry-climate or chemistry-transport models, usually feature no boundary conditions for the tagged tracers at the lateral (and top) boundaries (e.g. Li et al., 2012; Kwok et al., 2015; Valverde et al., 2016). Therefore, our approach allows for consistent zooming into the area of interest, including an apportionment of the contribution of emissions from different sources to ozone and its relevant precursors across the lateral and top boundaries of the regional model. Especially for chemical species with a long lifetime, such as ozone this is important as large parts of the ozone concentrations at a certain place are influenced~~

by long range transport or subsidence from the stratosphere. If the source apportionment is only performed in the regional model, long range transported ozone can not correctly be attributed to the emission sources themselves. More details of this tagging approach are given in Sect. 2.1.

The Lightninglightning NO_x emissions are calculated in EMAC only only in EMAC using a parametrization based on Price and Rind (1992), which is scaled to a global nitrogen oxide emission rate of ≈ 5 $Tg(N)\ a^{-1}$ from flashes. The calculated emissions are mapped to COSMO/MESSy and are subsequently emitted thereIn CM50 and CM12 we use the emissions from EMAC (i.e. with same geographical, vertical and temporal distribution), which are transformed on-line onto the grids of CM50 and CM12, respectively. This approach was chosen as the calculation of lighting-lightning-NOx is strongly coupled to the convection parametrisation (e.g. Tost et al., 2007). In different models and/or at different model resolutions convection occurs at different places and/or times and lightning emissions can differ largely. To Our approach was chosen to allow for an easier comparison between the results of different models and at different resolutions we used the same lightning-emissions. For studies with other scientific questions it would of course be desirable to calculate the lighting-emissions separately at every resolutionmodel instances.

Emissions of The calculation of emissions from soil-NO_x and biogenic isoprene (C_5H_8) are calculated is performed by the MESSy submodel ONEMIS (Kerkweg et al., 2006b), which uses the parametrisations of Yienger and Levy (1995) for soil-, and Guenther et al. (1995) for (described as ONLEM by Kerkweg et al., 2006b). Following the parametizations of Yienger and Levy (1995) and Guenther et al. (1995) the respective emissions depend on the meteorological conditions. In contrast to the lightning NO_x emissions, the soil-NO_x and biogenic emissions are calculated in EMAC and COSMO, respectivelythe COSMO instances separately. This leads to differences in the soil-NO_x and C_5H_8 emissions (see Fig. S5 S17 in the Supplement), influencing the calculation of the contributions. We have chosen this approach, because the land sea masks differ between models and model resolutions. If the emissions calculated by EMAC are simply emitted in the finer resolved used in the COSMO/MESSy model instances, some of the emissions would occur over sea (or vice versa). This could lead to artificial errors in the contribution analyses. In EMAC, the isoprene emissions calculated by ONEMIS are scaled with a factor of 0.6 (following Jöckel et al., 2006) and in COSMO with 0.45 (following Mertens et al., 2016).

For the present study

2.1 Tagging for source apportionment

For the source apportionment we apply the TAGGING submodel described by Grewe et al. (2017). The tagging method is a diagnostic method, i.e. the atmospheric chemistry calculations are not influenced. Due to constraints with respect to the computational resources (e.g., computing time and memory), the detailed chemistry from MECCA is mapped on a family concept, for which the tagging is performed. The tagged species are ozone, the family of NO_y, the family of NMHC, CO, PAN as well as OH and HO_2 in a steady state approach. The TAGGING submodel is applied in each model instance. At the lateral and top boundaries of CM50 and CM12 the tagged contributions are treated in the same manner as all chemical species, i.e. the mixing ratios of the tagged species are relaxed towards the mixing ratios provided by the driving model instance. This is depicted in Fig. 2, showing the relative contribution of the land transport emissions to ozone. EMAC calculates the contributions

Table 1. Overview of the submodels applied in EMAC and COSMO/MESSy, respectively. Both COSMO/MESSy instances use the same set of submodels. MMD* comprises the MMD2WAY submodel and the MMD library.

Submodel	EMAC	COSMO	short description	references
AEROPT	x		calculation of aerosol optical properties	Dietmüller et al. (2016)
AIRSEA	x	x	exchange of tracers between air and sea	Pozzer et al. (2006)
CH4	x		methane oxidation and feedback to hydrological cycle	
CLOUD	x		cloud parametrisation	Roeckner et al. (2006), Jöckel et al. (2006)
CLOUDOPT	x		cloud optical properties	Dietmüller et al. (2016)
CONVECT	x		convection parametrisation	Tost et al. (2006b)
CVTRANS	x	x	convective tracer transport	Tost et al. (2010)
DDEP	x	x	dry deposition of aerosols and tracer	Kerkweg et al. (2006a)
E2COSMO	x		additional ECHAM5 fields for COSMO coupling	Kerkweg and Jöckel (2012b)
GWAVE	x		parametrisation of non-orographic gravity waves	Roeckner et al. (2003)
JVAL	x	x	calculation of photolysis rates	Landgraf and Crutzen (1998), Jöckel et al. (2006)
LNOX	x		NO_x-production by lightning	Tost et al. (2007), Jöckel et al. (2010)
MECCA	x	x	tropospheric and stratospheric gas-phase chemistry (CCMI-base-01-tag.bat mechanism)	Sander et al. (2011), Jöckel et al. (2010)
MMD*	x	x	coupling of EMAC and COSMO/MESSy (including libraries and all submodels)	Kerkweg and Jöckel (2012b); Kerkweg et al. (2018)
MSBM	x	x	multiphase chemistry of the stratosphere	Jöckel et al. (2010)
OFFEMIS	x	x	prescribed emissions of trace gases and aerosols	Kerkweg et al. (2006b)
ONEMIS	x	x	on-line calculated emissions of trace gases and aerosols	Kerkweg et al. (2006b)
ORBIT	x	x	Earth orbit calculations	Dietmüller et al. (2016)
QBO	x		Newtonian relaxation of the quasi-biennial oscillation (QBO)	Giorgetta and Bengtsson (1999), Jöckel et al. (2006)
RAD	x		radiative transfer calculations	Dietmüller et al. (2016)
SCAV	x	x	wet deposition and scavenging of trace gases and aerosols	Tost et al. (2006a)
SEDI	x	x	sedimentation of aerosols	Kerkweg et al. (2006a)

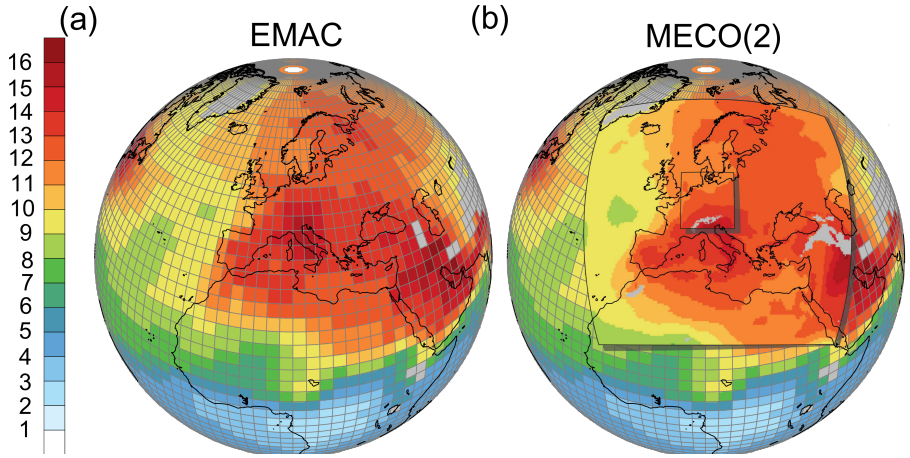

Figure 2. Relative contribution of land transport emissions to the ozone column up to 850 hPa (in %), averaged for July 2008; **(a)** the values calculated by the EMAC model and **(b)** the values calculated by MECO(2) with the two refinements covering Europe and Germany.

globally with a rather coarse resolution. With MECO(2) (Fig. 2b) the resolution over Europe and Germany is increased by the two COSMO/MESSy refinements. As the source apportionment is performed in EMAC, CM50, and CM12 - with the respective boundary conditions provided by the coarser resolved model instance - this approach allows for a consistent zooming into the area of interest within the global framework. In contrast to our approach, other tagging methods which are usually applied in regional chemistry-climate or chemistry-transport models feature no boundary conditions for the diagnosed contributions (i.e. tagged tracers) at the lateral (and top) boundaries of the regional model domain (e.g. Li et al., 2012; Kwok et al., 2015; Valverde et al., 2016). Therefore, these approaches have special categories tagging the contributions from lateral and/or top boundaries. In these cases long range transported ozone (or other species) cannot be correctly attributed to the emission sources themselves. Therefore, our approach allows for a consistent zooming into the area of interest, including an apportionment of the contribution of emissions from different sources to ozone and its relevant precursors across the lateral and top boundaries of the regional model instances. Especially for chemical species with a long lifetime, such as ozone, this is important as large parts of the ozone mixing ratios at a certain place are influenced by long range transport or subsidence from the stratosphere. It is important to note that this method is a classical down-scaling method and no grid-refinement technique, which means with MECO(2) for instance over Germany we calculate the contributions three times, once in each model instance (EMAC, CM50 and CM12). These results can be compared to investigate the influence of the model resolution on the results.

2.2 Analysis concept and performed model simulations

The goal of our study is to investigate how diagnosed contributions of different emissions to ozone in Europe are influenced by model uncertainties such as:

- the applied model,

- the resolution of the model,
- the resolution of the emission inventory, and
- the emission inventory.

For this analysis, four different ~~simulations were~~ MECO(2) simulations are performed which are named *REF*, *ET42*, *EBIO*, and *EVEU* (see Table 2). ~~The~~ In all simulation the same set-up for ~~EMAC is identical in all simulations using~~ the EMAC instance is applied, involving the MACCity emission inventory (Granier et al., 2011) with a resolution of $0.5°$ x $0.5°$. ~~For the COSMO model instances, however, the emission inventories as well as the resolution of the emission inventories are varied systematically in~~ The set-ups of the CM50 and CM12 instances (were applied) is varied systematically between the different simulations~~as described below~~. The concept for these variations is the following.

~~In~~ For the *REF* simulation ~~,~~ the same MACCity inventory is applied in EMAC~~and COSMO, using the finest possible resolution in every model instance~~, CM50 and CM12 at its finest available resolution. This means, that the MACCity emissions are transformed onto a grid of 2.8 x 2.8° resolution in EMAC and to a grid of 0.44 x 0.44° in CM50 (and 0.1 x 0.1° resolution in CM12). The different geographical distribution of the emissions is given in Fig. S16 in the Supplement). This simulation serves as reference~~, in order to disentangle effects of the resolution of the model from that of~~ . Differences between the results of the EMAC and CM50 (and CM12) can be attributed to different effects:

First, the dynamical core and physical parametrizations between EMAC and COSMO/MESSy differ, second the resolution of ~~emission inventories, and the different anthropogenic emission inventories. For the *ET42* simulation , the MACCity emissions are transformed to~~ these models differs and third EMAC and COSMO/MESSy calculate different soil-NO_x and biogenic C_5H_8 emissions. The latter due to the meteorology dependence and due to different soil types in EMAC and COSMO/MESSy.

The sensitiviy simulations help to disentangle these factors. The simulation *ET42* applies the identical emissions in CM50 and in EMAC, meaning the emissions are first transformed onto the coarse grid of EMAC (2.8 x 2.8°, T42) ~~, to investigate the impact of the~~ before they are applied at this coarse resolution in CM50. Accordingly, EMAC and CM50 use the same effective resolution of the ~~emission inventory. To study the influence of the different on-line calculated emissions of soil-and isoprene, the~~ anthropogenic emissions. By comparing the results from CM50 of *REF* and *ET42*, the effect of the emission inventory resolution can be analysed.

In the simulation *EBIO* ~~is performed. Here, the biogenic emissions~~ the biogenic C_5H_8 and soil-NO_x emission as calculated by EMAC are ~~mapped down to COSMO and applied instead of the biogenic emissions calculated by COSMO itself. In this case the emissions in COSMO are also scaled by 0.6. Finally,~~ transformed down and applied at the resolution of EMAC in CM50. By comparing the results from CM50 of *REF* and *EBIO* the effect of the differently simulated biogenic emissions can be analysed. These differences of the biogenic emissions are caused by different meteorological conditions simulated by EMAC and CM50.

Finally, the simulation *EVEU* is performed. In this simulation a different emission inventory for the emission sources shipping, land transport and anthropogenic non-traffic is used~~in the simulation *EVEU*~~. This emission inventory is only available for Europe with a resolution of $0.0625°$ x $0.0625°$ and is an outcome of the DLR-project 'Verkehrsentwicklung und Umwelt'

(Hendricks et al., 2017)(VEU, Hendricks et al., 2017). The results of this simulation are important to set the differences caused by the above discussed model related uncertainties into context with the uncertainties caused by emission inventories. Further, the finer resolution of the emission inventory allows to compare the results of CM50 and CM12 to investigate the effect of increased model and emission inventory resolution. The total emissions applied in all simulations are given in the Supplement in Table S2 to Table S10.

The simulated period of the *REF* simulation ranges from covers 07/2007 to 12/2010. All sensitivity simulations are branched off in 12/2007 from the *REF* simulation. The simulation period of the *EVEU* simulation ranges from 12/2007 to 12/2010. The simulation periods of the other simulations are given in Table 2. simulations *ET42* and *EBIO* cover just on year ending in 12/2008. Due to the high computational resources needed for the CM12 model instance, the CM12 instance is only activated for the period May to August 2008 and only for the simulations *REF* and *EVEU* (see also Fig. S14S15).

All chemical species, as well as the tagging diagnostics, are initialised from a 6-month spin-up simulation with EMAC only (period 01/2007–07/2007). This spin-up simulation was initialised with trace gas mixing ratios from the *RC1SD-base-10a* simulation described in detail by Jöckel et al. (2016). The soil-model TERRA of COSMO/MESSy is initialised with an output of a simulation without chemistry for the period 01/1983–07/2007. Further, MECO(n) is operated in the so called quasi chemistry transport model mode (QCTM-mode, Deckert et al., 2011; Mertens et al., 2016). In this mode chemistry and dynamics are decoupled to increase the signal-to-noise ratio for small chemical perturbations. For this climatologies are used within EMACThis means, that even though the emissions differ between the different simulations, each model instance (EMAC, CM50 and CM12) simulates the same meteorology in all simulations, which does of course not imply that the meteorology between the different model instances (EMAC, CM50 and CM12) is the same. In EMAC the QCTM mode is implemented applying the following climatologies: (a) for all radiatively active substances (CO_2 , CH_4 , N_2O, CFC-11 and CFC-12) for the radiation calculations, (b) nitric acid (heterogeneous chemistry ; for the heterogeneous chemistry calculations (submodel MSBM (Multiphase Stratospheric Box Model) and (c) for OH, O^1D and Cl for methane oxidation in the stratosphere (submodel CH4). In COSMO/MESSy only the climatology of nitric acid for the submodel MSBM is requiredcalculation of heterogeneous chemistry is needed. The applied climatologies are monthly mean values from the *RC1SD-base-10a* simulation.

For our comparison we focus on the period June–August (JJA) where the ozone production is largest. Further, we compare the results on the coarsest grid. Of course the finer resolved model instances provide additional information compared to the coarse model instance. On the grid of the finer modelinstace, however, the coarser model instance does not gain any information. Therefore, we investigate if the fine model instance provides an added value compared to the coarse model instance, on the grid of the coarse model instance.

Overview of the most important submodels applied in EMAC and COSMO/MESSy, respectively. Both COSMO/MESSy instances use the same set of submodels. MMD* comprises the MMD2WAY submodel and the MMD library. Submodel EMAC COSMO short description references AEROPT x calculation of aerosol optical properties Dietmüller et al. (2016) AIRSEA x x exchange of tracers between air and sea Pozzer et al. (2006)CH4 x methane oxidation and feedback to hydrological cycle CLOUD x cloud parametrisation Roeckner et al. (2006), Jöckel et al. (2006)CLOUDOPTx cloud optical properties Dietmüller et al. (CONVECT x convection parametrisation Tost et al. (2006b) CVTRANS x x convective tracer transport Tost et al. (2010)DDEP

~~x x dry deposition of aerosols and tracer Kerkweg et al. (2006a) E2COSMO x additional ECHAM5 fields for COSMO coupling Kerkweg and Jöckel (2012b) GWAVE x parametrisation of non-orographic gravity waves Roeckner et al. (2003) JVAL x x calculation of photolysis rates Landgraf and Crutzen (1998), Jöckel et al. (2006) LNOX x -production by lighting Tost et al. (2007) , Jöckel et al. (2010)MECCA x x tropospheric and stratospheric gas-phase chemistry Sander et al. (2011), Jöckel et al. (2010) MMD* x x coupling of EMAC and COSMO/MESSy (including libraries and all submodels) Kerkweg and Jöckel (2012b); Kerkweg et al. (2 MSBM x x multiphase chemistry of the stratosphere Jöckel et al. (2010)OFFEMIS x x prescribed emissions of trace gases and aerosols Kerkweg et al. (2006b) ONEMIS x x on-line calculated emissions of trace gases and aerosols Kerkweg et al. (2006b) ORBIT x x Earth orbit calculations Dietmüller et al. (2016) QBO x Newtonian relaxation of the quasi-biennial oscillation (QBO) Giorgetta and Bengtsson (1999), Jöckel et al. (2006) RAD x radiative transfer calculations Dietmüller et al. (2016) SCAV x x wet deposition and scavenging of trace gases and aerosols Tost et al. (2006a) SEDI x x sedimentation of aerosols Kerkweg et al. (2006a) SORBIT x x sampling along sun synchronous satellite orbits Jöckel et al. (2010) SURFACE x surface properties Jöckel et al. (2016)TAGGING x x Source apportionment using a TAGGING method Grewe et al. (2017)TNUDGE x x Newtonian relaxation of tracers Kerkweg et al. (2006b) TROPOP x x diagnostic calculation of tropopause height and additional diagnostics Jöckel et al. (2006)~~

Table 2. Overview of the applied MECO(2) simulation set-ups and simulation periods. For the EMAC instance the same set-up is applied in all simulations, but the ~~set-up~~ set-ups of the COSMO instances (~~both for~~ CM50 and CM12) ~~is~~ are varied systematically. More details are given in the text. The note 'calculated by EMAC' in the row 'biogenic emissions' means that the emissions, which are calculated by EMAC, are transformed to the COSMO grid during runtime via the MMD2WAY submodel.

Simulation		EMAC		CM50/CM12	
acronym	period	anthropogenic emissions	biogenic emissions	anthropogenic emissions	biogenic emissions
REF	07/2007-12/2010	MACCity, 2.8° x 2.8°	on-line calculated	MACCity, 0.5° x 0.5°	on-line calculated
ET42	12/2007-12/2008			MACCity, 2.8° x 2.8°	on-line calculated
EBIO	~~07~~12/2007-12/2008			MACCity, 0.5° x 0.5°	calculated by EMAC
EVEU	12/2007-12/2010			VEU, 0.0625° x 0.0625°	on-line calculated

3 ~~Difference in Ozone Production~~Model evaluation

To evaluate the performance of the different model instances and of the different simulations, we compare the model results with ground level observations of ozone and measurements from ozone sondes. For the evaluation we use observations by the European Monitoring and Evaluation Programme (EMEP, http://www.emep.int, Tørseth et al., 2012) and ozone sonde data from the world ozone database (WOUDC, http://woudc.org). The methodology is described in detail by Mertens et al. (2016). In comparison to Mertens et al. (2016), however, we here focus on average values for June to August 2008 instead of June and December 2008. A list of the used observation data is part of the Supplement (Section S1).

Table 3. Root-mean-square error (RMSE, in $\mu g\ m^{-3}$ and normalized mean-bias error (MBE, in %) of O_3 for EMAC and CM50 in comparison to ground-level observations. Shown are the averaged values for June to August 2008. The values are calculated from monthly mean values. The model values are height corrected as discussed in detail by Mertens et al. (2016).

	RMSE (in $\mu g\ m^{-3}$)		MB (in %)	
	EMAC	CM50	EMAC	CM50
REF	19.6	25.2	13.1	19.5
EVEU		22.7		16.4
ET42		26.0		20.5
EBIO		26.1		20.4

For a quantitative evaluation we chose the metrics RMSE (root meas square error) and MB (normalised mean bias error). The definition of both quantities is given in Appendix 1. Table 3 lists the RMSE and MB for the EMAC and CM50 instances for all simulations. As the set-up for EMAC is identical in all simulations the model results do not change. Generally, the models results are in agreement with the measurements. The RMSE is in the range of around 19 to 26 $\mu g\ m^{-3}$ and the MB in the range of 13 to 21 %. These deviations from the measurements are in the range of the results from comparable model systems (e.g. Knote et al., 2011; Stock et al., 2014). As already noted by Mertens et al. (2016), CM50 exhibits a larger positive ozone bias compared to EMAC. This bias is mainly caused by a more efficient vertical mixing in COSMO, as well as by a less stable boundary layer during night. The latter is a common problem of many models leading to diurnal cycles with too large ozone values during night, which results in an overall ozone bias (e.g. Travis and Jacob, 2019).

The coarser resolution of the emissions (*ET42*) as well as the different biogenic emissions (*EBIO*) between EMAC and CM50 contribute only partly to the bias of CM50 compared to EMAC. The CM50 ozone bias is larger in *ET42* and *EBIO*. The pattern of the ground level ozone mixing ratio bias of CM50 compared to EMAC is similar for all simulations (see Fig. 3). Generally, CM50 has a positive ozone bias compared to EMAC over most parts of Europe.

Only over the Mediterranean sea, CM50 simulates less ozone compared to EMAC. The lower ozone mixing ratios over the Mediterranean sea can partly be attributed to the coarser resolution of the emissions in EMAC compared to CM50, as the difference is lower in the *ET42* simulation (Fig. 3b). The simulated ozone mixing ratios of CM50 are up to 7.5 $nmol\ mol^1$ larger (JJA 2008) in *ET42* compared to *REF*. Averaged over area of the Mediterranean sea the increase of ozone is around 3 $nmol\ mol^{-1}$. If the soil-NO_x and biogenic emissions as claulcated by EMAC are applied in CM50 (*EBIO*) the ozone mixing ratios throughout the whole domain of CM50 increase by 1 to 3 $nmol\ mol^{-1}$. The differences are largest in South Eastern Europe, the Mediterranean Sea and over the Iberian Peninsula (Fig. 3c). Overall, however, the differences of the CM50 results between the *REF*, *EBIO* and *ET42* are small compared to the bias between EMAC and CM50. especially the positive ozone

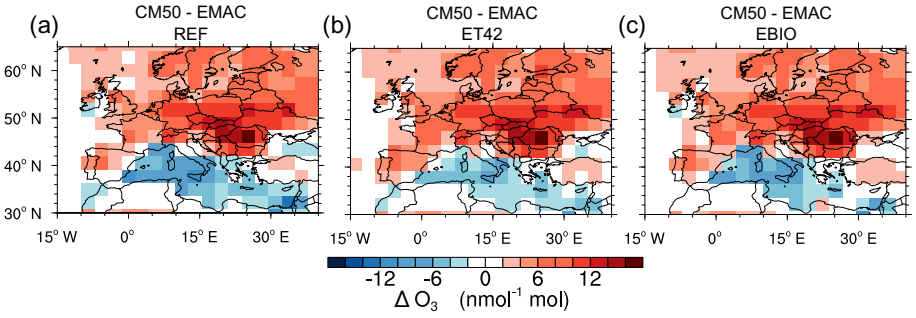

Figure 3. Difference between JJA 2008 averaged ozone mixing ratios (in nmol mol^{-1}) as simulated by CM50 and EMAC ('CM50 MINUS EMAC'); (a) *REF* simulation, (b) *ET42* and (c) *EBIO* simulation.

bias over Serbia and Bulgaria cannot be attributed to different biogenic emisisons or the coarser resolution of the emission inventories in EMAC compared to CM50.

Figure 4 shows scatter plots comparing observed and simulated ozone monthly mean concentrations at all considered stations of the EMEP network. The simulated concentrations for all model instances and simulations lie, with one outlier, around a factor of two of the measurements. As already discussed, the simulated ozone concentrations at most stations show a positive ozone bias. Only at some stations the simulated ozone concentrations are lower as the measured ozone concentrations. The ozone bias is very similar in all CM50 simulations. *EBIO* and *ET42* show almost the same bias as *REF*. Only the simulation *EVEU* shows a slightly lower positive ozone bias. Accordingly, the change of the anthropogenic emission inventory has a larger impact on the model results as the influence of the emission inventory resolution and the geographical distribution of the biogenic emissions.

To evaluate the simulated ozone mixing ratios in the free troposphere, the model results are compared to ozone sonde data (see Sect S1 in the Supplement for a list of considered stations). In total, 510 individual ozone sonde launches are considered for the year 2008. To compare the ozone sonde data with the model results, the vertical ozone profiles simulated by the model were sampled on-line at every time-step of the model at the location were the ozone sonde was launched. Drifts of the ozone sonde by winds are not taken into account. For every launched ozone sonde, we averaged the simulated vertical profiles in time over the measurement period (usually some hours). This temporally averaged vertical profile of simulated ozone mixing ratios is then compared to the measurements of the ozone sonde data. As the main focus of this comparison is the free troposphere, we restrict this analysis to all data in the range from 600 to 200 hPa.

The probability density functions (PDF) for the measured and simulated vertical ozone distributions are displayed in Fig. 5. The results show that in the free troposphere both model instances (EMAC, CM50) simulate a very similar vertical ozone distributions. Compared to the measurements, however, both model instances exhibit a positive ozone bias. Accordingly, the positive ozone bias of CM50 compared to EMAC is mainly confined to the planetary boundary layer.

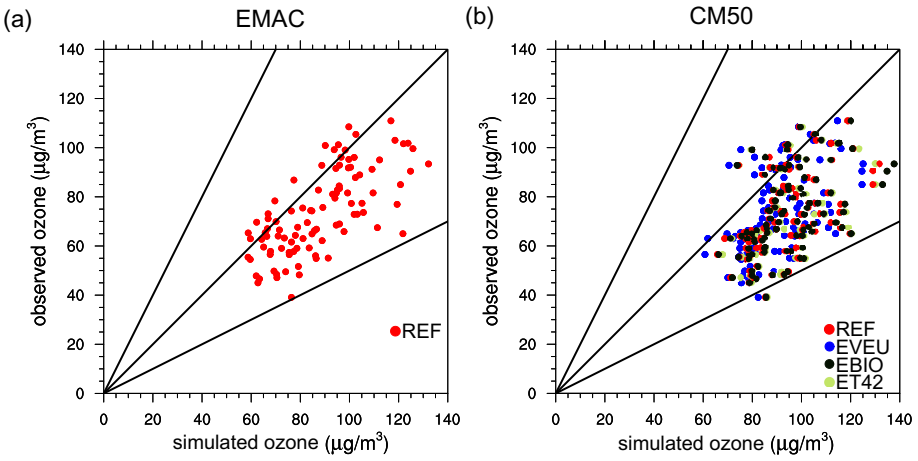

Figure 4. Scatter plot of the observed versus simulated ozone concentrations (in $\mu g\ m^{-3}$) for (a) EMAC and (b) CM50. Each dot represents a monthly mean value for one station in the period June to August 2008. The black lines indicate the 1:1 (observed and simulated concentrations are equal) line, and the range of a factor of two. For EMAC only the results of the *REF* simulation are shown, as the set-up of EMAC is identical in all simulations.

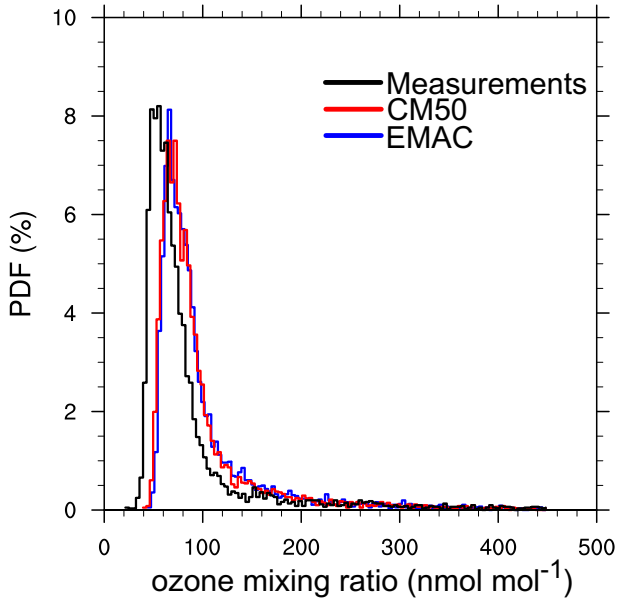

Figure 5. Probability density functions (PDFs) of observed (ozone sondes) and simulated vertical ozone contributions in the rangee of 600 to 200 hPa. Considered are 510 ozone sonde launches for 2008 in Europe.

3.1 Difference in Ozone Production

In a ~~first step, we analyse~~ next step, the difference of the ozone production (for the *REF* simulation) simulated by EMAC and CM50 ~~, respectively~~is analysed. For this, we consider the net ozone production (P_{O3}) defined as:

$$P_{O3} = ProdO3 - LossO3, \quad (1)$$

with the production ($ProdO3$) and loss rates ($LossO3$) as diagnosed by the chemical solver (for more details see Supplement of Grewe et al. (2017)).

We define ΔP_{O3} as $\Delta P_{O3} = {P_{O3}}^{CM50} - {P_{O3}}^{EMAC}$. ΔP_{O3} is largest in the lower troposphere (see Fig. 6a). As indicated by the negative numbers, CM50 simulates in general lower values of P_{O3} than EMAC. Zonally averaged P_{O3} is around 60 to 80 fmol mol^{-1} s^{-1} lower in CM50 as in EMAC, which corresponds to ~~10–20~~10 to 20 %. The largest differences (up to 100 fmol mol^{-1} s^{-1} or 40 %) are simulated over the Mediterranean ~~See~~ Sea (see also Fig. S1 in the Supplement).

To separate effects caused by the emission inventory resolution from the effects caused by the model resolution and specific model biases, Fig. 6b shows the differences of ΔP_{O3} between the *ET42* and *REF* simulation (${\Delta P_{O3}}^{ET42} - {\Delta P_{O3}}^{REF}$). The positive values indicate the effect of increased P_{O3} with reduced resolution of the emission inventory, which is caused by the dilution effect of the emissions on the coarse grid (e.g., Tie et al., 2010). The differences are largest in the Mediterranean area with an increase of P_{O3} in CM50 of up to 40 fmol mol^{-1} s^{-1} in *REF* compared to *ET42*. These differences are mainly simulated in the areas of the Alboran Sea and Balearic Sea, as well as in the areas of the Levantine Sea (see also Fig. S2 in the Supplement). The main reason for these differences are the dilution of the shipping emissions, and the large anthropogenic emissions in Israel if coarse emissions are applied. As the ozone production is strongly non-linear this dilution of the emissions leads to an artificial increase of the ozone production rate.

The other differences, which are not directly caused by the resolution of the anthropogenic emission inventory, are caused by a variety of other model factors ~~,~~ which cannot be disentangled ~~. Some of these factors are model specific temperature biases, differences of~~ in detail. The most important factor in this context is the enhanced vertical mixing in CM50 compared to EMAC, mainly in the boundary layer, but also due to stronger up- and downdraft massfluxes in CM50 compared to EMAC through convection. The enhanced vertical mixing transports ozone from above to the boundary layer and ozone precursors from the boundary layer more efficiently into the free troposphere. Further, differences in the land use classes between EMAC and CM50 lead to differences of the calculated dry deposition velocities, which affects ~~loss processes (like dry deposition) or differences in vertical mixing (see Mertens et al., 2016).~~ also ozone mixing ratios near the surface (see also Mertens et al., 2016).

4 Contributors to ozone in Europe

Figure 7 shows the absolute and relative contributions of different emission sources to the European ozone column up to 850 hPa as simulated by EMAC and CM50 for the *REF* simulation (see Table S1 in the Supplement for detailed definition of the tagging categories). The largest absolute and relative ozone contributors are the anthropogenic non-traffic and the biogenic

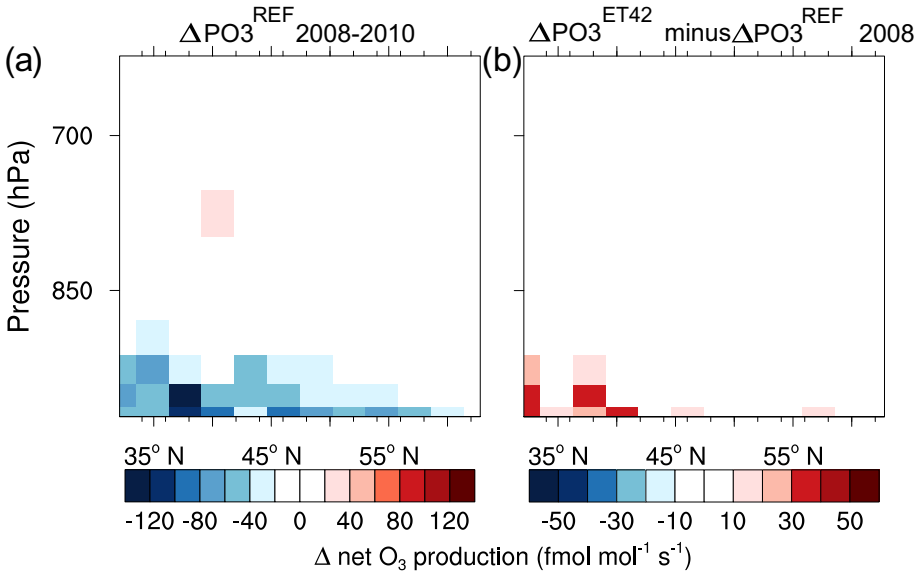

Figure 6. Zonally averaged differences of P_{O3} (ΔP_{O3}) between CM50 and EMAC (in fmol mol^{-1} s^{-1}). **(a)** ΔP_{O3} calculated from the results of the *REF* simulation for JJA 2008–2010. **(b)** differences of ΔP_{O3} between the *ET42* and *REF* simulations for the year 2008 only. The CM50 data have been transformed on the horizontal and vertical grid of EMAC.

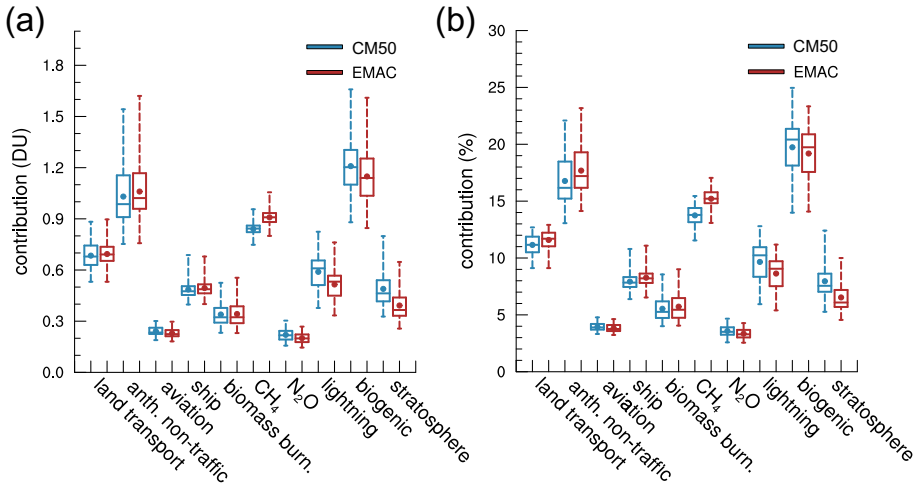

Figure 7. Box and whisker plot for the absolute (a, in DU) and relative (b, in %) contribution to the ozone column up to 850 hPa. The values are area-averaged over the CM50 domain. The lower and upper ends of the boxes indicates the 25th and 75th percentiles, the bars the medians, the dots the average and the whiskers the ranges of the timeseries for the JJA values from 2008–2010.

categories, both with contributions of more than 1 DU corresponding to more than 15 %. Both ~~models~~ model instances simulate similar absolute ozone contributions of the categories anthropogenic non-traffic ($\approx$ 1.0 DU), land transport ($\approx$ 0.7 DU),

ship (≈ 0.5 DU) and biomass burning (≈ 0.4 DU). For the biogenic category, CM50 calculates slightly larger absolute contributions compared to EMAC (see Sect. 4.2), but the differences are small compared to the temporal variability of the contributions. Further, CM50 calculates larger absolute contributions of the categories ~~lighting~~ lightning and stratosphere. ~~Due to~~ This affects mainly the categories land transport, anthropogenic non-traffic, shipping and biomass burning, where EMAC simulates 0.1 to around 1 percentage − point larger relative contributions compared to CM50. At the same time the increased vertical mixing in CM50 leads to an increase of the relative contributions of the categories stratosphere, lightning and aviation compared to EMAC~~ozone which is produced in the upper troposphere is transported downward more efficiently. This leads to overall larger ozone columns up to 850 in~~ . Here, the differences are in the range of 0.1 to around 1.5 percentage − points.

The positive ozone bias of CM50 compared to EMAC ~~(Mertens et al., 2016)~~indicates an too efficient vertical mixing in CM50 (see Sect. 3). Therefore, ~~EMAC simulates, despite similar absolute contributions of the anthropogenic categories, slightly larger relative contributions for these categories . Accordingly, CM50 simulates larger relative contributions to near ground-level ozone of~~ the ~~categories lightning and stratosphere compared to EMAC. These~~larger contributions of the categories stratosphere and lightning in CM50 compared to EMAC are likely an artefact of this too efficient vertical mixing. However, individual stratosphere-troposphere-exchange (STE) events are better represented in CM50 due to the increased resolution of CM50 (Hofmann et al., 2016; Mertens et al., 2016). Generally the correct representation of STE events poses a big challenge in most models (e.g. Zhang et al., 2011; Lin et al., 2012; Lefohn et al., 2014) and our results suggest also a large uncertainty of the STE contribution simulated by different models to ground level ozone.

The values which we discussed so far, however, are averages on continental scale. ~~In a next step the differences of the geographical distribution are analysed in more detail. Here, we focus~~ On the regional scale the differences can be much larger. Geographical distributions of the differences for the absolute and relative contributions as simulated by EMAC and CM50 are given in the Supplement (Figs S3 and S4). Next, we want to focus exemplary on the categories land transport, as one important anthropogenic emission source, and biogenic emissions~~(for~~ . For all other categories the differences are shown in Fig. S3 in the Supplement~~)~~. As discussed in Sect. 2, the biogenic emissions are calculated on-line by both ~~models~~ model instances depending on the meteorology and surface properties. While the total emissions are comparable, the geographical distribution, as well as the area averaged contribution, differ (see Supplement Fig. S17 and Tables S2 to S10). As differences of on-line simulated emissions are a typical ~~inter-model~~ inter-model difference, a detailed investigation of the influence of these differences is of interest.

4.1 Contribution of land transport emissions to ground level ozone

Averaged over JJA 2008 and the European area (defined as rectangular box from 10° W: 30° E and 32° N: 65° N, see red square in Fig. 1) EMAC simulates a relative contribution of the land transport emissions (denoted as O_3^{tra}) to ground level ozone of 13.1 %, while CM50 calculates a contribution of 11.9 %. A decrease of the emission resolution in CM50 increases the relative contribution to 12.1 % (*ET42* simulation), and the change of the anthropogenic emission inventory in CM50 increases the contribution to 12.7 % (*EVEU* simulation). In all cases similar absolute contributions of O_3^{tra} are simulated which range between 6.0 and 6.4 nmol mol^{-1}. ~~Accordingly, the~~ The area averaged values indicate that the inter-model differences

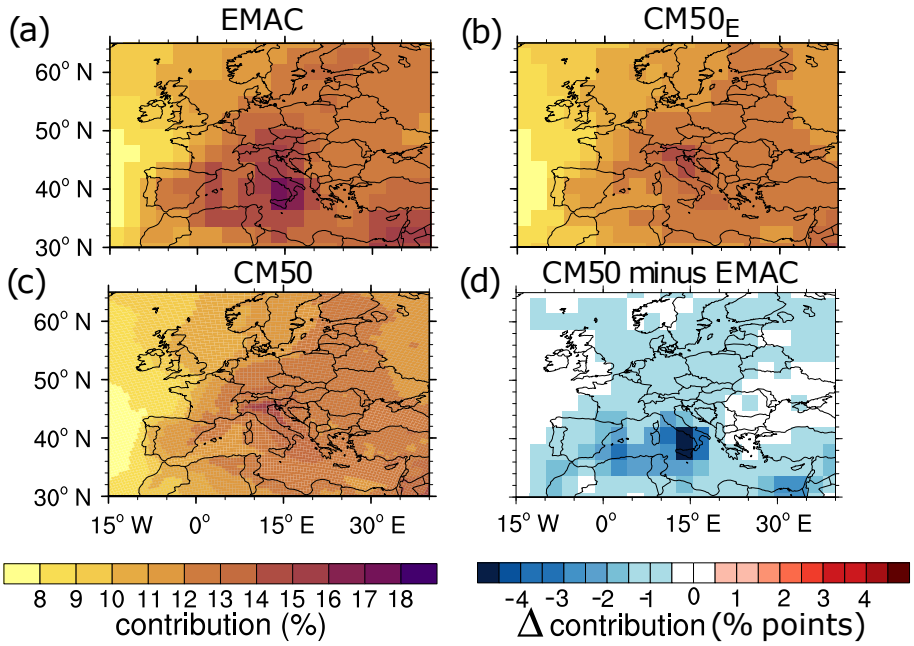

Figure 8. Comparison of the JJA averaged relative contribution of O_3^{tra} to groundlevel O_3 (in %) of EMAC and CM50: **(a)** results of EMAC, **(b)** results of CM50 transformed onto the EMAC grid, **(c)** results of CM50 on the original grid and **(d)** difference ('CM50 minus EMAC' in percentage points) on the coarse grid. **(a)**– **(c)** use the same (left) colour bar. Shown are the results of the *REF* simulation, averaged for 2008–2010.

as discussed in detail in Sect. 3 between CM50 and EMAC have a larger influence on the calculated contributions than the change of the anthropogenic emission inventory. The impact of the coarsely resolved emission inventory on the area averaged values is rather small. In general, the differences of the average contributions of O_3^{tra} simulated by the two ~~models~~ model instances (EMAC and ~~COSMO~~CM50), as well as simulated by ~~COSMO~~ CM50 for the four different simulations are ≈ 10 % at maximum. In comparison to this, the ~~inter-model~~ differences of the contributions to ground-level O_3 ~~with respect to~~ between EMAC and CM50 of the categories lightning and stratosphere ~~, which result mainly from the differences of the dynamics,~~ are much larger (≈ 20 % and ≈ 30 %, respectively).

Regionally, the differences in relative contribution of O_3^{tra} to ground level ozone (see Fig. 8) can be larger than the area averaged differences. In general, both ~~models~~ model instances simulate a comparable distribution with the largest relative contribution of O_3^{tra} in the Mediterranean region and contributions of around 8 % over the western Atlantic. ~~As discussed before,~~ These values are larger (10–18 %) over the continent. CM50 simulates ~~over large parts of Europe~~ a 0.5–1 percentage points lower relative contribution compared to EMAC~~, mainly caused by a decreased net ozone production and a~~. As discussed before, this can mainly be attributed to stronger vertical mixing and reduced ozone production (P_{O3}) in CM50 compared to EMAC. With increasing altitude the differences between EMAC and CM50 decrease (see Fig. ~~S4~~ S5 in the Supplement).

The largest differences of the relative contribution of O_3^{tra} to ground level ozone are simulated around the Mediterranean area. The differences over the Mediterranean Sea (up to 2 percentage points and more, corresponding to more than 10 percent) can partly be attributed to the coarse resolution of the emissions in EMAC compared to CM50. ~~As the analyses of the *ET42* simulation results shows, the~~ The coarse resolution leads to an artificial increase of P_{O3} (see Sect. 3.1) which in turn leads to an increase of the contribution from O_3^{tra} (and other anthropogenic categories). Accordingly, the results of CM50 of the *ET42* simulation shows regionally up to 3 $nmol\ mol^{-1}$ and 3 percentage points larger contributions of land transport emissions to ozone as the *REF* simulation (see also Fig. ~~S5~~ S7 in the Supplement). However, especially the large differences over Southern Italy and Sicily between CM50 and EMAC can not be attributed to the coarse resolution of the emissions. Here, EMAC simulates the largest contribution (up to 17 %) in the European region (especially around the Naples region with large land transport emissions), while CM50 simulates contributions of around 13 %. On the coarse EMAC grid most parts of Southern Italy are considered as sea, affecting especially the calculation of dry deposition in EMAC, as dry deposition of ozone is lower over sea as over land. Therefore, the coarse resolution of the land sea mask in EMAC compared to CM50 leads to an artificial underestimation of the ~~loss~~ozone dry deposition in EMAC. In addition, the coarse land sea mask leads to differences in the calculation of biogenic emissions. Especially over Sicily, EMAC simulates no biogenic emissions (including soil-NO_x) while CM50 simulates large emissions here (see Fig. ~~S8~~ S17 in the Supplement). Accordingly, soil-NO_x and anthropogenic NO_x do not compete in EMAC in this area and ozone is mostly formed from anthropogenic emissions. Compared to this artificial peak ~~produced~~ simulated by EMAC around Naples and over Sicily, CM50 shows the largest contribution (up to 15 %) around the Po ~~basin~~Valley. In this region, large amounts of emissions by land transport take place and ozone production is enhanced by stable and sunny weather conditions. The differences between EMAC and CM50 around the Naples region are even larger (up to 6 $percentage - points$, see Fig. S6 in the Supplement) for the extreme values (95th percentile) as for the means values which were discussed so far. Accordingly, extreme values are even stronger deteriorated as the mean values by the coarse land-sea mask problems discussed above.

The further increase of resolution from 50 km (CM50) to 12 km (CM12) impacts ozone and the contributions of ozone only slightly (see Fig. ~~S10~~ S11 in the Supplement). In general, we note a decrease of the absolute ozone values, as well as the absolute ~~contribution~~ contributions of anthropogenic emissions (including the land transport category) near the hotspot regions (e.g. Rhine-Ruhr, Munich, and Frankfurt), if the model resolution is increased (*REF* simulation). The increase of the resolution of the emission inventory (*EVEU* simulation) intensifies this effect, i.e. near the hotspots ozone values and absolute contributions of O_3^{tra} decrease further. In Southern and Eastern Germany, however, the ozone values increase. ~~Especially in Southern Germany~~ As a comparision of the contributions of the individual tagging categories shows, this is mainly caused by ~~the better resolved topography and larger contributions of stratospheric~~ an increase of the contribution from stratospheric ozone and from the CH_4 category. The first is partly caused by the enhanced topography in CM12 compared to CM50, further the convective up- and downdraft massfluxes are larger in CM12 compared to CM50. The larger contribution of ozone ~~.~~ ~~Accordingly, the absolute contribution of land transport emissions to ground level ozone decreases in this area.~~ from the CH_4 category (meaning more ozone formed by reactions involving CH_4 oxidation products) is consistent with the findings of the larger tropospheric oxidation capacity (i.e. lower methane liftetime) in CM12 compared to CM50 by Mertens et al. (2016).

~~Focusing on the~~ The relative contribution of O_3^{tra} to groundlevel O_3 averaged over Germany ~~we note a decrease~~ decreases in CM12 compared to CM50 (see Fig. 9). The difference is largest in Southern Germany, however, mostly below 0.5 percentage points (corresponding to less than 5 %). ~~As discussed above, this is mainly caused by increased ozone of stratospheric origin. Further, in Western Germany CM12 simulates a larger contribution of the category to ozone compared to CM50, which is consistent with the larger tropospheric oxidation capacity in CM12 compared to CM50 (Mertens et al., 2016).~~

In general, however, the differences of the ~~contributions of to ground level ozone for the~~ 95th percentile (see Fig. ~~S11~~ S12 in the Supplement) and the mean ~~between the results of~~ of the contributions of O_3^{tra} between CM12 and CM50 are much smaller compared to the differences caused by the different anthropogenic emissions inventory (e.g. the differences of the results of the *REF* and *EVEU* simulation). Accordingly, the differences of emission inventories dominate over differences caused by the resolution of emission inventories and models when comparing the results of CM50 and CM12.

What is not discussed here in detail is the influence of the difference of the shorter lived species, e.g. NO_2 or the tagged contributions to NO_y, which largely differ between the two resolutions. Here, maxima (e.g. in Stuttgart or around the Rhine-Ruhr area) are displaced in the coarser resolution (CM50) compared to the finer resolution (CM12). However, the direct influence of displaced precursors on ozone itself is not very large, because ozone formation usually takes place downwind of the source itself. Further, compared to previous studies investigating the influence of the model/emission inventory resolution on ozone (e.g. Wild, 2007; Tie et al., 2010; Markakis et al., 2015), it is important to note that we apply a chemistry-climate model in which not only the chemical processes are calculated on the finer grid, but also the meteorology. This can alter the results compared to studies applying simpler chemistry-transport models.

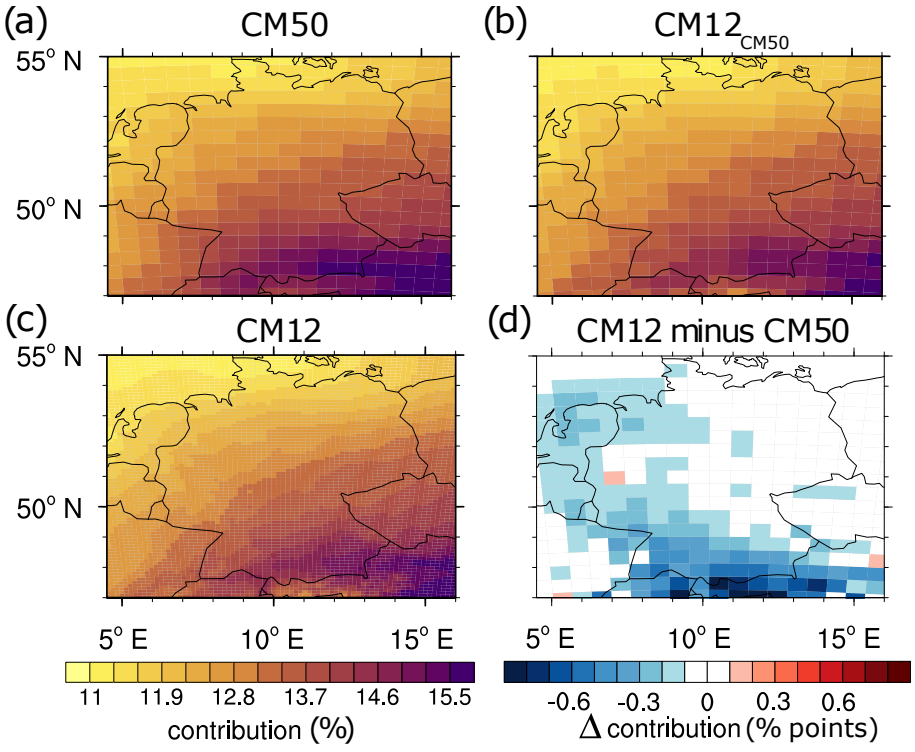

Figure 9. Comparison of the JJA averaged ground level contribution of O_3^{tra} to O_3 (in %) of CM50 and CM12: **(a)** results of CM50, **(b)** results of CM12 transformed onto the CM50 grid, **(c)** results of CM12 on the original grid and **(d)** difference ('CM12 minus CM50' in percentage points) on the coarse grid. **(a)**–**(c)** use the same (left) colour bar. Shown are the results of the *EVEU* simulation, averaged for 2008.

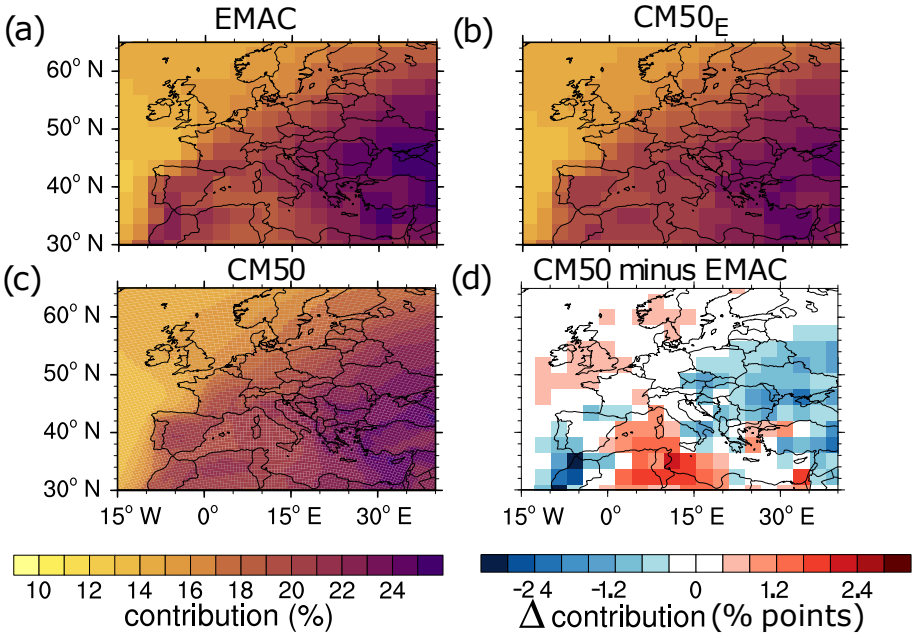

Figure 10. Comparison of the JJA averaged ground level contribution of O_3^{soi} to O_3 (in %) of EMAC and COSMO: **(a)** results of EMAC, **(b)** results of CM50 transformed onto the EMAC grid, **(c)** results of CM50 on the original grid and **(d)** difference ('CM50 minus EMAC' in percentage points) on the coarse grid. **(a)**– **(c)** use the same (left) colour bar. Shown are the results of the *REF* simulation, averaged for 2008–2010.

4.2 Contribution of biogenic emissions to ground level ozone

The JJA 2008 averaged relative contribution of ozone from biogenic emissions (mainly soil-NO_x and biogenic C_5H_8, denoted as O_3^{soi}) to ground level O_3 over the rectangular box defined as Europe (see Sect. 4.1) range from 19.0 to 19.6 % in all simulations. Hence, the differences of the relative contribution of O_3^{soi} to ground level ozone on the continental scale are rather small (below 5 %). The same is true for the absolute values, ranging from 9.3 to 9.7 $\mathrm{nmol\ mol^{-1}}$.

With respect to the geographical distribution (Fig. 10) both ~~models~~ model instances simulate a strong North-West to South-East gradient with relative contributions from O_3^{tra} of around 10 % over the Atlantic and more than 20 % over South-Eastern Europe. In contrast to the contribution of O_3^{tra}, EMAC does not show generally larger contributions of the biogenic category than CM50. Instead, EMAC simulates (~~in case of the~~ *REF* simulation) larger contributions (1–2 percentage points) over South-Eastern Europe and Morocco/Iberian Peninsula, while CM50 simulates around 1–2 percentage points larger contributions over large parts of the Mediterranean Sea as well as over Northern Africa. Also around the British Islands and Norway, the relative contributions of O_3^{soi} simulated by CM50 are larger by around 0.5 percentage points compared to EMAC. In total, CM50 ends up with 0.5 percentage points larger contributions of O_3^{soi} compared to EMAC. Similar as for the land transport category, the differences between the results of both ~~models~~ model instances decrease with increasing height, but the general pattern stay similarly (see Figs. ~~S7~~ S8 in the Supplement).

The differences between EMAC and CM50 are only partly caused by the different geographical distribution of the biogenic emissions in EMAC compared to CM50. When applying the same biogenic emissions as in EMAC in CM50 (*EBIO* simulation) the relative and absolute contributions of O_3^{soi} are increased mainly in the Mediterranean area by up to 2 percentage points and 3 $\mathrm{nmol\ mol^{-1}}$, respectively (see Fig. S9 and Fig. S10 in the Supplement). The characteristic dipole pattern ~~between EMAC and~~ with lower contributions of O_3^{soi} in South-Eastern Europe and larger contributions in Southern Europe and Northern Africa in CM50 ~~, however, stays similarbut the maximum is reduced (see Fig. S8 and Fig. S9 in the Supplement). These differences are~~ compared to EMAC remains similar. This pattern can partly be attributed to the coarse resolution of the shipping emissions in EMAC, leading to a positive ozone bias in the Mediterranean sea (see Sect. 3). The dipol pattern, however is neither caused by the coarse resolution of the emissions nor by different biogenic emissions, but mainly caused by ~~inter-model differences~~ the differences in meteorology between EMAC and CM50.

In general, we conclude that regionally differences of the relative and absolute contribution of O_3^{soi} caused by inter-model differences, emission resolution as well as different geographical distribution are up to 15 %. Averaged over Europe the differences are lower (10 %). Again, the differences are lower as for example the differences of around 30 % observed for the differences of the contributions to ozone from the stratosphere.

5 Discussion

So far, the results indicate that with respect to average values on continental scale, the differences caused by the ~~resolution~~ resolutions of the model/emission inventory are rather small. This confirms findings by Stock et al. (2013), reporting only a small influence of the global redistribution of megacity emissions (which can be seen as a locally decreased emission resolution) on the global ozone budget.

To summarise and quantify these differences in more detail, Fig. 11 shows the absolute (a) and relative (b) contributions of O_3^{tra} to ground level ozone ~~for the whole~~ averaged over the CM50 domain, as well as for the geographical regions defined in the Prudence project (Christensen et al., 2007). The results of EMAC are not analysed for these geographical regions, as due to the coarse resolution some regions would only consist of a few grid points.

Figure 11 shows that also on the scale of smaller regions, the absolute and the relative contribution of O_3^{tra} to ground-level ozone is only slightly influenced by the coarse resolution of anthropogenic emission inventories (*ET42*) as well as by a different geographical location or resolution of biogenic emissions (*EBIO*). This does not only hold for the mean O_3^{tra} contributions, but also for the extreme values expressed by the 95th percentile. Also the simulated differences for the biogenic and shipping category, which are affected much more by the changed variations of the emission inventories in the two simulations, are rather small (see Fig. ~~S12~~ S13 and Fig. ~~S13~~ S14 in the Supplement). The largest simulated differences of the contribution of shipping emissions to ground-level ozone between the *REF*, *EBIO* and *ET42* simulation are around 0.5 $\mathrm{nmol\ mol^{-1}}$ and below 0.5 percentage points, respectively. The largest change (95th percentile) of the biogenic category in the region Iberian Peninsula is around 0.7 $\mathrm{nmol\ mol}$ and 0.5 $\mathrm{percentage-points}$~~, respectively~~.

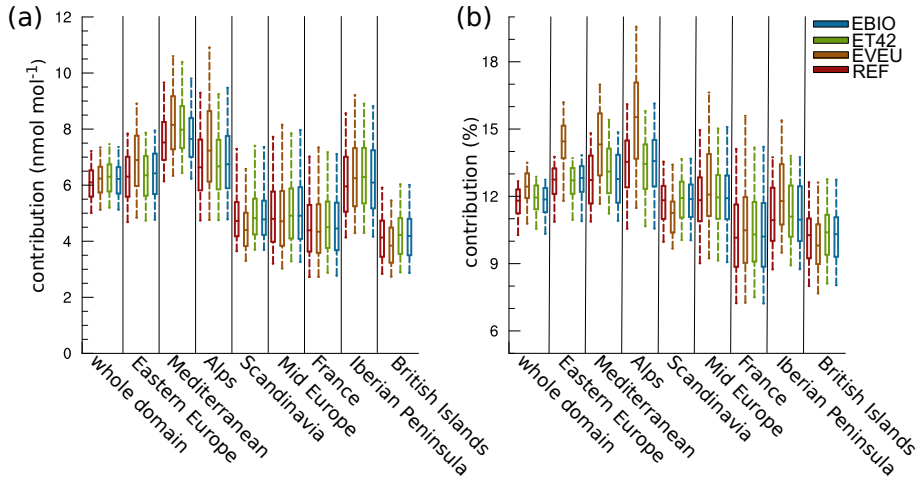

Figure 11. Comparison of the contributions of O_3^{tra} to ground level ozone for JJA 2008 between the four simulations. (a) displays the absolute contribution in $\mathrm{nmol\ mol^{-1}}$ and (b) the relative contribution to ground level ozone (in %). All values are area averaged over the respective region and are calculated using the results of the CM50 instance. The lower and upper end of the box indicate the 25th and 75th percentile, respectively, the bar the median, and the whiskers the 5th and 95th percentile of the timeseries for the JJA values from 2008 based on 3-hourly model output.

Compared to the differences of the contribution of O_3^{tra} between the *REF*, *ET42*, and *EBIO*, the differences caused by a changed emission inventory (*EVEU*) are larger. In the Mediterranean region, the mean and 95th percentile of the contribution of O_3^{tra} increases by 1 $\mathrm{nmol\ mol^{-1}}$ and 2 percentage points, respectively. In the Alps region the increase of the 95th percentile of the contribution is up to 1.3 $\mathrm{nmol\ mol^{-1}}$ and 3 percentage points, respectively. Similarly, also for the contribution of shipping emissions the differences are largest with the changed emission inventory (up to 1.5 $\mathrm{nmol\ mol^{-1}}$ and 1 percentage point). Accordingly, changes in the resolution of the emission inventory or the biogenic emissions can ~~effect~~ affect the contribution of anthropogenic categories (such as land transport and shipping). However, on the regional scale the main drivers of uncertainties are clearly the anthropogenic emissions ~~as well as inter model differences~~ and differences in the results of different models. As an example we found regional differences (cf. Sect. 4.1) of the contribution of O_3^{tra} to ground level O_3 between EMAC and CM50 of up to 20 % around the Naples region, which in this case can mainly be attributed to the coarse land-sea mask of EMAC, leading to the emission of land transport emissions over the sea.

The results of the model evaluation, however, are not very helpful in judging which of the two emission inventories are more realistic. Although, *EVEU* shows a smaller ozone bias compared to *REF* caused by reduced precursor emissions, it is unclear if lower anthropogenic non-traffic emissions in the VEU compared to MAC emission inventories is realistic.

6 Summary and Conclusions

In the present study, we are focusing on the question "Are contributions of emissions to ozone a matter of scale?'. To answer this question we compare the influence of the model, the model resolution, the emission resolution and the emission inventory on the results of ozone contribution analyses. For this we apply the MECO(n) model system which combines a global and a regional model by means of an on-line nesting technique. By applying the identical tagging diagnostics (source apportionment method) in the regional and global model and consistent boundary conditions, we are able to compare the results of model instances with different resolutions to investigate the influence of the model and emission inventory resolutions onto the diagnosed ozone contributions. Such analyses are important ~~to quantify~~ for quantifying uncertainties of ozone source apportionment studies, which arise due to limitations of the model and/or computational resources.

~~Our comparison showed, that with respect to~~ For the specific model set-up involving the global model EMAC and the regional model COSMO/MESSy our results show that simulated differences of ozone contributions on continental scale (e.g. Europe) ~~the differences simulated by our global and our regional model and two specific anthropogenic emission inventories~~ are rather small. The largest differences of the contribution of anthropogenic emission sources was up to 10 % for the contribution of land transport emissions to ground level ozone. However, the contribution of stratospheric ozone to ground level ozone calculated by EMAC and COSMO differs by up to 30 %~~, suggesting that inter-model differences with respect to downward transport of ozone can be larger.~~

~~Consistent with previous studies, a coarser resolution of the emission inventory and~~. One main reason for this large difference of the contributions of stratospheric ozone between the two models are the enhanced vertical mixing in COSMO/~~or of the model increases on average the ozone mixing ratios and the absolute contributions of emission sources to ozone. However, the relative contributions to ozone change only slightly. In general, the difference caused by the emission inventory resolution are smaller than differences arising from the different models and emission inventories.~~

~~Accordingly, the answer to the questions in the title is a clear 'it depends'. Questions such as 'What is the average contribution~~ MESSy compared to EMAC as well as stronger up- and downdraft massfluxes due to convection. Taking the comparison with the measurements into account the vertical mixing in COSMO/MESSy and the enhanced stratospheric contribution are likely too large. On the regional scale the differences between the results of COMSO/MESSy and EMAC are much larger. Here, we observed differences of up to 20 % for the contributions of land transport emissions to ~~ozone over Europe' can be answered with a global model and/or coarser resolved emissions , taking into account positively biased absolute contributions. On the regional scale, however, inter-model differences (caused partly by the model resolution) as well as effects introduced by specific emission inventoriesor the emission inventory resolution can be much larger. For the investigated combinations of models and emissions we observed~~ ground level ozone. This difference is mainly caused by the coarse land-sea mask of the global model instance, leading to emissions of land transport emissions over sea, different dry deposition of ozone and missing biogenic emissions. Taking the results of the same model instance (CM50) into acount the largest influence on the results are clearly caused by different emissions inventories. However, locally also coarse resolved emission inventories and differences of the biogenic emissions can lead to differences of up to ~~-~~20 %~~for the contributions of land transport emissions to ground level~~

ozone. Further, maxima areshifted by misplaced emissions or coarsely resolved land sea masks due to the coarser resolutionof the emission inventories or the models. In addition, we showed how the differences of the source apportionemnt results between different model instances can help to explain model biases and the physicial/chemical mechanisms causing these biases.

Apart from many model specific findings of this study, its results have important implications for other modelling studies and modellers applying source apportionment methods. These implications are:

- First, our study shows that average continental contributions of anthropogenic emissions are quite robust with respect to the used model and the used model resolution. This means that global models at coarse resolution can be used to perform ozone source apportionment in this global context.

- Second, our results also show that on the regional scale, the differences either caused by different models, but also by model resolution are much larger. These effects arise mainly near hotspot regions like the Po basin Valley or near major shipping routes in the Mediterranean Sea. However, especially in these areas, contribution analyses of anthropogenic emissions are very important and effects like spurious effects, such as artificially increased ozone levels and contributions caused by the coarse resolution of models and or emission inventories should be avoided. Hence, for regional analyses fine resolved models and emission inventories are required.

- Third, our results clearly indicate how large the spread between models with respect to STE is. The importance of stratospheric ozone, both in the global and the regional model, corroborates the necessity of tracing the contributions of stratospheric ozone to ground level ozone explicitly by the source apportionment methods. However, only few currently available methods used on the regional scale account for this process.

Further, this study shows how the application of a source apportionment method helps in explaining differences between the results of different models or model configurations. In this case, the larger ground level ozone mixing ratios simulated by COSMO compared to EMAC can partly be explained by a more efficient vertical mixing, which is supported by a larger contribution of the stratospheric ozone at ground level simulated by COSMO compared to EMAC.

Clearly, this study is only a first step to quantify the driving sources of uncertainties and especially the role of the model and emission inventory resolutions on the results of ozone contribution studies. Especially, as some processes like vertical diffusion or transport can heavily alter the model results, follow up studies need to take into account more (and more different) models to better estimate the inter-model differences when applying source apportionment methodsthe uncertainties caused only by differences of the meteorology simulated by different models. In addition, the two analysed anthropogenic emission inventories clearly do not reflect the whole spectrum of different emission estimates. Further, our analyses focused only on differences near the origin of the emissions. An increased resolution leads to a more realistic chemistry within the plumes downwind of the emission hotspots. This can affect the long range transport from different precursors and might influence regions far away from the emission region. Especially calculations of radiative forcings are very sensitive to ozone near the tropopause. In a coarsely resolved model, the overestimated absolute contributions might lead to a biased radiative forcing. This effect, however, is difficult to quantify and would require very fine resolved global chemistry climate models or 2-way-nesting capabilities, which

feed back information about the contributions from the fine back to the coarse grid. For a next step a further increase of the model and emission resolution should be envisaged. Even if we found only small differences between 50 and 12 km resolution this step would be important, as even with a 12 km grid resolution emissions are diluted over large areas. A finer resolution could reduce the dilution strongly. Such an analysis, however, is hindered by two aspects: First, consistent emission inventories (anthropogenic and natural) with a resolution of 1 km over areas, which are large enough to compare models on regional and global scale must be available. Second, requirements with respect to computational time of chemistry-climate models with $\approx$ 1 km resolution over large computational domains are very demanding, hindering detailed quantification of the differences caused by the resolution over long integration periods.

Code and data availability. The Modular Earth Submodel System (MESSy) is continuously further developed and applied by a consortium of institutions. The usage of MESSy and access to the source code is licenced to all affiliates of institutions which are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information, including on how to become licensee for the required third party software, can be found on the MESSy Consortium Website (http://www.messy-interface.org). The code presented here has been based on MESSy version 2.50 and is available in the official release (version 2.51). The namelist set-up used for the simulations is part of the electronic supplement. The data used for the figures 6 to 11 are part of the electronic supplement.

Appendix A: Definition of RMSE and MB

We define the root mean square error (RMSE) as:

$$RMSE = \sqrt{\frac{1}{n}\Sigma_{i=1}^{n}\left(\mathrm{O_3}_i^{mod} - \mathrm{O_3}_i^{meas}\right)^2}, \tag{A1}$$

where n is the number of data points, $\mathrm{O_3}^{mod}$ the simulated and $\mathrm{O_3}^{meas}$ the measured ozone concentrations.
The normalized mean bias error (MB) is defined as:

$$MB = \left(\frac{\overline{\mathrm{O_3}^{mod}}}{\overline{\mathrm{O_3}^{meas}}} - 1\right) \cdot 100, \tag{A2}$$

where $\overline{\mathrm{O_3}^{mod}}$ and $\overline{\mathrm{O_3}^{meas}}$ are the simulated and measured ozone concetrations averaged for all stations and month, respectively.

Competing interests. The authors declare that they have no competing interests.

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