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Interactive comment on "Attributing land transport emissions to ozone and ozone precursors in Europe and Germany" *by* Mariano Mertens et al.

Anonymous Referee #2

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Mertens et al. perform a source attribution study examining the contribution of different emission sectors to air pollution over Europe, with a focus on ozone as a pollutant, a special focus on emissions from the road transport sector, and a regional focus on Europe and Germany. They employ a uniform methodology for "tagging" the emissions of ozone precursors in a system of coupled models, allowing a consistent downscaling to be made from the global scale to the national scale. Furthermore, they compare two simulations performed with different emission inventories, showing the sensitivity of the sectoral contributions to the way in which the emissions from each sector are represented in the emission inventory. This combination of sensitivity and source attribution reveals some interesting information about the behaviour of of tropospheric ozone in the model system used, for example the particularly strong differences in the contribution of land transport emissions to the higher percentiles of the ozone distribution when a spatially more explicit inventory is used.

The manuscript is clearly within the scope of ACP, and the method clearly has a lot of potential to inform international air quality policy. Unfortunately the manuscript in its current form suffers from a number of serious flaws, which must be corrected before it can be accepted for publication.

Firstly, the quality of the written English is terrible. The manuscript is littered with grammatical and spelling errors, and written in a generally inaccessible style. I do not feel that it is my job as a reviewer to provide an exhaustive list of these errors. The authors should seek additional help to get the language up to an acceptable standard. I will give one example though: the very title of the manuscript contains a jarring error. The current title basically implies that ozone causes land transport emissions. Clearly this is the other way around. Land transport emissions happen first, and this leads to ozone production. A grammatically correct title could be "Attributing ozone and its precursors to land transport emissions in Europe and Germany".

Specific Comments

In the abstract, the authors state that tagging is "required" and that their method is the "only possible" way to examine global to regional scale effects. This language is way too strong and should be toned down before publication. This is especially true given that the authors themselves state on line 28 of page 25 that their results are "consistent" with a perturbation study, and also given the fact that the experiment design doesn't actually make a distinction between land transport emissions in Europe and the rest of the world.

Page 2, lines 26-27: while this is generally true on very small scales (eg. urban areas), the response of ozone to perturbation of precursor emissions in remote regions has been shown to be approximately linear. See for example Wild et al. (2012) and Turnock et al. (2018). Since the authors are also discussing long-range transport, some additional discussion of this here would be relevant.

Page 4, lines 3-4: Aren't the last two points in this list in fact exactly the same thing?

Page 4, line 5: I can see how using two different inventories can somewhat account for uncertainties in the emissions, but three years is way to short a period to account for interannual variability. I also do not see how the model uncertainty or the uncertainty in the choice of source apportionment method is accounted for at all in this experiment design. It's fine to mention that there can be a lot of uncertainty, but the authors should not claim to be doing more to address these uncertainties than they actually are.

Page 5, lines 32-35: These are the only lines in the paper where the authors discuss model evaluation. I understand that the model has been evaluated elsewhere, and the model is basically as {good,bad} as other models, but I would appreciate some more discussion about how the model performance could be expected to influence the conclusions of the manuscript. Since the authors also want to use their model to examine extreme ozone events (in Section 4.2), there must be at least some analysis of how well the model is capable of representing these events in comparison with observations.

Section 2.1: the authors need to do a lot more here to compare their source apportionment method with other methods in the literature. This is especially important, since the authors themselves have stated on Page 4 (line 1) that differences between source apportionment methods are an important source of uncertainty. Kwok et al. (2015) is already mentioned in Section 2.1, and Dunker et al. (2002) is mentioned in the introduction. Both of these studies use a regime-dependent attribution methodology, which is actually correctly acknowledged by the authors on page 26 in the Discussion section, but a discussion of how these methodologies differ from the methodology employed by the authors, and how this could be expected to influence the results of the study is required already in Section 2.1. Similarly, since the authors are also considering the global scale, they should also put their methodology into the context of the existing techniques for source attribution at the global scale. The authors already cite Emmons et al. (2012) elsewhere in the paper, but do not mention this work in Section 2.1, where it would be appropriate to have some discussion of how these methods differ, and how this might influence the results of the study. One very important difference is that Emmons et al. (2012) only consider NOx as a precursor of ozone, while the technique employed by the authors combines the effects of both NOx and VOC precursors. Similarly, the study of Butler et al. (2018) is also missing from the discussion. Butler et al. (2018) account for effects of both NOx and VOC as ozone precursors, but they make some very different design decisions to the technique employed by the authors. The authors must do more to put their method in the context of the previous work, and discuss the relative strengths and weaknesses of the approach they have chosen.

Also in Section 2.1, the authors could briefly mention how stratospheric ozone is tagged in their approach, since this does not fit into the framework of their Equation 2.

Section 2.2: The authors should make it clear that the tags are applied globally, with no distinction between emissions in Europe and the rest of the world. This is acknowledged later in the manuscript, but the reader would benefit from having this made clear already in this section.

Page 10, lines 17-24: For some additional context here, it would be nice to know how the proportional contributions of land transport to ambient modelled NOy compare to the proportional contribution of land transport to total NOx in the inventories. Is the contribution as would be expected from simply looking at the emissions, or is it disproportionally higher or lower?

Section 4.1, page 15: The authors rightly interpret the ozone due to land transport in DJF as coming from long-range transport. I also understand that the limits of the experimental design (one global tag for land transport) make it hard to say anything about long-range transport in JJA, when local photochemistry is more active. But could it be possible to try? For example, could they look at the the land transport contribution at the western boundary of the refined grid in JJA, and use this as a rough estimate of the contribution of land transport (and other sectors) in remote regions to baseline ozone in Europe? This could add a lot of value to the study and would be highly relevant for international policymaking.

Page 16, line 3: the seasonal cycle of photochemical activity also plays a role here.

Page 16, line 8: is there any way in this study to separate the influence of soil NOx and biogenic VOC? Or are these two different sources inextricably joined together into the "biogenic" sector?

Section 4.2: As mentioned earlier, it would be nice to know how well the model is capable of reproducing the extreme values of ozone as measured. If the model is doing a good job at this, then the results reported here could help to understand these extreme ozone measurements. If the model is not doing well at this, then the results reported here could potentially provide information about systematic model biases, and point the way towards improving the model. As it currently stands, it is not clear at all how these results should be interpreted.

Page 19, line 11: The region "Alps" includes the Po Valley. Does this mean that high mountains are in the same region as a polluted valley? The influences on air quality would be expected to be very different in these regions. High mountains will be more influenced by the free troposphere (and long-range transport), while the valley will be more influenced by local sources. Furthermore, "Alps" and "Po Valley" are used individually in this section and elsewhere in the manuscript. It is not always clear which region is meant. The authors could consider disaggregating this region into two subregions for their analysis (which could be quite informative), or at least being clearer about exactly which region they are referring to throughout the text.

Page 19, lines 15-16: the discussion about "uncertainties" in the inventory is very vague here. Could the large range in the contribution of land transport to extreme ozone when using EVEU emissions be related to the higher spatial heterogeneity and existence of more "hot spots" in this inventory compared with REF? There could potentially be some important information here about the need to get the distribution of NOx right in order to capture the high ozone events. A comparison of the REF and EVEU ozone timeseries with some measurements from urban background stations during extreme events could potentially add a lot of value here.

Page 23, line 4: the results are not "rather similar", but actually have some important differences, which are subsequently discussed. I think what the authors are trying to say here is that the contribution of land transport is similar in each case, but this is not the meaning which comes across.

Page 25, lines 25-26: This sentence basically conveys no meaning and could be easily deleted with no loss to the manuscript. Alternatively the authors could try to be clearer about what they mean here.

Page 25, last paragraph: if the previous work only accounts for the contribution of European land transport emissions to European ozone, and the current study also includes global emissions, then shouldn't the current study result in a higher contribution than the previous work? The opposite appears to be the case. Can the authors explain this apparent discrepancy?

Page 26, line 23: the authors appear to be concluding from the strong influence of the "biogenic" sector that soil NOx emissions are strongly influencing ozone. But couldn't this also be biogenic VOC? How do they separate the influence of these two different sources? A comparison with Butler et al. (2018) could be instructive here, since in that study the separate roles of NOx and VOC as ozone precursors were examined. Comparison of their Figure 3 and Figure 4 indicates that biogenic VOC make a larger contribution to European ozone in summer than biogenic NOx. The authors should discuss this here.

Page 28, lines 14-15: the future work proposed by the authors would indeed be extremely interesting from a policymaking perspective. If possible, they should also include as many other sectors as possible. This could help to inform decisions about

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Discussion paper

where emission reductions would be most effective.

Page 28, lines 28-29: again, it appears that the authors are over-interpreting their results when they conclude that soil NOx has a strong influence on European ozone levels.

Additional References

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