# Responses to the comments of anonymous referee #1

We would like to thank for the comments which helped to improve our manuscript. Please find below your comments in blue, our responses in black and modifications in the revised manuscript related to technical or specific comments in italic and inside quotes. In addition, we have updated the terminology of direct and indirect aerosol effects throughout the manuscript as well as in our replies to your comments, by replacing the term “direct aerosol radiative effect (DRE)” with “aerosol–radiation interactions (ARI)” and the term “aerosol indirect effects (AIE)” with “aerosol–cloud interactions (ACI)”. All modifications are highlighted in the revised manuscript.

The authors explore the summer surface O3 change over Europe in response to the potential aerosol emission change from 1990 to 2010 via their impact on direct radiation effect (DRE) using CAMx air quality model. They expand their study by taking into account the feedback of biogenic emission through the chain of aerosol emission change to radiation change and perform all studies at different chemistry backgrounds (i.e. base and high NOx emissions). This is an interesting study and the study content is suitable for ACP. I recommend publishing the paper after the authors make minor modifications suggested below.

General Remarks: It would be good if the authors could explain why it is better to use the designed emissions in 1990 instead of using the emissions in 1990 provided directly from emission inventories. The authors use emission inventory TNO-MACC-III for 2010. To derive the emission in 1990, they first analyze the observed PM10 between 90s and 2010 over 3 Netherlands stations and 3 Switzerland stations to infer potential enhancement factors of emission in 1990 verses in 2010. They then generate three potential emissions representing the situation in 1990 by increasing the emissions in 2010 with these enhancement factors. However, there are emission inventories, such as A2-ACCMIP that can provide emissions directly back to 1980. A2-ACCMIP is one of the multi-year emission datasets available from the international initiative AeroCom project for its second phase (A2) hind-cast model experiments (http://aerocom.met.no) (Chin et al., 2014). It might also be good to claim the study as a sensitivity study of O3 change in response to aerosol emission change via DRE. The impact of solar "brightening" on surface ozone is not limited to aerosol DRE. Aerosol indirect effect (AIE) is another potential pathway as the authors discussed in the paper. Although the authors indicated that AIE is not a driving reason for surface solar radiation (SSR) trend over Europe during the study period based on some previous studies, AIE is still a non-negligible factor as suggested by a recent study (Mian Chin 2018, personal communication). Furthermore, aerosol can impact ozone chemistry via heterogeneous reaction and the effects of photolysis and heterogeneous update are typically nonlinear (Bian et al., 2003). Feedbacks of O3 production and loss due to changes in O3 precursors leads to the complicated nonlinear feature.

We think that there was a misunderstanding about our approach to test the impact on ozone via the photolysis rates. The PHOT1–3 scenarios have the same emissions and meteorological input as the base case (BASE scenario). Therefore, we did not adjust any PM emissions in the three reported sensitivity tests, PHOT1–3. What we did was to adjust the PM only in the AOD calculations inside CAMx (in the in-line TUV), as described in Sect. 2.3.1. In other words, the induced forcing in the PHOT1–3 scenarios lies only in the AOD calculations. This approach excludes any potential impacts on the gas-aerosol chemistry, which would be the case if we would have adjusted the PM emissions or the PM concentrations generally in CAMx (and not just in the AOD calculation as we did). For example, consider the eq. (2) from the manuscript. Let $C$ be the PM concentration species for the base case (BASE scenario) and $p_f$ the adjustment factor that is applied to each PM species concentration. The product of $C \cdot p_f$ yields a new adjusted PM species concentration (that exists only in the AOD calculation) which we denote here as (referring to any of the PHOT1–3 scenarios) $C_{phot} = C \cdot p_f$. So, the actual concentration of PM species, $C$, does not change, except for secondary aerosols due to indirect impact of changes in the chemistry originating from the changes in the photolysis rates. However, we investigated this exception and we showed that the change in secondary aerosol is small (see Figs. S3 and S4) and any subsequent change in the AOD and the photolysis rates is expected to be negligible.

Although it was not our intention to use 1990 emissions in this study, we would like to point out that there are no reliable PM emissions in Europe before 2000 to use in regional models and those emissions that were estimated using various assumptions and gap-filling procedures for years before 2000 (for trend analyses) have very high uncertainties (Colette et al., 2016).

Specific comments 1. Page 1 lines 13-14 in Abstract: PHOT1, PHOT2, and PHO#3 is not in the effects of increased radiation on photolysis rates.

For a better clarification we reformulated the part of the sentence on page 1, lines 15–18 as follows:

*"… the effects of increased radiation between 1990 and 2010 on photolysis rates (with the PHOT1, PHOT2 and PHOT3 scenarios, which represented the radiation in 1990) and on biogenic volatile organic compounds (BVOCs) emissions (with the BIO scenario, which represented the biogenic emissions in 1990), …"*

2. Page 2 line 14: φ and σ depend not only on the gaseous species and air temperature, but also on air pressure for some VOCs.

We modified the sentence on page 2, lines 14-15 as follows:

*"φ and σ depend on the gaseous species and the air temperature T (K) as well as on air pressure for some species …"*

3. Page 2 line 15: Please add "O2, O3 and" before "water vapor". Please also add references of Bian et al., 2002 and Wild et al., 2000 at the end of this sentence.

Your suggestions were implemented in the manuscript (page 2, line 17).

4. Page 5 line 9: Is the two-weeks long enough spin-up for O3 chemistry?

Berge et al. (2001) showed that the impact of initial conditions on ozone is reduced to 10% or less with a spin-up time of approximately 3 days, while Jiménez et al. (2007) showed the same but for a spin-up time of 48 h. Other air quality modeling studies (Streets et al., 2007; Godowitch et al., 2008; Czader et al., 2012) focusing on ozone chemistry have used spin-up times between 2 and 10 days for simulating the summer season or summer episodes. Therefore, we are confident that the 2 weeks spin-up time was long enough.

5. Page 7 line 14: Could you elaborate on how to increase NOx emission?

As also stated in Table 1 in the manuscript, the $NO_x$ emissions were increased by a factor of 2 for each SNAP (Selected Nomenclature for Air Pollution) category based on the study of Oikonomakis et al. (2018). We added this clarification in Table 1.

6. Page 7 line 22: Do you really use the same emissions for both cases?

As also discussed above we used the same anthropogenic emissions and meteorology for all cases. For the PHOT1–3 scenarios we also used the same biogenic emissions, while for the BIO scenario the biogenic emissions were modified. We modified the sentence on page 8, lines 1–3 as follows:

*"In other words, we used the same meteorology and emissions for both cases (except for the BVOC emission sensitivity tests where we used different BVOC emissions) …"*

7. Page 9 line 29: It seems to me that you are not holding the aerosol coarse fraction constant during the period, but holding the absolute coarse model aerosol amount unchanged.

By "aerosol coarse fraction" we mean the particles with an aerodynamic diameter $2.5 < d \leq 10$. In CAMx, for the simulation of PM only two modes are considered: a fine one ($d \leq 2.5$) and a coarse one ($2.5 < d \leq 10$). The latter one we did not adjust/change in the PHOT1−3 scenarios and therefore it remained constant in line with our assumptions in page 9, line 29. In order to make our statements and assumptions more clear, we replaced the “aerosol coarse fraction” with “aerosol coarse mode”, wherever it was mentioned in the manuscript.

8. Page 10 lines 4-6: The criteria III should not be listed here since all PM2.5 data in this study are estimated based on PM10, while not measured.

The criteria iii is about the PM10 and PM2.5 data availability for 2010. In Sect. 2.2 we mentioned that both the $PM_{10}$ and $PM_{2.5}$ were provided by Airbase and NABEL networks and both products were measured at the respective sites.

9. Page 11 line 28: What aerosol components are included in FPRM?

FPRM (fine other primary) in CAMx refers to fine (<2.5 μm in diameter) primary particles other than those explicitly represented in the emissions and in the CAMx model. In the TNO-MAC-III emission inventory used in this study, the PM emissions are split to the following components: EC, OC, $Na^+$, $SO_4^{2-}$ and “Other minerals”. After the fractions of the first four species are calculated, the remaining part is assigned to the “Other minerals”, which contains other non-carbonaceous particles (Kuenen et al., 2014) and named as FPRM in CAMx.

10. Page 12 lines 18-20: To support the assumption in this study, the spatial distribution of AOD should be consistent with that of PM2.5, not PM10.

Ideally, we would also like to compare the spatial distribution of our simulated AOD changes with the respective ones of $PM_{2.5}$ concentrations. However, in Colette et al. (2017) only the simulated spatial distribution of the $PM_{10}$ concentration changes was shown. On the other hand, as discussed in Sect. 3, it was suggested that the decreasing trend in $PM_{10}$ concentrations was dominated by the reductions in the $PM_{2.5}$ concentrations (Barmpadimos et al., 2012; Tørseth et al., 2012), and thus we expect that the spatial distribution of the changes in $PM_{10}$ concentration to be similar to that of $PM_{2.5}$ concentrations. Moreover, Tørseth et al. (2012) showed that the geographical distribution between the changes in $PM_{10}$ and $PM_{2.5}$ concentrations, for the period 2000–2009, was similar for several EMEP monitoring sites. Therefore, we believe that the spatial distribution of the changes in $PM_{10}$ concentrations is a good proxy for that in $PM_{2.5}$ concentrations, especially for the areas which are not strongly influenced by sea salt and African dust particles.

11. Page 13 line 22: Where is the J(NO2), surface, column?

The $J(NO_2)$, is reported at ground-level. We made the following modifications in the text and figure captions:

page 14, line 4: “*The simulated (in the base case) ground-level photolysis rate of* $NO_2$, $J(NO_2)$, *…*”

page 14, lines 5–6: “*The modeled continental mean absolute (relative) differences in ground-level* $J(NO_2)$ *…*”

page 14, lines 7–8: “*The spatial distribution and relative changes are the same for the ground-level photolysis rate of* $O_3$, $J(O_3 \rightarrow O^1D)$, *…*”

caption of Figure 6: “*Seasonal daytime (10:00–18:00 LMT) mean* $J(NO_2)$ *at ground level …*”

caption of Figure S3 in the Supplement: “*Seasonal daytime (10:00-18:00 LMT) mean* $J(O_3 \rightarrow O^1D)$ *at ground level …*”

12. Page 13 line 20 to Page 14 line 2: Other chemistry and physics processes may join with the photochemistry to impact tracer change in a nonlinear way.

That is true, but in this study with the PHOT1–3 scenarios we investigated and isolated only the impact of aerosol–radiation interactions (ARI) on the chemistry and only via photolysis rates, excluding the impact of ARI on meteorological processes.

## References

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