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Interactive comment on "Analysis of European ozone trends in the period 1995–2014" *by* Yingying Yan et al.

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Referee #1

The manuscript investigates the European mean, 5th and 95th percentile daily, daytime and nighttime ozone trends between 1995 and 2014, using surface observations from the EMEP network and the EMAC model. The manuscript is well written and organized and the level of the English language is good. It is suitable for publication in ACP after addressing the minor issues I have listed below.

We thank the reviewer for comments, which have been incorporated to improve the manuscript.

General Comments

1. Why do the authors use only EMEP stations? There is also other networks available such as AirBase so that there can be an urban background vs. regional investigation of the ozone levels. I am aware that the EMAC model on a coarse resolution is not suitable to investigate the observed trends but limiting it only to the observations would I think increase the value of the paper.

In the revised manuscript, we have added the Airbase data to analyze the ozone levels and changes over rural, suburban and urban sites (Fig. 2), and also incorporate these results in conclusion.

In the revised Sect. 2.1, we have added the Airbase data selection: "As the measurements from EMEP network are carried out under the "Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe", the monitoring sites are located where there are minimal local influences, and consequently the observations are representative of relatively large regions (Torseth et al., 2012). In order to compare the observed ozone levels and changes over urban, suburban and rural sites, we also use the hourly measurements over 1995–2012 from the European Environment Agency Airbase system (https://www.eea.europa.eu/data-and-maps/data/airbase-the-european-air-quality-database-8#tab-figures-produced; available years: 1973–2012) (Schultz et al., 2017). After applying the same data selection criteria above, we get a total of 685 sites (289 for urban, 150 for suburban and 246 for rural)."

In the revised Sect. 3.1, we have added the Airbase ozone data analysis: "Annual and seasonal mean daytime and nighttime ozone mixing ratios averaged over the EMEP sites and Airbase sites are shown in Fig. 2. Ozone mixing ratios are maximum over the spring-to-summer season and minimum over the fall-to-winter season for different type of station classification. For annual mean ozone, the concentrations both in daytime and at night over rural sites (EMEP sites and Airbase rural sites) are higher than those averaged over the Airbase suburban and urban sites. Although the EMEP (93 sites) ozone and Airbase rural (246 sites) ozone are calculated based on different number

of sites, the ozone trends (shown in each panel in Fig. 2) for annual and seasonal means are similar both during daytime and at night. For the Airbase suburban and urban sites, ozone has increased rapidly with the statistically significant growth rates of 0.09–0.83 μg/m3/y, except that a decline of -0.19 μg/m3/y (P-value < 0.01) is also visible for suburban summer ozone during 1995–2012. These suburban and urban ozone enhancements (0.20–0.59 μg/m3/y for annual means; P-value < 0.01) contrast with the slight rural ozone decrease (-0.09 – -0.02 μg/m3/y for annual means; with an increasing trend for winter ozone and a decreasing trend for summer ozone). As the EMAC model version used here has a coarse resolution, which is not suitable to investigate the observed contrast ozone trends among the urban, suburban and rural stations, we focus on the analysis of ozone levels and changes over the regional background areas monitored by EMEP network in the following results."

2. Although the model is well-documented, I think a little more information can be provided for the model properties influencing ozone such as the chemical scheme. Also, more information on how the emissions are used in the model can be useful. Finally, other natural emissions such as dust, sea-salt as well as biomass burning must be explained. The biomass burning during summer time in southern Europe can have significant impacts on O3 levels, which can explain some year-to-year variability.

In the revised Sect. 2.3, we have added the information of chemical scheme: "The chemical mechanism in the simulations considers the basic gas-phase chemistry of ozone, odd nitrogen, methane, alkanes, alkenes and halogens (bromine and chlorine). Here we use the Mainz Isoprene Mechanism (version 1; MIM1) to account for the chemistry of isoprene and additional non-methane hydrocarbons (NMHCs). This mechanism in total includes 310 reactions of 155 species and is included in the submodel MECCA (Jöckel et al., 2010; R. Sander et al., 2011)."

Also more emission information has been shown in the revised Sect. 2.3: "Anthropogenic and biomass burning emissions in the model are incorporated as prescribed sources following the Chemistry-Climate Model Initiative (CCMI) recommendations

(Eyring et al., 2013), using the MACCity (Monitoring Atmospheric Composition & Climate/City Zero Energy) emission inventory, which includes a seasonal cycle (monthly resolved) for biomass burning (Diehl et al. 2012) and anthropogenic emissions (Granier et al. 2011). Additionally, the emissions are vertically distributed as described by Pozzer et al. (2009). Since the total NMVOCs (non-methane volatile organic compounds) values for anthropogenic sectors are not provided by the MACCity raw dataset, they are recalculated from the corresponding species (Jockel et al., 2016). Emissions from natural sources have been prescribed as well, either as monthly resolved or annually constant climatology. The spatial and temporal distributions of biogenic NMHCs are based on Global Emissions InitiAtive (GEIA). In addition, the emissions of terrestrial dimethyl sulfide (DMS), volcanic SO2, halocarbons and ammonia are prescribed mostly based on climatologies. The ocean-to-atmosphere fluxes of DMS, C5H8, and methanol are calculated by the AIRSEA submodel (Pozzer et al., 2006) following the two-layer model by Liss and Slater (1974). The emissions of soil NOx (Yienger and Levy, 1995;Ganzeveld et al., 2002) and biogenic isoprene (C5H8) (Guenther et al., 1995;Ganzeveld et al., 2002) are calculated online using the submodel ONEMIS. The lightning NOx emissions are calculated with the submodel LNOX (Tost et al., 2007) following the parameterization by Grewe et al. (2001). This scheme links the flash frequency to the thunderstorm cloud updraft velocity. Aerosols are included in the simulation, although their heating rates and surface areas (needed for heterogeneous reactions) are prescribed from an external climatology rather than interactive chemistry. Further details of the model setup on the emissions, physical and chemical processes as well as the model evaluation with observations can be found in Jöckel et al. (2016)."

3. It would also be interesting to show the spatial evaluation of the MAC model and discuss if there are regions with higher biases than others and why.

In the revised Sect. 3.3, we have shown the spatial evaluation of EMAC modeled ozone with the revised Fig. 1: "Fig. 1 also shows the spatial distribution of observed and modeled mean ozone mixing ratios, as well as the modeled biases for every five

years during 1995-2014 over the selected 93 sites. It is shown that for most monitoring stations the model overestimates the observed background ozone concentrations with the bias up to 15 μg/m3. Ozone overestimation has been observed also in other EMAC simulations when compared to satellite data (Jöckel et al., 2016). Relatively frequent overestimations (> 10 μg/m3) occur over the coastal and marine sites where the coarse model resolution mixes the polluted air over land with cleaner air masses. Underestimation of modeled ozone also occurs over several sites located at the central Europe. These simulated ozone underestimations are probably due to the underestimation of precursor emissions (especially NOx) discussed by Oikonomakis et al. (2017)."

Specific Comments

Lines 107-109 is a repetition of lines 94-96.

We have removed the lines 107-109.

Section 2.2. lacks motivation for why these analyses will be done for, although it is obvious. I think few lines would improve the flow and readability of the section.

We have revised the first sentence in Sect. 2.2: "To help investigate the underlying effects of climate variability on ozone variations and trends, we relate the monthly variability of ozone to 2-meter temperature relevant to the European ground-level meteorology."

Lines 147-149: Please write here explicitly how the emissions are kept constant? Are they fixed to 1995 or the mean of the period etc: : :?

This sentence has been revised: "We also conducted a sensitivity simulation in which the anthropogenic emissions were kept constant (at the 1994 levels), to represent a scenario with fixed emissions throughout the years where observations are available to investigate the effects of emissions on ozone trends."

Line 190: The supplement figure should be referred here.

We have added the supplement figure (Fig. S6) reference in the revised sentence.

Line 231: the trends written in the text are slightly different than those on the plots, please double check.

We have modified the trends in the text.

Check the alphabetical order in the Referece list.

We have rearranged the reference list according to the alphabetical order.

Krotkov et al. (2016) is missing in the text.

We have added this reference in the text.

Langner et al. (2004) is missing in the reference list.

We have added Langner et al. (2004) in the reference list.

Change Lelieveld et al. (2000) with Lelieveld and Dentener (2010).

We have added the reference: Lawrence and Lelieveld (2010).

Fig. 1. It would be more interesting to see the e.g. annual mean O3 distribution rather than the surface elevation.

The revised Fig. 1 have added to show mean ozone mixing ratios for every five years during 1995-2014 over the selected 93 sites.

Legends should be added to all figures with time series plots.

We have added legends in the time series plots.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2017-1077/acp-2017-1077-AC1-supplement.pdf

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