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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 #2

General comments

This paper by Yingying et al., investigates long-term trend in near-surface ozone in Europe by analysed observations part of the EMEP network. Moreover, it provides some very interesting hints about the different weights that change in European anthropogenic emission and "climate" variability have in determining the observed long-term tendencies. The paper is well written and within the goal of ACP, the topic is more than relevant. Here, I addressed a few major and minor points that must be considered before final publication in ACP.

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

MAJOR POINTS

1) One major point that must be carefully addressed by authors is the statistical significance of the tendencies reported in the paper. As an instance the Mann-Kendall test must be applied to the different subset of data to verify the actual existence of a “trend”. Otherwise, the authors can only discuss about “tendencies”. It is questionable to discuss and attribute tendencies that are not statistically significant, i.e. not different from zero. As an instance, statistical significance of tendencies/trends must be indicated in Table 3.

Thanks for the suggestion. In the revised manuscript, to address the statistical significance, all trends in the text and tables (Table 2 and Table 3) are performed with an F-test at the 95% confidence level.

2) By reading the paper is not clear to me how the authors aggregate data. Are the monthly percentiles (line 96-98) the average of the corresponding percentiles at each single station or the percentiles obtained for the whole data set (i.e. by considering all the ozone data observed at the 93 stations) for each specific month? I think the first “metric” would be much more robust that the second: : :

Yes, we calculate the monthly percentiles with the first method above to get the corresponding percentiles at individual station in each month. We analyze the ozone trends and variation for different percentiles at each station (Fig. 5, Fig.11, Fig. 12, and Fig. S2). Averaged over the 93 sites, we then also calculate the trends of different percentile ozone concentrations over the whole Europe. To make it clear, this sentence has been revised: “The monthly 5th, 50th and 95th percentile ozone concentrations for each period (per hour, daytime, nighttime and diurnal) are derived from the lowest, middle and highest 5th percentile hourly ozone mixing ratios of the corresponding period at individual stations in each month. Averaging over the 93 sites, we then also calculate the trends of different percentile ozone concentrations over the whole Europe."

3) The analysis concerning the impact of climate variability is promising but it need more attention: it is not novel that near-surface O3 respond to air-temperature (used as proxy of meteorological conditions favorable for photochemical production and accumulation). I would see a more deep discussion (and possibly analysis, see my comment about Fig S9) about the specific processes underlying this "climate variability". The authors mentioned (and reported by Figure S9) an influence of NAO but without any specific comments/explanation (I also suggest to discuss possible implication of NAO to air-mass transport regimes). As suggested by the Referee#1, biomass burning occurring at continental scale is an issue for near-surface ozone, especially under heat-wave or dry conditions. A cross-correlation analysis with number or geographical distribution (burned area) of open forest fire numbers can be useful to assess this point. For a large subset of year (i.e. since 2000), MODIS data can be used.

We have moved and revised the Fig. S9 to the main text and discussed more deeply in the revised Sect. 4.2.2: "Fig. 11 shows the correlations between the monthly mean 2-meter temperature and the monthly mean, 5th and 95th percentile ozone for diurnal, daytime and nighttime concentrations. Most of these site-by-site correlations are statistically significant (P-value < 0.05 under a T-test; shown as triangles in Fig. 11) with high fraction (66%–91%) of sites for which significant correlation exist. For each metric (mean and percentiles for diurnal, daytime and nighttime), it corroborates the high correlations over central Europe with statistically significant values up to $\sim$0.82 (P-value < 0.01). It indicates that the surface ozone mixing ratios are highly sensitive to enhanced air temperature, being favorable for photochemical O3 production, which has been reported by previous studies (Lin et al., 2017; Yan et al., 2018 and references therein). For different seasons, ozone variations in fall are most closely affected by temperature (Fig. S9), followed by the spring and summer ozone. The weakest linkage between ozone and temperature is in winter with few sites for which significant correlation exist especially for 95th percentile. In contrast to the positive cor-

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relations over central and southern stations, ozone concentrations over the northern and western sites are negative and significantly correlated with temperature, associated with statistically insignificant correlations at several sites located in the transition regions from positive-correlation to negative-correlation (Fig. 11). This may be related to the influence of the Northern Atlantic Oscillation (NAO; a dominant mode of winter climate variability in the North Atlantic region including Europe; higher correlations with ozone in winter shown in Fig. S11), which had an opposite impact on ozone over northern and western compared to central and southern Europe (Fig. S10). This is because the positive NAO phase is associated with enhanced pressure gradient between subtropical high pressure center (stronger than usual) and Icelandic low (deeper than normal). It can result in more and stronger winter storms crossing the Atlantic Ocean on a more northerly pathway, and consequently lead to warm and wet air in northern Europe. Compared to the impact of temperature, the effect of NAO on ozone are relatively modest with much lower correlations (Fig. 11 and Fig. S10). The correlations of less than 30% of the sites pass the significance test (P-value < 0.05). These results underscore that the large-scale climate variability affects the inter-annual variability of European background ozone."

Thanks for the suggestion of necessary discussion in biomass burning. Many previous studies have shown the linkage between forest fires under heat-wave and surface ozone. Here we have added this discussion in the revised Sect. 4.2.1: "Especially, in August 2003, coinciding with a major heat wave in central and northern Europe, massive forest fires were observed from the Terra and MODIS satellite in many parts of Europe, particularly in the south and most pronounced in Portugal and Spain (Pace et al., 2005; Hodzic et al., 2006, 2007; Solberg et al., 2008). Long-range transport of fire emissions have been found to give rise to significantly elevated air pollution concentration and proved to be contributed to the European ozone peak values in August 2003 (Solberg et al., 2008; Tressol et al., 2008; Ordóñez et al., 2010)."

4) Line 394-395: the role of China emissions (even if reasonable) is not supported by

data or analysis in this paper. If not strong evidences are added, this statement must be strongly understated or presented with much more caution. I'm wondering if you can use EMAC to make sensitivity study on China emission trends by playing with the MACCity inventory:

We agree with the referee that the sentence is not accurate. Although a sensitivity run can be performed with the EMAC model, we believe this to be out of the scope of this manuscript. Many studies have shown the impact of intercontinental transport on European ozone (i.e. Derwent et al. 2008, West et al. 2009a and West et al. 2009b). However, how claimed by the referee and shown by the aforementioned studies, emissions from other regions have larger impact on European ozone than Chinese emissions. Therefore we reformulated the sentence as: "Slower rates of ozone reduction during nighttime are suggested to be combined effects of reduced titration due to lower NOx emissions, and an increase in the global background ozone concentrations during this period, probably due to growing precursor emissions worldwide since 1995, which has been predicted by Lelieveld and Dentener (2000) based on atmospheric chemistry – transport modeling, and corroborated by satellite observations (Richter et al., 2005; Krotkov et al., 2016)." We also revised the conclusions.

SPECIFIC COMMENTS

Line 71: annual "surface" 5th...maybe "surface" ozone concentration?

We have modified this sentence.

Line 96: please, better elucidate the aggregation process to obtain the calculated percentiles

Have revised; please see our response to major comment 2.

Line 170: which is the number sites characterized by negative trend ?

We have added this information: "For the ozone trend of 95th percentile at individual station, 84 sites (90%) are characterized by decreasing trend in daytime and 78 sites

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(84%) at night (Fig. 5 and Fig. S2).”

Line 187-190 and Table 3. Are these tendencies/trends obtained by averaging single trends/tendencies at each station or what else? Please specify.

These trends are calculated with the 5th, 50th, and 95th percentile ozone concentrations averaged over the 93 sites, not obtained by averaging trends at each station. We have specified in the revised text.

Line 202. Some comments are due to the absence of diurnal cycle for 5th percentile in winter. I would expect a diurnal cycle in NOx anthropogenic emissions that can affect O3 diurnal cycles and subsequently its trends: : :

Thanks for the suggestion. We have updated this sentence: “The slight growth rates in the 5th percentile ozone are approximately equally distributed at the level of 0.1 ± 0.12 μg/m3/y (P-value > 0.05), probably due to the absence of ozone diurnal cycle, affected by NOx anthropogenic emissions, for 5th percentile especially in winter.”

Line 214: did you calculate the average of trends or trend of averaged ozone over the whole Europe. In this latter case, you put together sites with very different inhomogeneous in term of ozone variability. As an instance, in summer, ozone is strongly dependent by geographical regions and latitudes: : :This is also evident by your Figure 4.

Here the trends are calculated with the averaged ozone over the whole Europe. To show the different inhomogeneous in term of ozone variability at different stations, we calculate the spatial standard deviation of trends following the last function in the revised Sect. 2.1. The ozone trends at each station and the average of trends are also show in Fig.5 and Fig. S2.

Line 233: the annual trend for emep stations here reported are different from those in Table 3. Why?

Here the annual ozone trends (Fig. 5) are the average of trends at each station. The trends in Table 3 are calculated with the averaged ozone over the 93 sites.

Line 234-235:please comment these geographical differences and provide possible reasons

We have added the possible reasons: “These geographical differences in ozone trends are probably explained by the effects of a general decrease in European anthropogenic precursor emissions, being partly offset by those of climate variability (see Sect. 4.2 for discussion of Fig. 11 and Fig. S10).”

Line 237: what do you mean with “regional trend contrast”. Contrast in respect to what?

Here “regional trend contrast” means the geographical differences in ozone trends. We have updated this sentence: “The geographical differences in ozone trends are most significant in spring with an average growth rate of 0.01 μg/m3/y (Fig. 5).”

Line 251: why did you investigate the correlation with 95th percentile? What do you want to proof?

We calculate the correlation between the exceedances and 95th percentile ozone to show their interannual consistence.

Line 275: is the trend overestimation (especially for 95th percentile, i.e. lower decrease with time) due to the O3 overestimation since 2010?

Thanks for the suggestion. Yes, the ozone overestimation since 2010 may be the dominant reason for the trend overestimation. We have added this comment in the revised text.

Line 297: what the reason of the enhanced trends in the 5th percentiles?

We have added the possible reason: “The possible reason for these simulated enhanced ozone trends is the overestimation of the decline of European anthropogenic ozone precursor emissions (decreasing more rapidly than observed) in EMAC.”

Line 359: Figure S9 need to be shown in the main body of paper and it deserve more attention/comments/explanation. As an instance, what the possible impact of NAO variability to transport regimes?

We have moved this figure to the main text and please see our response to major comment 3 for detail discussion.

Figure S8-S9: please identify the sites with statistically significant correlation and provide in the paper the fraction of sites for which significant correlation exist for each metric (mean, percentiles) with T and NAO.

In the revised Fig.11 and Fig. S10, we have identified the sites with statistically significant correlation and also shown the fraction of sites for which significant correlation exist for each metric (mean, percentiles) with T and NAO in the figures. We have also discussed it in the text; please see our response to major comment 3.

Line 352: are these correlation calculated over the 20-yr period? Since NAO effect are strongly dependent by season (see Pausata et al., ACP, 2012), Fig S8 and S9 should be disaggregated as a function of different seasons.

Yes, these correlations are calculated over 1995-2014. We have shown the seasonal correlations in the revised Fig. S9 and Fig. S11 and also added some discussion in the revised Sect. 4.2.2.

Line 379: it may be useful if the fraction of sites with statistically significant trends is provided.

Have added: “Results show that although reductions in anthropogenic emissions have lowered the peak ozone concentrations (sites with statistically significant trends: 91 out of 93 sites; 98%), especially during daytime in the period 1995–2014, the lower level ozone concentrations have increased (sites with statistically significant trends: 71 out of 93 sites; 76%) continually since 1995 over Europe.”

In the "Conclusion section" it should be stressed that 20-yr is a time frame too short

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for depict climate tendency (formally a 30 yr period is necessary). I agree that some “large-scale” processes like NAO can influence near-surface O3, thus possible change of these regimes under a changing climate can have serious impact on ozone.

We have added this discussion in the revised conclusion: “We note that our analysis over 1995–2014 is a timeframe too short for the analysis of climate tendencies (formally a 30-year period is necessary). Thus, here the climate related variability is mainly driven by the large-scale processes like NAO and heat wave occurrence, which may be influenced by climate change.”

Reference: Derwent, R. G., et al. "How is surface ozone in Europe linked to Asian and North American NOx emissions?." Atmospheric Environment 42.32 (2008): 7412-7422. West, J. Jason, et al. "Effect of regional precursor emission controls on long-range ozone transport–Part 1: Short-term changes in ozone air quality." Atmospheric Chemistry and Physics 9.16 (2009): 6077-6093. West, J. Jason, et al. "Effect of regional precursor emission controls on long-range ozone transport–Part 2: Steady-state changes in ozone air quality and impacts on human mortality." Atmospheric Chemistry and Physics 9.16 (2009): 6095-6107.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2017-1077/acp-2017-1077-AC2-supplement.pdf

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