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Interactive comment on "Evaluation of near surface ozone over Europe from the MACC reanalysis" *by* E. Katragkou et al.

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We thank the reviewer for his/her comments. We responded to all of the points raised and changed the revised manuscript accordingly. Attached below our detailed response.

Main comments R2.1 "It is not clear to me why the authors do not make more use of the CTRL simulations in their comparisons. While the CTRL simulation does not extend to 2012 like the MRE, comparison of the MRE statistics for 2003-2012 (table 1) and 2003-2010 (table 2) suggests little difference when the extra 2 years are included. If the authors restricted all their analysis to the common 2003-2010 period, they could add the CTRL results to the seasonal cycle plots. I would be interested in seeing this, particularly as the CTRL simulation's seasonal cycle agrees better with observations

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(at least marginally). Might an extended comparison of MRE against CTRL hint at further drivers of observation/MRE discrepancies?"

This paper was thought to be an extended evaluation of the reanalysis product with respect to near surface ozone. For anyone wishing to have this paper as a reference for the evaluation of the ECWMF reanalysis product, we believe it would be better to include the whole period of the reanalysis (2003-1012). In the current manuscript we provide the basics of the comparison between the ctrl and the reanalysis (Fig 3 and Table 2). The extended report on the impact of assimilation on surface ozone from 2003 to 2010 is available as a VAL technical report, Deliverable D84.2 “Validation report on the Comparison of surface ozone in the global (2003- 2010) and regional reanalysis (2011) over Europe”. The basic findings of our analysis are robust and do not depend on the length of the period examined.

R2.2 "The language is often imprecise when discussing the comparisons. For example, what are “acceptable temporal correlations” (P1085, L20)? What does “reasonably well” (P1091, L2) mean? In addition, for the discussion on CTRL vs MRE, if r = 0.74 to 0.49 is “slightly reduced” (P1087, L10), what should we make of the bias improvements discussed on P1087, L1-5?"

In response to this comment we modified the manuscript accordingly: i) “acceptable temporal correlations” the expression was removed and the discussion on the correlations was rewritten. ii) The sentence was rewritten: “Comparison with ozonesonde measurements at different locations (Fig. 7) indicate that MRE ozone profiles reproduce the basic structure of the profile, overestimating in most cases ozone below the 850hPa.” iii) The word “slightly” is deleted and the reduction of R in the assimilated experiment is discussed more thoroughly.

R2.3 "The font size on the figures is too small and there are often too many panels to give a useful picture of what’s going on (esp. fig 5 and fig 7). In addition, the authors could consider plotting the biases and correlations on maps like Figure 1 (e.g. coloring

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the dots by the r and FGE values)."

New plots are provided in the revised manuscript with larger fonts.

Specific comments (including technical corrections)

P1078, L24: “and MACC-II: : :”

Done

P1079, L8: Define ECMWF (you do it for all other acronyms)

Done

P1079, L12: year-long -> long

Done

P1079, L20: “(AQME)”

Done

P1079, L25-: Near surface ozone is not very important as a GHG

We remove the sentence referring to tropospheric ozone and its role as GHG.

P1080, L2-: Mention chemical loss and deposition

The sentence “It can be destroyed photochemically or by dry deposition at the surface” was added.

P1080, L7: “soil, vegetation”

Done

P1080, L25: “Monks, 2000”

Done

Section 2.1: Would be useful to know which species/observations are assimilated

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which are relevant for ozone

The ozone data assimilated in MRE are listed in Table 1 of Inness et al. (2013). A comment has been inserted in the revised manuscript.

P1081, L18: Define “variational bias correction”

In the variational scheme biases are estimated during the analysis by including bias parameters in the control vector. The bias corrections are continuously adjusted to optimize the consistency with all information used in the analysis.

P1081, L19 (and several other places): Mind that -> Note that (former sounds like an admonishment)

The sentence is re-written: “The assimilation correction on ozone is due to the stratospheric and total ozone column.”

P1082, L9: What are “types 1-3”? Need more detail

Joly and Pech (2012) used Linear Discriminant Analysis on the pollution measurements of the AIRBASE network to discriminate the rural stations from the most polluted, urban and traffic stations. This statistical process is specific for its measured pollutant and using nine percentiles from 10% to 90% as fixed arbitrary thresholds, ten classes have been defined. The first three classes for the case of O3 discriminate reasonably the stations with rural characteristics that are the most representative of the large scale. ‘Types 1-3’ are corrected to ‘classes 1-3’ in the manuscript.

P1082, L14: “corresponding observational data” (data can be from a model too)

Done

P1082, L26: Is there any rationale for these regions beyond geographical convenience? E.g. can you demonstrate that similar processes control ozone in each region. This is important for making inferences about the regional statistics.

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We agree with the reviewer that this is an important issue, when averaging stations within a region. Our regional selection is arbitrary. Indeed the geographical convenience was our initiative to split the regions and the fact the similar regions have been used in previous climate-oriented studies (Christensen, J. H. and Christensen, O. B.: A summary of the PRUDENCE model projections of changes in European climate by the end of this century, Clim. Change, 81, 7–30, doi:10.1007/s10584-006-9210-7, 2007).

It has not be used a statistical cluster analysis to objectively discriminate regions with distinct ozone characteristics. This is part of our on-going work. However, in the case of ozone, there are a number of difficulties for an objective way to discriminate regions with distinct characteristic from station data. This is because even within a small region with similar large scale ozone features, the stations may differ significantly in terms of the ozone behavior depending on the distance from sea, the elevation and the distance for pollution sources. This becomes even worse for regions with small number of stations. A nice example is the case of Mediterranean with a small number of stations and with different ozone characteristics of the maritime rural EMEP stations from the continental rural classified AIRBASE stations. In our analysis we did a geographical compromise, when calculating ozone averages.

P1083, L1 (and Fig 1): Could you indicate the region codes on the figure? Additionally, using the full name for the region in the text makes for easier reading (there are some instances where just “BI” etc are used)

Done: Region codes are explicitly indicated in Figure 1 caption.

P1083, L16-21: This paragraph could do with re-phrasing and making into <1 sentence.

The sentence was split as shown below: "We have also to take into consideration that the NOx observations are affected strongly by local emissions. Furthermore there are known issues with interference by oxidized nitrogen compounds (e.g. HNO3, PAN and other organic nitrates) for ground-based NO2 measurements by most commercially available NO2 instruments using molybdenum converters, hence leading to an

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overestimation of NOx concentrations (Steinbacher et al., 2007)."

P1085, L3: Be clear that you're using R to refer to the seasonal cycle, rather than correlating the whole time series (or time series of DJFs etc). The latter might be interesting though to investigate interannual variability.

Done. "Seasonality" was corrected to "interannual variability".

P1085, L5: SD -> standard deviation (at least the first time)

Done

P1086, L3-6: This information can go in the caption

Done

P1086, L11-: This is repeating the point in L8 (i.e. not "On the other hand")

Done. The first sentence was deleted.

P1087, L27: however, -> but

Done

P1088, L3 (and throughout): depicted -> found

Done

P1088, L14: revealing -> causing (?)

"Revealing" changed to "causing"

P1089, L18: "captures the shape: : :cycles quite well, but with a: : :"

Done

P1090, L14 (and throughout): You've changed from "ozone" to "O3". I much prefer the former for easier reading.

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Done

P1091, L16: suggests that -> is consistent with (and then “being resolved: : :”)

Done

P1092, L5-: Whether an environment is “NOx-limited” will also depend on the mix of VOCs (their reactivity, propensity to form NOy etc), and presumably the VOC mix differs across Europe. We agree with the reviewer that the split between NOx-sensitive and VOC-sensitive conditions is correlated with the ratio of reactivity-weighted VOC mixture to NOx and this ratio differs across Europe. The sentence was modified accordingly.

“In global scale, nitrogen oxides (NOx) are the limiting precursors for O3 production throughout most of the troposphere, and also directly influence the abundance of the hydroxyl radical concentration in the troposphere (e.g. Crutzen, 1988). At regional scale for rural environments with NOx values less than a few parts per billion by volume, O3 formation is NOx limited (Liu et al., 1987) and therefore almost independent of hydrocarbon concentrations, depending of course on the ratio of reactivity-weighted VOC mixture to NOx which may differ from region to region across Europe (Beekmann and Vautard, 2010).”

Beekmann, M. and Vautard, R.: A modelling study of photochemical regimes over Europe: robustness and variability, Atmos. Chem. Phys., 10, 10067-10084, doi:10.5194/acp-10-10067-2010, 2010.

P1093, L1 -: Do you know that the NOx diurnal cycle is all chemical, with no transport? In general, these arguments might be more convincing if you were able to demonstrate them with (e.g.) a box model

We agree with the reviewer that this is an important issue. We used a box model with the CBIV chemical mechanism to calculate ozone production efficiencies for typical summer conditions using initial conditions for NOx and other gaseous species from the MACC model at BI, IP and ME. We calculated that 3 to 4 molecules of O3 are produced

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for every molecule of NOx oxidised at BI and ME, and up to 5 pbbv at IP. The above values agree well with ozone production efficiency estimates from previous studies for summer at rural semi-polluted sites with NOx more than a few ppbv in Europe and US (Chin et al., 1994; Derwent and Davis, 1994; Rickard et al., 2002). Furthermore, the box model estimated that ozone production efficiency values are comparable to the near surface ratio $\Delta O3/\ \Delta NOx$ ($\Delta O3$ increased over the day; ΔNOx decreased over the day) shown in Figure 10, which is roughly 3 for BI, 3.5 for ME and 10 for IP. Additionally, we have also estimated MRE $\Delta O3/\Delta NOx$ ratio values at 925 hPa (above near surface but within the atmospheric boundary layer) being roughly 3.5 for BI, 3 for ME and 4 for IP, which is in good agreement with the box model calculations.

Nevertheless, diurnal meteorological patterns of wind speed and boundary layer height, that lead to higher dilution of primary pollutants at daytime than at nighttime, may also contribute for the diurnal pattern of NOx in Figure 10 (see Figure 1, following the diurnal cycle of wind speed and boundary layer height). This is supported from the fact that CO which is a species with much longer chemical lifetime than NOx has a similar diurnal pattern with NOx (not shown in the manuscript but shown in the Figure below).

The text in Section 4.2 was modified accordingly and the relevant references were added (see below). The figure shown below was added as Figure S1 in the revised manuscript.

References Chin, M., Jacob, D. J., Munger, J. W., Parrish, D. D., and Doddridge, B. G., 1994: Relationships of ozone and carbon monoxide over North America, J. Geophys. Res. 99, 14,565–14,573.

Derwent, R. G. and Davis, T. J., 1994: Modelling the impact of NOx or hydrocarbon control on photochemical ozone in Europe, Atmos. Environ. 28, 2039–2052.

Rickard, A. R., Salisbury, G., Monks, P. S., Lewis, A. C., Baugitte, S., Bandy, B. J., Clemitshaw, K. C., and Penkett, S. A.: Comparison of measured ozone production

efficiencies in the marine boundary layer at two European coastal sites under different pollution regimes, J. Atmos. Chem., 43, 107–134, 2002.

P1093, L24: ": : :adequately capture the seasonality,: : :"

Done

P1095, L27: Final paragraph should be aligned to the left margin.

Done

Table 2: The FGE, MNMB and R should be centered above the MRE and CTRL columns.

This comment will be delivered to the Editorial Office, since all Tables and Figures are edited.

Interactive comment on Geosci. Model Dev. Discuss., 8, 1077, 2015.

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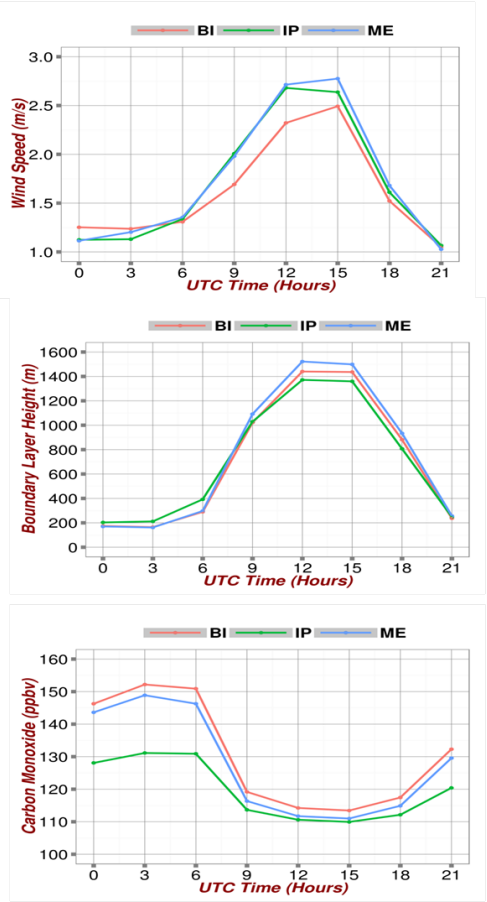

Fig. 1. Mean summer diurnal cycle of wind speed (m/s), boundary layer height (m) and near surface carbon monoxide (ppbv) for the sub-regions BI, IP and ME based on MRE.

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