**Atmospheric
Chemistry
and Physics**
Discussions

Interactive comment on "Quantifying the contributions of natural emissions to ozone and total fine PM concentrations in the Northern Hemisphere" *by* A. Zare et al.

Anonymous Referee #1

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The study of Zare et al is principally concerned with updating the DEHM chemistry-transport model with respect to various pre-cursor emission estimates, SOA formation and sea-salt generation. It should be stated that for some of the emission types (e.g. Wildfires) the choice of inventory is not optimal considering what is freely available to the modelling community from data portals at the current time. All model updates have been introduced simultaneously and there are a number of sensitivity studies performed to examine the 'natural' combined contribution towards PM2.5 distributions throughout Europe. Comparisons are made against measurements mainly in the US and Europe showing that even after the updates DEHM exhibits a low bias in PM

concentrations, although potential reasons are not discussed in much detail but simply listed. A second part of the paper then moves towards the contribution of different 'natural' emission sources on tropospheric ozone at the surface, which has already received attention in the literature over the past decade as part of many independent studies. There is no validation of the surface O3 distribution included therefore the reader is left guessing as to quality of the model performance when using non-yearly specific emission inventories for e.g. the wildfire component. The paper has 15 figures included making it quite long and the quality of writing is fair although the analysis is not robust enough in a number of places. For publication in ACP I would therefore like there to be more focus on the missing components towards explaining the PM2.5 bias at the expense of removing the tropospheric ozone component. One suggestion would be to utilise the additional sensitivity studies in terms of understanding and quantifying the most plausible explanations for the significant PM2.5 low bias in DEHM. This could also be extended for other regions using the e.g. AERONET network. I am finding it difficult to take away a new message from this manuscript or a direction in which emphasis should be applied to tackle the underestimation of PM concentrations which occur across a number of independent models. General Comments:

(i) The grammar should be checked throughout the manuscript as there are many instances where it is not correct. (ii) The use of ozone and O3 should be consistent throughout the manuscript (iii) The use of references could be significantly improved by using more recent studies especially concerning the new parameterizations, emission estimates introduced into DEHM and the many previous works concerning the influence of emission type on regional and global surface ozone distributions. (iv) The unit of ppb is associated with mixing ratios rather than concentrations which typically have the unit of μg m-3. (v) Although the method of introducing variability in biogenic emission estimates is sufficient the choice of averages for wildfire distribution and emission is not satisfactory for capturing the well documented inter-annual variability in wildfire emissions. Especially considering the importance of this emission source to PM2.5 distributions in e.g. Scandinavia (vi) Although SOA is introduced into the model is it

interactive with the photolysis via additional scattering and absorption of photolysing light? Please provide details. (vii) Wildfires are strongly influenced by agricultural practises in the tropics so this is cannot be defined as a Natural emission source as done throughout this manuscript.

Specific comments

Pg 16777, ln 4: Show that the exceedence of biogenic emissions over anthropogenic emissions occurs for the more recent emission estimates by updating references to e.g. Lamarque et al., 2010 and Guenther et al., 2012, which are used later on in the text. For what compounds does this occur?; ln 9: " ... are expected to change in the future ... "; ln 14-16: Update references e.g. direct comparisons of recent biogenic estimates for a number of BVOC species has been performed in Williams et al, 2013; ln 21: provide reference for modelling studies; ln 27/29: "natural O3" is O3 from natural sources? To help the reader please define what percentage changes are introduced.

Pg 16779, ln 7-8: Use capitals before acronyms.

Pg 16781, ln 2: The work of Price et al (1997) has been significantly updated in Ott et al (2010). For instance the estimate for the cloud-to-ground and cloud-to-cloud flash distribution has been modified for the northern hemisphere. Can the authors provide some reason for using the old method considering this is a recently implemented parameterization? Can the authors discuss what impact using a 8 year average for have on predicting the inter-annual variability in LiNOx during 2006. ; ln 8 The range given in Schumann and Huntrieser (2007) is 5±3 Tg N/yr rather than 1-20 Tg N/yr. Please correct.; ln 20: replace "the model" with DEHM; ln 25: How does the global annual emission compare to the estimates given in Steinkamp et al. (2009)?

Pg 16782, ln 3: why not use the NH3 anthropogenic estimates provided in Lamarque et al (2010) in order to maintain a consistent set of emissions; ln 13: what is the reference for the Wild Animal NH3 emissions totals and what is the corresponding value for Domestic animals? ln 15: replace "released" with emitted, plural of wildfire

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Pg 16783, ln 4-9: Most state-of-the-art global CTM's use monthly mean distributions from wildfires for specific years from e.g. GFEDv2. Why do the authors not use these readily available estimates for 2006? What type of injection heights are used for introducing fire emissions (e.g. Dentener et al., 2006)? The year 1997 had a strong signal due to El-Nino in the intensity of fires therefore may not be directly applicable to a more normal year such as 2006.

Pg 16784, ln 25: "...HTs, Henze and Seinfeld (2006) ...."

Pg 16786, ln 5: remove comma after salt and DEHM

Pg 16787, ln 19-20: Mention the effects on the strong seasonal cycle in NH biogenic emissions on the annual surface averages (i.e.) are the maximal mixing ratios similar during boreal summertime?; ln 25: replace summation with 'cumulative total". Define the latitude and longitude values of the domain.

Pg 16788, ln 3: A range of global monoterpene emissions is provided in Arneth et al., 2008. How does the annual total in DEHM lie within this range? ln 9: replace "validate" with compared; ln 10-11: provide latitude and longitude for each site as done for Thompson Farm; ln 13: "in time periods". This should be replaced by "days of the year" as time periods infers a specific year; ln 35: "... with corresponding the measurements "

Pg 16789, ln 1-2: Although the relative magnitudes are similar a correlation co-efficient of 0.34 shows that the timing of the emission is quite different. Please add a sentence regarding this in the text.; ln 15: the simulation is for 2006 therefore the comparison is not for corresponding years as currently described in the text; Introduce Figure 3, and the related discussion, immediately after Tables 1 and 2. Move more general reasons for the mismatch to the end of Section 3.1.; ln 23 replace "East Asia" with South-East Asia, replace "on both" with during; ln 25: Higher SOA in the NH due to higher temperatures, the tropics only has a small seasonal cycle.

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Pg 16790, ln 1: What is the difference in temperature in the tropics between January and July? There is little seasonal cycle in monoterpene emissions in the tropics. ln 3: there is a stark contrast between Africa and South America with respect to SOA concentrations. Can the authors explain why considering the distribution of vegetation and monoterpenes (probably related to POA distribution)? ln 5-6: remove anthropogenic from the discussion if negligible. This could be confusing and misinterpreted as a link between anthropogenic emissions and SOA. ln 10: Has POA been defined? ln 18: "tropic" should be "equatorial regions"; ln 10-24: the discussion here is not clear and should be re-written. ln 29: repalce "the model" with DEHM

Pg 16791, ln 3-4: Given that the total global SOA in DEHM is near the lower limit of 2 Tg/yr surely it is not so surprising that the model underestimates the measurements. What is the corresponding global total of POA's and how does this compare with the literature values? Ln 5-6: Is this probably due to good meteorology driving MEGAN? ln 3-14: The findings related to OA could be interpreted in a better way by segregating the US into e.g. East and West and showing the corresponding correlations. This would add to the discussion as POA will be higher in the East than the West. This could be done by introducing an additional Table which focuses on the correlation co-efficient and biases in different states similar to what is done for Europe.

Pg 16793, ln 1-7: The authors should comment on the differences in agreement between seasons and potential causes of the significant underestimation during the wintertime. From figure 9 it can be assumed that the sea-salt distribution is not the cause of the under-estimation but rather the other PM components in the system. It has been shown before that SOA contributes between 1-8% of OA therefore the missing component seems related to one of the other aerosol types. For instance how good are the BC emissions? Further discussion should be added on such details. Sect 4: Has there been some past validation of the surface O3 distribution in DEHM over Europe in previous work? Please add a few sentences related to this. Ln 11: "the latter simulation" . . . is this the BASE simulation?

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Pg 16794, ln 10: Unless you are able to differentiate between wildfires and biomass burning then the burning component is not strictly natural but mostly related to agricultural practises; ln 22-25: there have been many studies related to the influence of lightning on surface O3 such as Zhang et al. (2003), Aghedo et al (2007), Holmes et al. (2013) so this sentence is not correct and should be modified.

Pg 16798: The observational data in Fig 10 could be segregated into different regional components e.g. Scandinavia and the different sensitivity tests related to e.g. wildfires used for a better quantification of the contribution. The inter-annual variability in wildfire incidence will be important for capturing events though.

Pg 16799, ln 1-2: Please include the latitude and longitude limits of each region; ln 23-next page: This section can be significantly shortened to a few sentences as the effects of non-linear O3 chemistry are well known in global modelling.

Pg 16801, ln 22-24: This sentence should be moved out of the conclusions to the discussion. Can the previous comparisons be valid for these simulations which adopt different emission inventories for NOx?

Pg 16802, Final paragraph: Again there is new discussion arising related to the under-prediction of PM2.5 in Europe which should not be in the conclusions but in the appropriate section.

Tables:

General: Replace "NO. data" with 'Number of observations'.

Table 4: replace "Lightnings" with "Lightning"

Figures

Fig 2: Use a logarithmic Y-axis to allow better visibility between the different types of monoterpenes. The units should be Tg species/yr.

Fig 4: The colour scale needs changing to 0.1 between 0.1-0.7 as there is currently

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too much green which hides the variability in e.g. Europe.

Fig 7: Using a total average across the US masks the fact that the agreement in the West seems better than in the East.

Fig 9: Legend ". . . a combined source functions . . . "

Fig 11: Add "No-NE" to the panel relating to the sensitivity simulation and 'Control" to the BASE panel relating to the simulation. Also "relative percentage differences" .

References:

Aghedo, A. M., Schultz, M. G. and Rast, S, The influence of African air pollution on regional and global tropospheric ozone, Atms. Chem. Phys., 7, 1193-1212, 2007.

Arneth, A., Monson, R. K., Schurgers, G., Niinemets, ÇŢ., and Palmer. P.: Why are estimates of global terrestrial isoprene emissions so similar (and why is this not so for monoterpenes)?, Atms. Chem. Phys., 8, 4605-4620, 2008.

Dentener, F., et al, Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescribed datasets for AeroCom, Atmospheric Chemistry and Physics, 6, 4321-4344, 2006

Holmes, C. D., Prather, M. J., Sovde, O. A. and Myhre, G, Future methane, hydroxyl and their uncertainties: key climate and emission parameters for future predictions, Atmos. Chem. Phys., 13, 285-302, doi:10.5194/acp-13-285-2013, 2013.

Ott, L. E., K. E. Pickering, G. L. Stenchikov, D. J. Allen, A. J. DeCaria, B. Ridley, R. F. Lin, S. Lang, and W. K. Tao, Production of lightning NO and its vertical distribution calculated from three dimensional cloud scale chemical transport model simulations, J. Geophys. Res., 115, doi:10.1029/2009JD011880, 2010.

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