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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Multi-model evaluation of short-lived pollutant distributions over East Asia during summer 2008" *by* B. Quennehen et al.

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Please find below detailed replies to your comments. Since we have made significant changes to the manuscript, we invite you to read our replies together with the new version of the manuscript attached as a pdf.

Multi model evaluation of short-lived pollutants over East Asia during summer 2008 (reviewer 1) This paper reports on an exercise where six global and 1 regional model are performing the same model experiment (i.e. using the same ECLIPSE emissions, summer 2008) and compare model results with a collection of measurements. While the paper pulls together an interesting set observations, the paper is not well framed in terms of defining a key-question or hypothesis. For instance can the models be used

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to constrain emissions? Can the set of models be used to provide better estimates of air pollution, RF, or any other parameters, compared to earlier studies. While the region of interest is highly relevant, it is not clear why the problem is tackled with six global models, and only 1 regional model. It is also not clear what the new findings are, compared to what is already known. Authors' reply: We thank the reviewer for giving us the opportunity to explain more clearly the rationale our study. We appreciate their remarks and suggestions. The Introduction of the new version of the manuscript has been rewritten in order to clarify the purpose of this study and to better explain the objectives and rationale. It includes the following text:

"Evaluating the CLimate and air quality ImPacts of Short-livEd pollutants∼(ECLIPSE) project developed new emission inventories for present-day global SLP emissions as well as future scenarios designed to benefit both air quality and climate with a focus on Asia and Europe (see Stohl et al. (2015) for discussion of the ECLIPSE rationale and summary of results)." Âń An important component of ECLIPSE is the so-called reality check to evaluate model performance over pollutant source (Europe : Stohl et al. (2015), China/Asia∼, the focus of this paper) and receptor (Arctic : Eckhardt et al. (2015)) regions. In these evaluations, the ECLIPSE models were run with the same present-day ECLIPSE emission inventory∼(ECLIPSEv4a) for 2008 and 2009. Note that the same global models were used to estimate sector/regional emission responses and, in a sub-set of cases, to predict, using the ECLIPSE emission scenarios, future atmospheric composition and associated impacts on climate and air quality. The ECLIPSE global chemistry-climate models may not be the most suitable tools to assess air quality impacts, however they are the tools used to evaluate climate and air quality impacts together. To address this point, a regional model is also included in the evaluation, and one of the global chemical-transport models is run at relatively high horizontal resolution (50 km) compared to the other global models. Âż

We note that the reviewer has mainly focused their remarks on the analysis of the trace gas results. The new findings about trace gas results including a discussion about the

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causes for model discrepancies are now detailed in Sect. 3.6. We have also expanded the revision of the paper to include further analysis of the aerosol results (Sect. 4.3).

I could imagine that the paper would be better placed in the ACP sister journal, GMD (characterizing model performance), an option that the editor and authors should seriously consider. In summary I think the paper in its current form has to little scientific novelty to justify publication in ACP. The authors could consider however choosing a more in depth discussion of certain aspects of the evaluation. In my comments below I mention some potential deepening of the analysis. I suggest that such work doesn't necessarily have to be done with all models, but additional sensitivity studies with 1 or 2 models could make the paper more relevant and attractive. Authors' reply: We thank the reviewer for their suggestions. Following consultation with the Editor, we decided to follow the recommendation of both anonymous referees to analyse out results in more depth and continue with ACP. We have therefore substantially rewritten the paper providing additional scientific analysis that was, as the reviewer points out, missing from the submitted version of the manuscript. In addition, this paper does not focus on describing model developments, it aims to quantify the ability of models to accurately simulate the spatial and vertical distributions of trace gases and aerosols over Asia in order assess uncertainties related to model estimates of air quality and climate impacts of SLPs. The discussion and analysis of the results in the paper have been substantially updated. New findings are now detailed in two discussion sections on the trace gases (Sect. 3.6) and aerosols (Sect. 4.3). This includes further analysis of causes in model-observation discrepancies based on, for example, trace gas ratios and modelled diurnal cycles, in the case of ozone. We also included a sensitivity study, using one model, to quantify the impact of additional emission mitigation (vehicles, power/chemical plants) in Beijing province in the context of the Beijing Olympic and Paralympic Games (August/September 2008).

Detailed comments: P 11051 l 5 troposphere includes surface=>better tropospheric columns Authors' reply: "troposphere" has been replaced by "tropospheric columns"

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where necessary in the text.

l 6 I don't think a mismatch of NO2 between global models and a few stations can be conclusive on emissions, given coarse resolution of the models. The regional model should be more representative but even then it will depend much on the vicinity of local sources. Authors' reply: We agree with reviewer #1 that this conclusion was too general and not backed up by the results. The abstract and text in the main paper (e.g. Sections 3.3, 3.6) have been revised to better reflect the finding of this study with regard to NO2 distributions and NOx emissions.

l. 9 I think to some extent this provides the argument that coarse resolution models can not be used for such detailed comparison in polluted areas. This is not new knowledge. Authors' reply: We agree that it is difficult to compare global models with surface data collected in urban locations, for example. On the other hand, it is important to assess these models over a variety of scales and locations ranging from urban to rural and on continental scales. Such models need to be able to capture average patterns in terms of gradients from polluted to background environments as well as vertical distributions of pollutants. As explained above, these models have been used in the ECLIPSE project to assess air quality and climate impacts together. With regard to the representativeness of the model/observations comparison we now explain, in several places in the text, which observations we have used, for which purpose and which spatial scales they represent.

For example, in the Introduction: "In order to assess model performance over East Asia for air quality, as well as climate, we use a variety of different datasets covering the urban, regional, and continental scales. Ozone, aerosol and precursor data at surface sites in urban and rural locations are used, together with CAREBEIJING aircraft data collected in the lower troposphere south of Beijing, to evaluate model performance in terms of local and regional pollution from major emission regions. Continental scale horizontal and vertical transport of ozone and aerosols, important for radiative impacts, are assessed downwind of the main emission regions using aerosol lidar data as well

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as satellite aerosol lidar and tropospheric ozone, CO and NO2 column data."

l. 10 What are the important conclusions? Are the global model RFs all wrong? What did we learn? Authors' reply: The Abstract and Summary section have been rewritten to make the our conclusions much clearer to the reader. We have also, as noted above, included 2 new sections discussing possible causes for the trace gas and aerosol discrepancies compared to the observations and the implications of these findings for assessment of air quality and climate impacts. Several points are highlighted that contribute to model uncertainty with regard to radiative forcing estimates. Here are the main conclusions :

" Models show systematic positive biases in ozone, especially at rural surface locations, and compared to satellite data downwind of major Chinese emission regions. The general underestimation of CO over and downwind of emissions is linked to this, most likely due excessive destruction by OH, suggesting that CO lifetimes are too short. Reasons for ozone differences varies between models but appears to be linked to model ability to simulate VOC and NOx regimes in polluted and less polluted environments. This may also be linked to inter-model spatial variability in compared to NO2 surface data and NO2 satellite column data. The latter, however, indicates a possible underestimation in Nox emissions over Korea and Japan as well as under (over)-estimation of emissions to the south/east (west) of the Chinese NCP emission region. These findings point to the need to employ adequate model resolution to improve simulated responses to emissions when moving from ozone titration to ozone production regimes within large polluted conurbations, their surroundings and downwind. Overestimation of Asian ozone and its transport downwind implies that radiative forcing from this source may be overestimated. Sensitivity analyses, based on one model, suggest that emission mitigation over Beijing cannot explain these discrepancies.

Satellite-derived AOD measurements were reproduced quite well by the models over China even if evaluation of individual aerosol components over Asia overestimate ECLIPSE model-mean surface BC and sulphate aerosols in urban China in summer

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2008. The effect of the short-term mitigation measures taken during the Olympic Games in summer 2008 is too weak to explain divergences between the models and observations. Our results rather point to an overestimation of emissions close to the surface in urban areas, particularly for SO2. A potential reason for this is the fact that the spatial distribution of power plant emissions has been changing dramatically in the last decades in China Liu et al. (2015), a change that has not been captured well in the ECLIPSEv4a dataset. A strong underestimation by ECLIPSE models of aerosol loadings has been identified over northern India, suggesting that the emissions of BC and precursors of other aerosols are underestimated in the ECLIPSEv4a inventory. Improvements have already been included, such as higher emissions from kerosene lamps, in the ECLIPSEv5 dataset. Model deficiencies in the representation of pollution accumulation due to the Indian monsoon may also play a role. The underestimation of the scattering aerosols in the lower troposphere, above the boundary layer, suggests too much vertical transport of pollutants towards the free troposphere and/or insufficient deposition in the boundary layer, leading to overestimated aerosol residence times in models."

p. 11055 l. 15 Although of course attractive to use, CAREBEING was taking place in a period of lower than usual emissions, while this was not considered in the models. This is of course strange, but perhaps only minor fraction of analysis is affected? Not clear. Why not use one model with 'lower' emissions in the period and region of interest? Authors' reply: A sensitivity run has been performed using the WRF-Chem model to quantify the local and regional effects of emission mitigation in Beijing Province (June-September 2008) based on the emission mitigation plan described in Wang et al. (2010). Pollutant emissions from transport, industrial and solvent use sectors were reduced in the Beijing region from 1-15 August 2008. Prior studies are discussed briefly in the Introduction and details and findings are discussed mainly in Section 3.6 and also in Section 4.3: The following text is included in Section 3.6: "To assess the possible impact of emission mitigation measures in Beijing during the period analyzed in this study, the WRF-Chem model was run for 2 weeks (1 to 15 August 2008) with

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reduced pollutant emissions from the transport, industrial and solvent use sectors were reduced following the mitigation strategy during the Olympics described in Wang et al. (2010). For example, emissions of all species in the transport sector were reduced by 75% in Beijing and 20% in the area 200 km from Beijing, corresponding to eight model grid cells around Beijing in this model. Emissions linked to the industrial sector or to solvents were reduced by 50% in the same region. Most pollutant concentrations are reduced resulting, for example, in lower CO, by about 30 ppbv, locally in and around Beijing in the emission reduction run compared to the base run. This results in ozone reductions of up to 6-7 ppbv in the region of Beijing. Based on these results, it appears that these reduction measures cannot explain the discrepancies between the models and the observations discussed earlier."

Fig. 1 shows the difference between surface concentrations in the base simulation and surface concentrations in the simulation where the emissions have been reduced during the Olympics for two pollutants: CO and ozone. In general, most pollutants are reduced (CO, ozone, SO2, BC, OC, SO4) in Beijing and the impact of emissions abatement is too low to explain discrepancies in the models compared to observations.

Fig. 1: Differences between surface concentrations in CO (left) and ozone (right) in the base simulation and surface concentrations in the simulation where emissions have been reduced during the Olympics using the WRF-Chem model.

p. 11056- table 1: Is it correct that NorESM was not running 2008? If so, why is it then included? Authors' reply: NorESM is a chemistry-climate model. As such, it was not nudged to meteorology, but forced using reanalyses of sea surface temperature for 2008 (now noted in Table 1). Compared to the other models, driven by ECMWF or NCEP analyses, NorESM has a monsoon circulation that penetrates to far to the north over China. NorESM relative humidities are also higher over China, for example, compared to ECMWF and NCEP analyses which is likely to affect ozone photochemistry as well as aerosol formation. These factors, linked to model representation of the Asian monsoon, are discussed in the results sections. NorESM was included in this

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evaluation to provide consistency with companion papers (e.g Stohl et al., 2015) and because it was used to derive present-day and future emission impacts on air quality and climate.

p. 11056 Can you provide some statement on the effect of not including seasonality in most emissions? (Especially since the focus on the short period). Here a sensitivity experiment would have been appropriate. Authors' reply: As mentioned in the text (P. 11056, L. 21-22), all ECLIPSE models used a seasonal cycle for the domestic heating sector, based on Streets et al. (2003). WRF-Chem uses seasonal cycles for all emission sectors, and only two other sectors (energy, industry) exhibit small variations ($\sim$28 %) relative to the mean emissions. Stein et al. (2014) highlighted the importance of including a seasonal cycle for the CO emissions to correct a low bias in the northern hemisphere found in many previous global studies, especially in winter, but it is unlikely that this is having a significant effect over Asia in summer where CO is already underestimated.

p. 11057 l. 11 There is some strange reasoning: on the on hand dust emissions in WRF-CHEM are too high, and two lines later it is argued that it doesn't matter anyway, so why leave it out? Authors' reply: The reviewer is correct: the text in the manuscript was unclear. According to the literature, dust sources should not impact East Asia during summer. In the current version of WRF-Chem dust emissions are too high at this time year leading to unrealistic amounts of dust at high altitudes, and thus, excessive AODs and backscattering values (Saide et al., 2012). The following text has been added to the manuscript : Âń WRF-Chem provides online dust and sea-salt emissions but only the latter are used in the ECLIPSE simulations due to an overestimation of dust loads, as reported by Saide et al. (2012). The main dust sources in East Asia are located in dry regions of China and Mongolia, north of the Himalayas (Taklamakan, Gobi and Gurbantunggut deserts). Most of the dust events occur in spring (Huang et al., 2013) whilst in summer, due to the Asian summer monsoon ïňĆux, rather little dust is transported to coastal areas (Kim et al., 2007). Thus, neglecting this source in

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WRF-Chem summertime simulations is not expected to introduce large bias in modeled aerosol loads. Âż

p. 11059 Was NorESM nudged, or calculating own meteorology? If the latter, why was it included? Authors' reply: NorESM is a chemistry-climate model. It was not nudged to meteorology, but forced using reanalyses of sea surface temperature for 2008. See previous reply on this point.

p. 11059 what means FNL? Authors' reply: As defined p. 11059, FNL means "final" analyses. This has been clarified in the text.

p. 11061 Does it make sense to compare WRFCHEM with the IASI ozone profiles? Authors' reply: Yes, in our opinion, it makes sense to compare WRF-Chem with IASI ozone columns since the model provides results up to 20 km. The IASI sensor is most sensitive at 4 and 16 km as indicated by the averaging kernel (not shown). In addition, WRF-Chem ozone profiles are completed between 20-40 km.

p. 11061 explain the resolution of IASI and how models at different resolution where compared. One month or one year? Authors' reply: Ozone columns are compared on a monthly basis (August 2008). IASI data have been averaged on a 1x1° grid and model results are scaled to this grid. The following sentence has been added in section 3.1: "IASI data are averaged on a 1x1° grid and model results were scaled to this grid."

p. 11062 indeed global models have difficulties putting the correct location of the monsoon flux. Authors' reply: We agree with this comment. As pointed out by the reviewer this may be having an influence on the NorESM modelled aerosol results. This point is now considered in section 4.3. NorESM-derived AODs is slightly underestimated in northern India. This is an indication that the effect of pollution accumulation due to the Indian monsoon is not adequately represented. NorESM has indeed lower winds associated with the Indian monsoon (Fig. 1).

p. 11063 l. 20 Which time period? interesting to see that the models are having

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so different bias/error with regard to NO2. However I would like to be sure that they really used the same emissions. Then some dedicated experiments could point to explain why the difference are so large (lifetime/mixing/deposition)? Authors' reply: All the models use the same emissions, which have been specifically designed for the ECLIPSE project. Further analysis of model discrepancies in NO2, CO and ozone is now included in the new Section 3.6. For this purpose we compared available model results to observed trace gas ratios at different locations. In the case of polluted locations differences compared to emitted and observed trace gas ratios (e.g. CO/NOx) are used to indicate a lack of chemical processing or mixing of air masses. We also use, for certain models, ratios of NO:NO2 and ozone:NOz (NOy-NOx) to examine whether models are in VOC or NOx limited regimes with respect to photochemical ozone production.

p. 11064 If air quality is an issue, the authors should worry also about the vertical resolution of the first layer, the height of the emissions, the height of the measurements, and how the model was sampled. Authors' reply: All the models prescribe surface emissions. The first vertical layer height varies between 25 and 192 m depending on the model and we did not find any significant correlation between the first layer height and modelled pollutant concentrations. Model results were sampled from the first layer in grid cells corresponding to station locations. This point is now addressed in Section 3.4: "There is significant variability in modelled NO2 compared the observations at polluted and rural sites. This could be caused by differences in model vertical resolution near the surface although no correlation was found between the height of the first model layers and pollutant concentrations."

p. 11065 Given the lifetime of CO, it would be instructive to learn what OH concentrations the models are having now, and what would be needed to reproduce the right gradients? Can the coarse resolution models be used for this? Author's reply: We now include a more detailed discussion about the differences between the model results and CO observations in Section 3.6. We were able to examine OH concentrations

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in certain models. It appears that the underestimation in CO is linked to the overestimation in ozone, particularly downwind of China. As explained previously, the use of complementary datasets covering different environments and spatial scales allows us to draw more firm conclusions. The following text is included in Section 3.6: "The ECLIPSE models also systematically underestimate CO downwind compared to surface data over Korea, Japan, and compared to IASI CO data over Japan and downwind over the north-western Pacific Ocean. Whilst inclusion of additional seasonality in the ECLIPSE emissions (already included for domestic combustion), might improve agreement in winter and spring (Stein et al., 2014), this is unlikely to explain these summertime differences. Low model CO appears to be linked to the clear overestimation in modeled ozone at rural sites and compared to IASI 0-6$\sim$km column data. Excessive ozone resulting in too much destruction of CO by OH and may suggest that modeled CO lifetimes are too short. This hypothesis is consistent with the findings of Monks et al. (2015) who concluded that, in models run with the same emissions, differences in OH$\sim$(chemical schemes) are a more likely cause of the systematic CO underestimation in the Northern Hemisphere and the Arctic than differences in vertical transport. Indeed, we find that surface August mean modeled OH (not shown) is higher in the NorESM model (due to the penetration of the monsoon flux) compared to, for example, TM4-EPCL and WRF-Chem over the main Chinese emission regions. In contrast, excessive modeled CO over the central Pacific, where concentrations are low, may be due to the position of the Pacific anticyclone in the meteorological analyses used by the majority of models. A shift in the position of the anticyclone to the south could result in this pattern of negative (positive) biases over the north (south) Pacific potentially as a result of transport that is too zonal. This may also explain low modeled CO in the Arctic noted by Monks et al. (2015)."

p. 11066 l 25 I understand that properly taking into account aerosol scattering is a big thing for NO2 retrievals. Was this done at these locations. What could the error? Authors' reply: The observed NO2 columns were retrieved from GOME-2 satellite measurements using an algorithm described by Boersma et al. (2004). The NO2 columns

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are retrieved with a precision of 35-60 % and the main uncertainties are dominated by uncertainty in the estimate of the tropospheric air mass factors (AMF) (Boersma et al., 2004). Cloud fraction, surface albedo and profile shape are the most important factors influencing the AMF estimation uncertainties. Boersma et al. (2004) concluded that the aerosol correction is indirectly incorporated into the cloud correction and that an independent aerosol correction procedure would lead to differences of less than 10% in the correction factor. The following sentence has been added in Section 3.3 (Tropospheric NO2 columns): "Column retrievals do not include corrections for aerosol scattering, which are estimated to less than 10 % by Boersma et al. (2004)"

p. 11068 l. 25 What was assumed for size distribution for particulate emissions? Authors' reply: Particulate emissions are distributed differently depending on each model: three models only considered total mass (EMEP, NorESM and OsloCTM2), TM4-ECPL includes all the emitted mass in the fine mode, HadGEM3 distributed biofuel and fossil-fuel combustion in modes centred on 150 and 60 nm, respectively, both with a sigma of 1.59. ECHAM equally distributes ship, industrial and power-plant emissions in the accumulation and coarse modes, respectively centred on 75 and 750 nm with sigma of 1.59 and 2.0 and other emissions are equally separated between the Aitken mode, centred at 30 nm and the accumulation mode whereas biomass and fossil fuels are emitted in a single mode, centred at 15 nm with a sigma of 1.8. WRF-Chem distributes the emissions in its 8 size bins centred on 60, 117, 234, 468, 937.5, 1875, 3750, 7500 nm and with the following percentages: 25, 25, 15, 15, 7.5, 7.5, 2.5, 2% respectively.

References: Boersma, K. F., H. J. Eskes, and E. J. Brinksma (2004), Error analysis for tropospheric NO2 retrieval from space, J. Geophys. Res., 109, D04311, doi:10.1029/2003JD003962

Liu, F., Zhang, Q., Tong, D., Zheng, B., Li, M., Huo, H., and He, K. B.: High-resolution inventory of technologies, activities, and emissions of coal-ïňĄred power plants in China from 1990 to 2010, Atmospheric Chemistry and Physics, 15, 13 299–13 317, doi:10.5194/acp-15-13299-2015, http://www.atmos-

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chem-phys.net/15/13299/2015/, 2015.

Saide, P. E., Spak, S. N., Carmichael, G. R., Mena-Carrasco, M. A., Yang, Q., Howell, S., Leon, D. C., Snider, J. R., Bandy, A. R., Collett, J. L., Benedict, K. B., de Szoeke, S. P., Hawkins, L. N., Allen, G., Crawford, I., Crosier, J., and Springston, S. R.: Evaluating WRF-Chem aerosol indirect effects in Southeast PaciïňĄc marine stratocumulus during VOCALS-REx, Atmospheric Chemistry and Physics, 12, 3045–3064, doi:10.5194/acp-12-3045-2012, http://www.atmos-chem-phys.net/12/3045/2012/, 2012.

Stein, O., Schultz, M. G., Bouarar, I., Clark, H., Huijnen, V., Gaudel, A., George, M., and Clerbaux, C.: On the wintertime low bias of Northern Hemisphere carbon monoxide found in global model simulations, Atmos. Chem. Phys., 14, 9295-9316, doi:10.5194/acp-14-9295-2014, 2014.

Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S. M., Tsai, N. Y., Wang, M. Q., Woo, J.-H., and Yarber, K. F.: An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, J. Geophys. Res.-Atmos., 108, 8809, doi:10.1029/2002JD003093, 2003.

Wang, B., Shao, M., Lu, S. H., Yuan, B., Zhao, Y., Wang, M., Zhang, S. Q., and Wu, D.: Variation of ambient non-methane hydrocarbons in Beijing city in summer 2008, Atmos. Chem. Phys., 10, 5911-5923, doi:10.5194/acp-10-5911-2010, 2010.

Worden, H. M., Cheng, Y., Pfister, G., Carmichael, G. R., Zhang, Q., Streets, D. G., Deeter, M., Edwards, D. P., Gille, J. C., and Worden, J. R.: Satellite-based estimates of reduced CO and CO2 emissions due to traffic restrictions during the 2008 Beijing Olympics, Geophys. Res. Lett., 39, L14802, doi: 10.1029/2012GL052395, 2012.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/15/C12288/2016/acpd-15-C12288-2016-supplement.pdf

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