

Interactive comment on “Observations and modeling of air quality trends over 1990–2010 across the Northern Hemisphere: China, the United States and Europe” by J. Xing et al.

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We would like to thank the reviewer for a very thoughtful and detailed review of our manuscript that helped to improve the paper. We address all the points raised by the reviewer as follows. We basically followed all the comments and revised manuscript accordingly.

[Comment]: The manuscript says that an evaluation of the WRF meteorology will be the subject of a separate paper. I think that there should be at least some evaluation of WRF in the current paper. This could be simply be some summary statistics of model

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performance.

[Response]: we agree with the reviewer and have provided additional description about the WRF performance evaluation in the revised manuscript (P6 L17-P7 L1), as below: “WRF performance for the simulation of hourly surface temperature (T), relative humidity, wind speed and direction was evaluated through comparison with observations from NOAA’s National Climatic Data Center (NCDC) Integrated Surface Data (ISD with lite-format) which provides hourly (or with 3-hour interval) meteorological observations over a long historical period across the globe. The mean bias of T, wind-speed and direction over the simulation domain is -0.4 K, 0.4 m s⁻¹ and -3 degree respectively, within the benchmark range suggested by Emery et al. (2001) for retrospective regional-scale model applications which is $\leq \pm 0.5$ K, $\leq \pm 0.5$ m s⁻¹ and $\leq \pm 10$ degree respectively.”

[Comment]: The authors are comparing CMAQ model output (which is I think at 108 km resolution) with the AQS and EU-AIRBASE data, which are primarily from urban areas. Each of these sites are representative of much smaller regions. I do not think this comparison is appropriate. The AQS and EU-AIRBASE data should be averaged over the 108 km grid cells before comparing with the model to obtain a more valid analysis.

[Response]: we agree with the reviewer that the averaged AQS and AIRBASE data are more appropriate for the comparison against with simulations on a 108km resolution. We have reworked these two networks and updated all the numbers in the revised manuscript. In most of cases, the performance gets slightly improved. The NMBs for SO₂ in AQS/AIRBASE are changed from -46%/-12% to -38%/-18%; the NMBs for NO₂ in AQS/AIRBASE are changed from -54%/-57% to -48%/-54%. Such updates have been noted in the revised manuscript (P8 L18-L21), as below: “Sites in US-AQS and EU-AIRBASE are typically closer to urban areas and may be impacted by local pollution and features sub-grid to the model resolution, thus are representative of much smaller regions. To obtain a more valid analysis, the US-AQS and EU-AIRBASE data were averaged over the 108 km grid cells before comparing with the model.”

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[Comment]: p. 25457, lines 17-19 and p. 25458, line 8: the earlier text mentions nested regional domains at finer resolution, and then in the later text specified the three sub-regions used in the analysis. However, no specific finer resolution is mentioned. This leaves the reader unclear as to whether the sub-regions are or are not nested. I've assumed they are not. Please clarify the text.

[Response]: the three sub-regions are not nested. We have clarified it in the revised manuscript (P6 L8-9), as below: "We selected three sub-regions, i.e., eastern China (20–40 N, 100–125 E), eastern US (28–50 N, 100–70 W) and Europe (35–65 N, 10W–30 E), for further analysis and comparison with measurements. These three sub-regions are parts of the original northern hemispheric domain and no nested simulations were conducted."

[Comment]: p. 25459, lines 16-17: lightning NOx emissions are said to be from Price et al., 1997. This paper indicates the total global emission is 12.2 Tg/yr. This amount is well above the most well-accepted values of 2 - 8 Tg/yr (Schumann and Huntrieser et al, 2007, ACP). Please provide some indication of what the impact of this likely too large emission value is on NOx, O3, and nitrate.

[Response]: We have included a brief discussion about this bias in the revised manuscript (P25 L20-P26 L3), as below: "However, the model estimates still suffer from uncertainties in emissions (in regards to temporal variation and speciation), coarse spatial resolution and subsequent impacts on representation of non-linear atmospheric chemistry. The lightning NOx emissions used in this study (Price et al, 1997) are likely overestimated by 0.5 to 5 times compared to more recent study (Schumann and Huntrieser et al., 2007) and may contribute to some extent to the overestimation of NOx, O3 and nitrate concentrations."

[Comment]: p. 25460, lines 5 - 8: define the acronyms

[Response]: the acronyms have been defined in the revised manuscript, but in the previous section (P 4).

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[Comment]: p. 25463, line 7-8: Some statistics on model precipitation vs. observed should be provided. Then, the authors could more definitively say whether precipitation bias is the reason for the underestimation.

[Response]: We appreciate the suggestion from the reviewer, and we further investigated the WRF performance of the precipitation. The precipitation was underestimated domain-wide by from 4% (in summer) to 65% (in winter). We provided the statistics on the performance of precipitations and clarified the reason for the bias in the revised manuscript (P11 L22-P12 L5) as below: "Some studies also found similar under-prediction in their simulations and they attributed such low biases to the uncertainty in precipitation and overestimation of wet-scavenging. However, precipitation simulated in this study is underestimated domain-wide by 4% (in summer) to 65% (in winter). Wang et al (2009) found similar underestimation of precipitation from -31% to -41%, but SO42- was over-predicted because higher SO2 emissions were used. Future investigation of the low bias in predicted SO42- is still necessary."

[Comment]: p. 25464, lines 4-7: Were these previous modeling studies at much finer resolution? If so, then resolution may not be the issue.

[Response]: These previous modeling studies were conducted at finer resolutions of 36km/12km. We agree with the reviewer that the original statement is vague. We have rephrased that in the revised manuscript (P13 L7-L15), as below: "The correlation between the observed and simulated EC concentrations is high with $R > 0.5$, though the model significantly underestimates the concentrations. NMB up to -74% which is worse than previous modeling studies utilizing relatively higher spatial resolution (Zhang et al., 2009; NMB= -15.4 to 8 %; Eder and Yu, 2006; NMB= -6 %), but the magnitude of NMB is comparable with Wang et al. (2009) (NMB= 101.7%) which also utilized coarse spatial resolution. Some previous CMAQ modeling studies (Tesche et al., 2006; Appel et al., 2008) with higher spatial resolution also found the similar underestimation of EC, indicating other factors besides model resolution, such as uncertainties of PM speciation profiles used to estimate the EC emissions might

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also contribute to such low biases."

[Comment]: p. 25465, line 8: maybe 'eastern AQS' instead of 'mid-east AQS'. 'Mid-east' is not a commonly used term to describe locations in the US.

[Response]: as the reviewer suggested, we replaced the "mid-east AQS" by "eastern AQS" in the revised manuscript (P14 L13).

[Comment]: p. 25466, line 13: "....capture these trends, yielding trends more similar to those of the emissions"

[Response]: as the reviewer suggested, we modified this sentence into "the model was unable to capture these trends, yielding trends more similar to those of the emissions" in the revised manuscript (P15 L21-L22).

[Comment]: p. 25467, line 24: should "NOx- and VOC-limited regimes" be reversed?

[Response]: we thank the reviewer for pointing this out; this typo has been fixed in the revised manuscript (P17 L9), as below: "a likely switch of O₃ chemistry from VOC- to NOx-limited regime which usually goes along with the transition from urban to rural area"

[Comment]: p. 25472, lines 5 - 7: The authors should note that in China the rate of O₃ increase was much smaller during 1995-2002, which was the period when VOC emission growth was much greater than that of NOx emissions. This result indicates greater sensitivity of ozone to NOx emissions than VOC emissions.

[Response]: we thank the reviewer for this good suggestion. We have included this finding in the revised manuscript (P22 L9-L12), as below: "The ratio suggested is less than 1 indicating greater sensitivity of ozone to NOx emissions than VOC emissions. It's also obvious to see that the rate of O₃ increase was much smaller during 1995-2002 which was the period when VOC emission growth was much greater than that of NOx emissions in China."

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Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/14/C11212/2015/acpd-14-C11212-2015-supplement.pdf>

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 14, 25453, 2014.

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