

Interactive comment on “High-resolution simulation of link-level vehicle emissions and concentrations for air pollutants in a traffic-populated East Asian city” by Shaojun Zhang et al.

Anonymous Referee #2

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This study develops a high-resolution motor vehicle emissions inventory for a city in China, models the inventory, and evaluates the modeling results against ambient monitoring data. Main findings of this paper are that it is important to capture the spatial heterogeneity of the vehicle fleet mix across the urban domain, and to use local information on emission factors. Overall, the authors present a novel approach to mapping vehicle emissions, especially in cities where traffic activity and emission factor data are not as readily available. This is a major accomplishment and worth replicating in other cities. In regards, to the second major aspect of this study, the air quality modeling, I have some concerns that I believe need to be addressed more fully in revision.

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My comments mostly refer to the treatment of atmospheric chemistry in the dispersion model. With major revision, I do believe it is possible for this manuscript to be considered for publication in Atmospheric Chemistry & Physics.

General Comments

(1) My concerns with respect to the air quality modeling are with the treatment of chemistry, and how background levels are estimated. Unless the following concerns can be addressed, I believe that statements that quantify the fractional contribution of motor vehicle emissions to ambient concentrations observed should be removed (bottom of page 15 and top of page 16), and commentary restricted to qualitative statements.

(i) More detail is needed on how a dispersion model like AERMOD accounts for chemistry, especially for NO₂. Given that authors present high-resolution air quality maps, it seems important to capture spatial gradients that may arise due to interactions between ozone and fresh NO emissions; i.e., ozone tends to be suppressed near highways, and NO₂ elevated (Murphy et al., 2007). On page 10, Lines 15-17, the authors mention using ozone data to account for the oxidation of NO to NO₂, but do not describe how. How many monitoring sites are used in this calculation? Where are they located, and what is their proximity to roadways? What is the timescale of the NO → NO₂ conversion employed in AERMOD, and how was this estimated from observations?

Murphy, J. G., Day, D. A., Cleary, P. A., Wooldridge, P. J., Millet, D. B., Goldstein, A. H., and Cohen, R. C.: The weekend effect within and downwind of Sacramento – Part 1: Observations of ozone, nitrogen oxides, and VOC reactivity, *Atmos. Chem. Phys.*, 7, 5327-5339, doi:10.5194/acp-7-5327-2007, 2007.

(ii) It is also not clear how AERMOD treats the loss of NO₂ to PAN and HNO₃. In an urban mass, these products of NO₂ can comprise up to half of daytime NO_y (= NO_x + PAN + HNO₃, see Pollack et al., 2012). If the authors' only account for the production of NO₂ from fresh NO emissions, without accounting for the loss of NO₂ from daytime chemistry, then the NO₂ concentrations simulated from local vehicle emissions shown

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in Figure 7 could be overestimated. Consequently, the authors may overestimate the motor vehicle contribution to ambient NO₂. The importance of the loss term will depend on the photochemical age of the air mass that reaches the monitoring site, which could vary by time of day, wind speed/direction, and synoptic events. A robust estimation of the local vehicle contribution to ambient NO₂ (as shown in Figure 8) should take into account both the production and loss of NO₂.

Pollack, I. B., et al. (2012), Airborne and ground-based observations of a weekend effect in ozone, precursors, and oxidation products in the California South Coast Air Basin, *J. Geophys. Res.*, 117, D00V05, doi:10.1029/2011JD016772.

(iii) As I understand, the NO₂ observations shown in Figure 8, are daily concentrations from a single monitoring site (Page 16, Lines 10-13). Is this the average of 24-hours of data? The use of daily averages could be influenced by nighttime chemistry, which presumably would not be taken into account with AERMOD. To avoid these complications, it is better to restrict the model comparison to daytime values only.

(iv) I found the description of the CMAQ model (on Page 10, Lines 20-26) used to estimate regional background and cross-boundary transport lacking. This is important since background levels (Page 10: 304 ug/m³, 27 ug/m³, and 23 ug/m³ of CO, NO₂, and PM2.5, respectively) are as big or much larger than the motor vehicle contribution to these pollutants (Page 15: 88 ug/m³, 22 ug/m³, and 1.3 ug/m³ of CO, NO₂, and PM2.5, respectively) in Macao. For example, what was the domain of the CMAQ model used? Did it include a much wider region that encompassed other cities/provinces of China? What meteorological data and chemical schemes were used to run the model? What were the chemical and meteorological boundary conditions used to drive the CMAQ model? On Page 10, Lines 20-24, the authors mention turning off local stationary and mobile source emissions, but it is not clear what emissions inventory was used to drive the background concentrations of CO, NO₂, and PM2.5 elsewhere. What about shipping emissions, which are sources of NO_x? The emissions and meteorological data used to drive the 4 km x 4km CMAQ model need to be described in detail; the

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CMAQ model is as critical as the AERMOD model in calculating the local vs. regional contribution. Since the prevailing wind direction is from the northeast (shown in Figure S5), it appears there would be a strong influence from emissions occurring in Hong Kong and other major cities in Southeast China.

(v) If the authors' used CMAQ to calculate background concentrations, why isn't CMAQ also used to quantify the local contribution of vehicle emissions to ambient concentrations of CO, NO₂, and PM2.5, along with AERMOD? Some of these concerns I have raised with regards to chemistry could be mitigated with a chemical transport model like CMAQ. If there is similarity in the result between AERMOD and CMAQ in the local vs. regional contribution, then chemistry may not play such an important role and the modeling results presented may be valid.

Specific Comments

(2) Page 6, Lines 6-15: What were the criteria used that defined a "typical" road link? Especially, how were the 5 road links investigated for the entire day chosen? For example, in Figure S3, it appears that many of the observations were on arterial and residential roads, and relatively few observations on freeways. However, I would think it would be more important to characterize the freeways since they have much higher traffic volumes, and account for a significant fraction of vehicle traffic. Also, it would help to create a map similar to Figure S6, showing traffic volumes for each link simulated using the TransCAD model, and to also highlight which links were surveyed.

(3) Page 6, Lines 27-28: It would help to show a line with trucks in Figure S2 to illustrate this point.

(4) Page 7, Line 2: To extrapolate to other hours using Equation 3, how consistent is the temporal variability observed across road links? If they are consistent, then it is appropriate to spatially model traffic flows for the 6 PM hour only, and to extrapolate traffic patterns to other times of the day. However, if they are not, I would imagine it is better to run the traffic model for each hour of the day. To support the assumption

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that temporal variability is consistent across space, Figures 2 and 3 would benefit from estimating uncertainty bands for each road class shown.

(5) Section 2.4: It is not clear from the description here of the EMBEV-Macao model whether gross-emitters are taken into account with the emission factor data collected between PEMS and remote sensing. Average emission factors could be significantly underestimated if gross-emitters are not included (Bishop et al., 2012). Also, how are cold start emissions taking into account? It is ok to reference prior papers, but I think it is important to address these issues explicitly here.

Bishop, G. A., et al. (2012), Multispecies remote sensing measurements of vehicle emissions on Sherman Way in Van Nuys, California, J. Air & Waste Management Association, 62.

(6) Page 9, Lines 17-18: It is important to describe here the advantages and disadvantages of Gaussian models in relation to the other types of models. As highlighted in Comment 1, I have concerns over whether Gaussian models can accurately model constituents that undergo complex chemistry, including NOx-VOCs-O3 and secondary aerosols, which are pertinent to this study.

(7) Page 14, Lines 20-24: The authors mention that traffic loop detector data is collected in many Chinese cities. Is traffic loop detector data not being collected in Macao? If so, it should be mentioned here.

(8) Section 3.3: The locations of the ambient monitoring locations should be shown on a map somewhere (e.g., Figure S1).

(9) Figure 8: Why are results not shown for CO? It seems relevant to the model evaluation described (Page 15, Lines 29-31).

(10) Page 16, Lines 16-20: Another source of uncertainty are effects due to chemistry (see Comment 1).

(11) Table 4. For the most part, the fleet-averaged emission factors seem reasonable,

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except for MDPV-Gasoline and LDT-Gasoline. Why are emission factors for CO and THC nearly as large as motorcycles, presumably with two-stroke engines, which are expected to have the highest emission factors for these pollutants?

(12) Tables 5 and 6. Too many significant figures are shown, especially for CO2. Probably no more than 3 significant figures are justified given uncertainties in emission intensities.

(13) Figure S5. Where are weather stations located? Should be shown on Figure S1.

Minor Comments (14) Page 5, Line 1: I believe there is a mistake here, that "vehicle classification f" should read "vehicle classification v".

(15) Page 7, Line 23: Better to report amount of data in hours collected rather than seconds; as a reader it is hard to comprehend how much data was collected using the latter units.

(16) Page 15, Line 22: I believe there is a mistake here, "Table 6" should read "Table 7".

(17) City boundaries shown in Figure 7 and Figure S7, are hard to see. Suggest darkening the boundaries.

(18) Figure S2. The vertical axis labeling is confusing. Instead of ratios, I think fraction of total traffic counts better describes what is being shown.