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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "High-resolution quantification of atmospheric $CO_2$ mixing ratios in the Greater Toronto Area, Canada" *by* Stephanie C. Pugliese et al.

**Anonymous Referee #1**

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The authors develop a high-resolution CO2 emissions inventory (SOCE) for the Toronto region, and evaluate it against in ambient CO2 measurement. The SOCE inventory appears to perform well against ambient observations, along with FFDAS, a commonly used CO2 inventory. The main advance of this study is sectoral information provided that FFDAS does not, which is significant. The sectoral information is useful to cities in their efforts to mitigate greenhouse gas emissions. Also, not many cities have urban CO2 monitoring networks from which to evaluate emission inventories. This study provides a useful framework by which to perform evaluations in the future.

Overall, I found the manuscript to be well written, figures to be clear, and findings well



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supported. I do have some questions about the methods used to construct the bottom-up inventory, especially with the use of carbon monoxide (CO) to scale up to CO2 emissions. Most of the rest of my comments are minor in nature and relate to clarity. My recommendation is that this manuscript be published with a moderate amount of revisions.

General Comments

1. As I understand, there is provincial-level estimates of CO and CO2 that are reported by the Canadian National Inventory Report (NIR), with broad sectoral information (e.g., area, point, on-road, off-road). Then there is also a gridded inventory of CO emissions that is processed at 2.5 km x 2.5 km. The authors' reconcile the provincial-level CO2/CO estimates with the gridded CO inventory to arrive at a gridded CO2 inventory.

My main critique with this approach is with the CO2/CO emission ratios reported. Some of these values seem unbelievable, which reflect potential problems in either CO2 or CO emissions reported by the NIR (more likely from CO). For example, the on-road CO2/CO emissions ratio reported here (Line 247) is 29.5 g CO2/g CO. This is equivalent to a CO emission factor of 33.9 g CO/kg CO2 or 110 g CO/kg fuel (using a carbon fraction of 0.85 g C/g fuel for gasoline). Roadway studies report tailpipe CO emission factors from gasoline cars at 10-20 g/kg fuel [McDonald et al., 2013]. The factors reported here appear too high. Also, based on the point source emission factor of 313.1 g CO2/g CO (Line 239), $\sim$0.5% of the carbon emitted is as CO and the rest from CO2. This is a very small number in the denominator from which to scale to CO2 emissions, introducing potentially large uncertainties in industrial CO2 emissions.

Ultimately, I found the reporting of CO2/CO emission ratios distracting and not central to the inventory constructed. What I believe the authors' are really doing here is using the gridded CO inventory as spatial/temporal proxies for CO2 emissions, and downscaling CO2 emissions from the provincial-level to grid cells by sector. Rather than report CO2/CO emission ratios that are dubious, I suggest reframing inventory





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methods to emphasize the use of CO as spatial/temporal proxies for CO2.

2. In Section 2.4, too many significant figures are reported in emission inventory estimates, which suggest a high degree of certainty in emissions that is unwarranted (especially for CO). Suggest using 2-3 significant figures at most.

Specific Comments

3. Line 207. How are CO2 emissions from the Canadian National Inventory Report (NIR) estimated? Are these based on energy or fuel use statistics, or from engineering calculations? Since CO2 emissions is the focus of this paper, it would be helpful to include a few sentences on the basis for how CO2 emissions are estimated in national reporting.

4. Equation 1. The "total" subscript was confusing to me. I believe what the authors' mean is "sector" in the third term of the equation (e.g., total area, point source, on-road, etc.) and "sub-sector" in the first two terms.

5. Section 2.4.1. Typically when I think of area emissions they are dispersive sources that include residential, commercial, AND industrial sources. Suggest that this source category be renamed to something like "Area industrial emissions".

6. Lines 232-234. The combustion efficiency is not actually that variable, it is just that CO emissions are almost negligible from point sources and hence why the CO2:CO ratio is variable due to a tiny denominator (see Comment 1). Suggest re-wording of this sentence.

7. Lines 255-256. Lawn equipment and other small two- and four-stroke gasoline engines (e.g., snow equipment) have been shown to be a significant source of CO emissions [Gordon et al, 2013; Volckens et al., 2007; Bishop et al., 2001]. Where would they show up in the APEI, or are they included here? More importantly, how are off-road gasoline engines specifically accounted for in this study, which contribute high amounts of CO, but consume small amounts of fuel? Other off-road diesel equipment would





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consume significantly larger amounts of fuel than off-road gasoline engines, but have much lower CO emission factors. Not properly accounting for off-road emissions of CO between gasoline and diesel engines could affect the scaling of off-road emissions of CO to CO2.

8. Lines 363-365. Some more description of the FLEXPART model is needed here, or could warrant a short paragraph in the methods section. Specifically, how many hours was the back trajectory simulated for? Was this run for each site? I'm guessing the emission inventories are then multiplied by the footprint to arrive at concentrations, and then compared with ambient CO2 measurements. Also, a reference to FLEXPART and some description of the model would be helpful to a reader unfamiliar with the model.

9. Line 365. This appears to be the first mention of the TAO site. It would be helpful to describe this location in more detail in Section 2.2. Also, it is not clear why looking at gradients between Downsview and TAO is a useful metric. Is it because TAO is a downwind site, whereas Downsview is downtown? In general, for a reader unfamiliar with Toronto, it would be helpful to describe locations as urban or rural more explicitly throughout Results and Discussion.

10. Lines 492-494. Focusing on wintertime months, while easier from a modeling perspective, would bias our understanding of CO2 emissions towards wintertime sources. The sources and spatial patterns of emissions vary between winter and summer. For example, peaking plants could be important in summertime [Farkas et al., 2016]. Seems like we should try to understand both periods.

11. Figure 5. This plot could benefit from some uncertainty bands on the CO2 measurements, such as the standard deviation or 95% confidence interval of the mean. In this way, it will be easier to discern the variability in CO2 concentrations, as well as the significance of the model improvements.

12. Figure 7. I cannot see the line for the biogenic sources, though it is called out in the text (Line 490).

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