

## ***Interactive comment on “High-resolution quantification of atmospheric CO<sub>2</sub> mixing ratios in the Greater Toronto Area, Canada” by Stephanie C. Pugliese et al.***

**Anonymous Referee #2**

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In this study, CO<sub>2</sub> emissions for most sectors within Ontario were estimated using CO as a starting point, along with sector-specific CO<sub>2</sub> to CO ratios from a province-wide emission inventory. Various CO<sub>2</sub> emission inventories, including existing estimates as well as the newly-developed estimates in this study, were then used as input to a weather and chemistry/transport model to predicted CO<sub>2</sub> concentrations. Modeled concentrations were compared against observations at four monitoring sites in Southern Ontario. Sector-specific tracking of CO<sub>2</sub> emissions led to the conclusions that, during winter months, the daytime increase in CO<sub>2</sub> above background was dominated by vehicle emissions, whereas at night, the increase was dominated by wintertime natural gas combustion for space heating in residential and commercial buildings.

C1

The CO<sub>2</sub> emission factor from natural gas combustion. I calculate by carbon balance that the value for pure methane should be  $42 \text{ mol/m}^3 \times 16 \text{ g/mol} \times 44 \text{ g CO}_2 / 12 \text{ g C} = 2464 \text{ g CO}_2 / \text{m}^3$  of natural gas burned (assuming gas temperature of 15 deg C = 60 deg F). Real natural gas may include inert gases such as nitrogen and carbon dioxide. There may also be some incomplete combustion in the residential sector, though I expect those adjustments to be minor for the space heating sector in question. The cited emission factor (1879 g CO<sub>2</sub>/m<sup>3</sup>) may be too low, the authors should explain what assumptions underpin this emission factor, which dominates CO<sub>2</sub> enhancements at night in their modeling.

The authors should say more about seasonality and diurnal patterns of emissions. Presumably many residential users turn down the heat at night, and some furnaces (esp. residential) run only during winter months. Have such effects been accounted for in formulating the CO<sub>2</sub> emission inventory for southern Ontario?

It would be helpful to say more about how motor vehicle CO emissions were estimated, in particular the spatial and diurnal distribution of traffic, and also the gasoline/diesel traffic split. The use of a single CO<sub>2</sub>/CO ratio is problematic for multiple reasons. (1) the mix of gasoline versus diesel-powered vehicles varies spatially (e.g., on highway/city streets and in urban/rural areas). The diesel truck fraction tends to be much higher on major highways traveling through more sparsely populated rural areas (e.g., highway 401 outside of Toronto). The diesel CO<sub>2</sub>/CO ratio differs from the corresponding ratio for gasoline engines. Also (2) the emissions of CO are elevated during cold engine starting (and especially so during winter). Therefore the CO<sub>2</sub>/CO emissions ratio varies spatially and by time of day. The ratio should be higher on highways and lower in residential areas in the morning when vehicle engines are started under cold conditions. The method used in this study for estimating CO<sub>2</sub> emissions from vehicles (by ratio to CO) is therefore questionable and only provides a rough approximation to a more complex reality.

Editorial suggestions:

C2

Line 133, observational program Egbert: the word 'site' is missing

Watch sig figs in reporting emissions and calculating CO<sub>2</sub>/CO emission ratios. It is not reasonable to report emissions or emission ratios with 4-5 figures of accuracy.

Line 224: CO<sub>2</sub> emissions should be rounded to 23.5 Mt and CO emissions should be rounded to 219 kt (even that is optimistic precision) and the ratio should be reported as 107 kt CO<sub>2</sub>/kt CO.

The same excessive precision issue is again of concern at lines 239, 247, 274-75, 283, 311, 314, 318, and in Table 2

The paper uses too many acronyms, which makes the paper harder to read. Suggest omitting some of the more obscure ones such as PIA, BBTCA, and NEE (the last one is defined on line 290 but not used anywhere else in the manuscript).

Line 359: diel should be diurnal

Line 365: what does TAO stand for? Since the site was operational during the period of interest for the modeling, this site should be described as part of section 2.2 rather than suddenly appearing in the manuscript at this point.

In Figure 3, the resolution is coarse and it is not easy to discern differences among the three panels shown in this Figure. The first two panels (a) and (b) are almost indistinguishable. A legend showing the color scale is missing in this Figure.

In Figures 2 and 7, the marine contribution is negligible and should be omitted to simplify these figures. The point source panel in Figure 2 is not particularly helpful either.

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