

The study presents a technically detailed implementation of a Lagrangian model adapted for use with column data, X-STILT, focusing on emission ratios of CO:CO₂ on sub-city scales using XCO measurements from TROPOMI and XCO₂ measurements from OCO-2 and OCO-3. The level of technical detail is quite good (with a few exceptions, see comments below), and it is nice to see that the adapted X-STILT code has been made publicly available. The approach is interesting, particularly in its attempt to not make use of emissions inventories as part of the prior information. However some potential sources of error have been overlooked, particularly with respect to the high-frequency variability of the CO:CO₂ ratio, which seems to be assumed to be constant during the time between measurements. This is not something that the current study can really correct, but it needs to be included in the discussion as a clear limitation of the current results. Despite these limitations, the study is certainly appropriate for publication in ACP once these concerns are addressed. A good proof-reading of the paper is also needed before resubmission: while it was almost always clear what was meant, and the paper was well-written and easy to follow, there were lots of missing articles etc. that the native speakers among the co-authors could clear up quickly.

Major concerns:

The authors have gone to great pains to try to correct for temporal shifts between the measurements, considering the impact of the different meteorological conditions and averaging kernels. What was not taken into account is changes in the CO:CO₂ ratio over the course of a day. While this may not be as critical for heavy industry and power generation, other sectors (e.g. traffic) have highly heterogeneous emission ratios in time, depending also on traffic patterns. Having the XCO and XCO₂ measurements offset by even a couple of hours complicates this approach considerably, and might also cloud the proposed analysis of temporal trends in the emission ratio over the year. That does not mean that nothing can be learnt from this approach, only that this neglected error source needs to be explicitly described. In any case, GeoCarb data will make such analyses considerably easier in the future.

Minor concerns:

What the authors mean by “FF” needs to be made clear. The implication is that the emission signature of fossil fuels is being measured directly, which is clearly not the case. Emission from combustion, sure. There is no capacity to separate e.g. biofuel from fossil fuels in this approach, and this needs to be made clear.

L37: While this is true for some air pollutants such as CO, this is not true for many CO₂. In fact, increasing efficiency during combustion activities increases the amount of CO₂ and NO_x emitted (unless the latter is scrubbed) (e.g. Lama et al., 2020). The key to reducing emissions of greenhouse gases is to reduce combustion, period.

L223-224: Why use a rectangle as the source function? Was this to be “independent of emission inventories”, while still knowing that emissions are located within the city?

L231: Regarding the second point about excluding observations elevated by another city: yes, this excludes enhancements outside the model-defined urban plume, but what about enhancements from other cities that might also be contributing to enhancements within the urban plume? This seems to be the case for the XCO values shown in Figure 3c. Is there any way to correct for these values? Especially given that there do not seem to be XCO₂ values over the same area?

L235: Is there any way to mark on here which soundings were used to define the background? Some more quantification of the information would also be useful here. Which latitude range was used to define the background for the OCO-2 swath? In Figure 3a I don't see any soundings outside the urban

plume region... For 3b it is somewhat clearer, aided by the coastal cut-off (although point sources near the coast are clearly in the “background”, but how the area would be defined for Figure 3c is really vague. With this level of detail, the approach would be hard to replicate.

Section 2.2.4: It’s unclear how the observation uncertainty is computed here. Each sounding has a reported uncertainty that comes with the data product – is this the retrieval error? The “measurement noise” seems to be equated with the standard deviation of the retrievals within a TROPOMI footprint. But this isn’t really what is usually meant by measurement noise. It is not completely clear how these two types of errors (one per OCO-2 sounding and one per larger TROPOMI sampling) are combined, only that the retrieval errors are aggregated in a “standard-deviation-of-mean” manner, which sounds like it’s being divided by the square root of the number of soundings. This assumes that the measurements and their errors are independent of one another, which is not the case. This almost certainly underestimates the measurement uncertainty on the X_{CO_2} term. A formula here is certainly needed.

Technical/language comments:

L43-45: I’m confused by this sentence. Maybe should “Benefit” be “Benefiting”? In any case, the sentence should be rewritten to make it clearer, or even split into two sentences, starting with the second half, i.e. something like “The ratio... between tracers is reported. This has the benefit that errors ... cancel out.”

L46: would remove “their”

L50: ...difficult to detect?

L53-54: Rewrite this, something like: “Given its much longer lifetime, CO is much easier to interpret...” The “on the other hand” doesn’t fit here as written.

Figure 1 caption: “The x-axis indicates...”

L82: such gradient -> either “such a gradient” or “this gradient”

L87-88: in vertical -> in the vertical

L89: by the gaps in prior literature -> by gaps in the existing literature

L92: adopt or adapt?

L97: quantify accurately -> accurately quantify

L101: implication and limitation -> implications and limitations

L107: surrounded -> surrounding

L109: urban plume that is the spatial extent -> urban plume, defined as the area

L111: that estimated -> that are estimated

L113: requires estimate -> requires an estimate

L116: would remove “much”

L155: accounted for AK -> accounted for the AKs?

L156: As result -> As a result

L160: evaluations -> evaluation

L163: interfere -> interfere with; also: more explanations in Sect. -> for more explanation, see Sect.

Figure 2 caption: The citation for the Google Maps data in the last sentence doesn't sound quite right – please check what it's supposed to be (i.e. adopted the Google Maps what?)

L166: and atmospheric transport model -> and an atmospheric transport model

L175: for sounding-specific -> for the sounding-specific

L178-181: I think I understand what is meant here, but it's a bit hard to parse. When I hear “pathways” I'm thinking of chemical reactions, and I'm not sure what is meant by an air parcel being “tied” to somewhere, or correction terms being “attached” (perhaps “applied” would fit better)? In any case this should be rewritten for clarity.

L193: corresponding for -> corresponding to

L196: wind condition -> wind conditions

L200: by sounding -> by the sounding

L201: Because AK -> Because the AK

L204: If -> By

L208-209: I guess the meteorological conditions and the AK profile are specific to the sounding, not just the sensor?

L209: condition -> conditions

L216: combines -> combine

L216-217: This seems a bit backwards – isn't the first method more independent of information about emissions, unlike the two modelling-based approaches?

L218-219: improve it over what?

L222: soundings within -> soundings as within

L258-262: I would split this into two sentences.

L265: lasts for -> ranges from

L267: are -> is

L273: Observed uncertainty of XCO₂ are -> Observation uncertainty of XCO₂ is

Figure 4: Please add a coastal outline in panels c and f to make it easier to interpret. (Masking water would also be an option.)

Figure 4 caption: lightgray should be two words.

L301: I think a word is missing. Maybe: “Those industry coverage *maps* are then convolved...”

L311: I would remove “originating” here, it is more confusing than helpful.

L313: When you write “too low valid soundings” do you mean “too few valid soundings” or “too low enhancements”?

L313-314: remove “the few”

L319: interfered by wind shift -> affected by shifting winds

L329-330: Recommended change: "Again, the colored contours and curves in Fig. 5 indicate neither the intensity of concentrations nor flux fields (as no prior emissions are used), but rather the likelihood of urban plumes determined solely by atmospheric dispersion."

L332: but problematic -> but becomes problematic

L335: remove "cases"

Figure 5: in figure label, it should be "caution" rather than "cautions"

L364: I would suggest replacing "atmospheric movement" with "transport". Also, something seems to have gone wrong with many of the subscripts in this paragraph (e.g. E_{CO} instead of E_{CO} in LaTeX syntax).

Figure 6 caption: The second sentence and the second-last sentence seem to repeat the same information. Remove one? Also, it is mentioned here that only regression lines with positive slopes were chosen from the Monte Carlo experiment. What proportion of these lines needed to be removed?

Figure 6: the γ_{bio} term shown on the plots should be in units of ppm, right? Also, is this somehow different from the ΔX_{bio} discussed previously? If not, please make this consistent.

L385: remove second comma, also change "them" to "these overpasses".

L392: in time -> in the time

L406: tend -> tends

L408 (and elsewhere): perhaps "industry-dominated" might be more appropriate in some places than "industry-dominant"?

L432: i.e., less -> i.e., those less

L452: the Solar-Induced -> remove "the"

L455: function -> functional

L456: either "interfering" -> "interfering with" or "interfering" -> "affecting"

L456: "wind directional shift induced by" -> "the shift in wind direction due to"

L464: spontaneously -> simultaneously

L464: do you mean "a future geostationary satellite (i.e. GeoCarb)" or "future geostationary satellites"?

Figure 8 caption: Text could use a bit of work. Suggestion: "indicate the urban plumes between two times differ significantly that a simple plume rotation fails to fix" -> "indicate that that urban plumes between the two overpass times differ so much that they cannot be brought into agreement with a simple plume rotation". Also: what is the meaning of θ_1 and θ_2 ? Are they shifted or not?

L479: begun -> began

L483: take -> takes

L492: to coarser -> to the coarser

L494: may locate -> may be located

L500: against OH -> against the OH

L503: contributed to 21.2% -> contributed 21.2%

L503: but negligible -> but a negligible

L504: season, the -> season, and as such the

L504: remove "likely", "encapsulated" -> "included", "yield" -> "have"

L506-507: "...whether ... remains unclear" -> "it is unclear whether AVOCs... of interest."

L512: "can help" -> "to"

L513-514, and L521: inventory -> inventories

L515: footprint -> footprints

L520:

L527: in informing locations of -> to provide information about

Supplement:

Figure S1 caption: average -> averaged

Figure S3 caption: unique to each satellite sounding given unique -> unique to each satellite sounding, giving a unique

Figure S3 caption: Column footprint -> The column footprint

Figure S4 caption: "these resultant normalized fraction $P_{ind}(x, y)$ informs the influence on the observation at a given sounding (white rectangle) due to heavy industry. Lighter the color, stronger impact from heavy industry in LA." -> "these resultant normalized fractions $P_{ind}(x, y)$ show the influence of heavy industry on a given sounding (white rectangle). The lighter the color, the stronger impact from heavy industry."

Figure S5 caption: "of urban-background" -> "of the urban-background"; "Since biogenic" -> "Since the biogenic"; "Two sets of footprint totals" -> "The two sets of footprints"; "difference in AK" -> "difference in the AKs"; "Higher the footprint ratio, larger the discrepancies" -> "The higher the footprint ratio, the larger the discrepancy"

Figure S6 caption: "during two" -> "during the two"

Figure S7 (and others): Is the date format in the plots (YYYYMMDD) consistent with the Copernicus guidelines?

Figure S7 caption: "close to the noon, daytime carbon sink dominant leading" -> "close to noon, the daytime carbon sink dominates, leading"; "nighttime respiration dominant," -> "respiration dominates the biogenic fluxes," (I would remove "nighttime" because winter cases are also mentioned.)

Figure S9 caption: "with or without the account of the urban-rural biogenic gradient over Zibo on June 21, 2020" -> "over Zibo on June 21, 2020, with and without taking the urban-rural biogenic

gradient into account”; “light grey shading denote” -> “light grey shading denotes”; “Such positive gradient” -> “Such positive gradients”

Figure S10 title: A bit awkward, would suggest “Log-normal distributions of ER_{CO} [ppb-CO / ppm/CO₂]

Figure S10 caption: “on log-normal” -> “on a log-normal”; “stewed” -> “skewed”; Also, aren’t the values skewed towards the higher end (i.e. positive skew or right-skewed) if the mean is higher than the median?

Figure S11: Should the title for panels c) through f) be Sectoral CR_CO or ER_CO? Also, I guess panel f) should be marked “residential” rather than “resident”? (Perhaps “road traffic” might also be better for d)...))

Reference in the review:

Lama, S., Houweling, S., Boersma, K. F., Eskes, H., Aben, I., Denier van der Gon, H. A. C., Krol, M. C., Dolman, H., Borsdorff, T., and Lorente, A.: Quantifying burning efficiency in megacities using the NO₂/CO ratio from the Tropospheric Monitoring Instrument (TROPOMI), Atmos. Chem. Phys., 20, 10295–10310, <https://doi.org/10.5194/acp-20-10295-2020>, 2020.