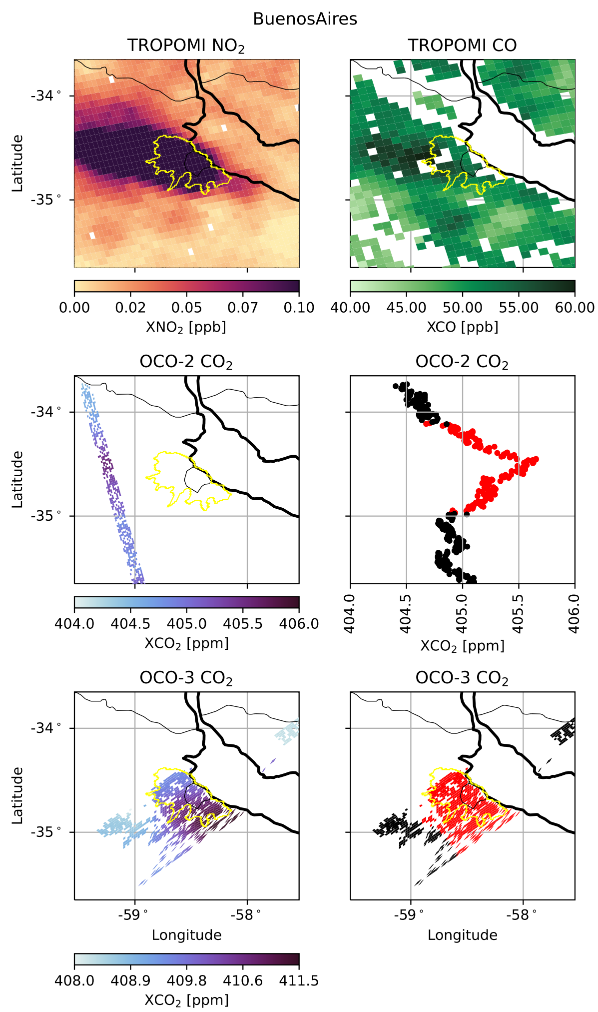
Response to Anonymous Referee #1

We thank Referee #1 for helpful comments and suggestions. We have addressed each of the comments below. Referee comments are in *red italics* and our responses are in Roman font.

> *The methodology section would benefit greatly from the inclusion of more figures to illustrate the various steps of the analysis and aid the reader's understanding. I find this particularly important since several satellite products are used, each with distinct spatial coverage and overall data handling. For example, the differences in spatial coverage between the various satellite are not shown, nor is the colocation of OCO-2 and TROMPOMI/OCO-3 SAM data (section 3.1), or the masking that results from taking only using data from the sparser production (section 3.3.3). Similarly, the plume extraction presented in section 3.2 is not shown, nor is the impact of smoothing on the TROPOMI/OCO-3 data (section 3.3.1). At a minimum, a figure showing the different spatial sampling of the three satellites should be included.*

We have added an additional figure (below) to more clearly show the ground tracks of the three satellites we have used in the paper. These are examples of the various satellite instrument ground tracks over Buenos Aires taken on 11 January 2019 (top 4) and 19 September 2019 (bottom 2). Red points indicate the urban enhancement plume.



There appear to be two distinct methods used for the enhancement calculation depending on which product is used in the analysis; a cross section taken downwind of the city (OCO) and the enhancement over the entire bounding area for the city (TROPOMI). The manuscript jumps around between these various methods, which is difficult to follow. I suggest clarifying the these two complementary methods within the text or with some sort of figure summarizing the methods.

Yes, there are distinct methods as the referee has described depending on which ratio is being computed and from which instruments. We have clarified the text by adding the following sentences:

OCO-2 CO_2: "To compute enhancement ratios, coincident TROPOMI CO and NO_2 enhancements are selected at the locations of the OCO-2 ground track."

OCO-3 CO_2: "Coincident TROPOMI enhancements are selected at the locations of the OCO-3 SAM measurements."

NO_2:CO: "... only direct observation of the cities within their bounding areas are considered when deriving NO_2:CO enhancement ratios."

Specific comments

Representativeness of enhancement ratios – Due to a limited number of overpasses (e.g. 1 for Toronto), how representative do you think some of these ratios are to the timescale of the inventories? I believe a brief discussion concerning this is warranted since the number of overpasses per city varies, while it appears conclusions about the validity of inventories are drawn from all cities regardless of the number of overpasses used in the ratio calculation.

We have computed enhancement ratios for each overpass individually for each city to quantify the variability in enhancement ratios across overpasses. Most of the overpasses are in the summer, which would bias our results to summertime emissions, and, as you point out, in other cities, such as Toronto, the number of overpasses is extremely limited. As the satellite records lengthen, this representativeness bias should reduce somewhat. At the moment, however, most of the inventories are reported annually, so sub-sampling the inventories is not yet possible. We have added a brief discussion of the representativeness issue in the Discussion:

"The overpasses that successfully pass our filtering criteria are biased toward sunnier conditions and are most often collected in summertime, and some sites have very few overpasses (e.g., Toronto). If the enhancement ratios change seasonally, as expected, this type of analysis could cause a representativeness error, in which the comparisons between the measured enhancement ratios and the reported annual inventory ratios are systematically biased. Currently, the EDGAR and MACCity inventories, which provide CO and NO_2 emissions, do not report sub-annual emissions, so comparing to seasonal inventory ratios is not possible. With longer satellite time series providing more opportunities for wintertime enhancement ratios, we will be able to compute robust annual enhancement ratios to compare with the annual inventories."

Do you expect the biosphere to impact your CO2 or CO enhancements, and subsequently your ratios?

Yes, the biosphere will impact our enhancement ratios, but it should be a relatively small effect. Wu et al. [2022] calculate CO:CO_2 emission ratios over 4 cities including Los Angeles. In the paper they derive fossil fuel and biogenic anomalies using the X-STILT transport and SMuRF models. They find biogenic XCO_2 anomalies far smaller than those of fossil fuels. They provide the overpass example of Zibo on 21 June 2020, which contains the largest biogenic urban–background contrast that they studied. The biogenic anomalies ranged from 0 to 0.4 ppm while the sounding-level fossil fuel CO_2 enhancements ranged from 2 to 7 ppm. They find that typically the aggregated biogenic anomaly (i.e., summed for all footprints) stays low, with an absolute value of < 0.3 ppm. The aggregated fossil fuel CO_2 enhancements will be roughly 2 orders of magnitude larger (figure 2, S7, S11). We have added the following paragraph to the discussion:

"One potential source of error in this analysis is from biogenic emissions of CO and cycling of CO_2, but we expect these effects will be small. Under the assumption that there is less vegetation within urban boundaries than outside the urban region, biogenic CO and CO_2 emissions have the potential to affect the urban-rural gradients, especially during the growing season. According to recent studies, however, these gradients are significantly smaller than the enhancements we measure, suggesting that urban CO and CO_2 enhancements are dominated by fossil fuel emissions [Plant et al., 2022; Wu et al., 2022]. Further modeling analysis will be necessary to apply this kind of analysis to smaller emission sources."

> *Lines 191-194 – How are the secondary sources identified?*

The secondary sources from cities are identified using the European Commission Joint Research Centre's (EC-JRC) Global Human Settlement layer Urban Centres Database (GHS-UCDB). The secondary sources from power plants are identified using the Carbon Monitoring for Action (CARMA) database. We have added this to the text.

> *Lines 194- 199 – What explains the discrepancy between the MERRA-2 winds and the plume direction, and the resulting variability in overpass retention rates? Is it errors in the wind direction and/or issues with the automatic filtering scheme since manual inspection and correction is required?*

The MERRA-2 model output is at a spatial resolution of 0.5° latitude × 0.625° longitude with a 3-hourly temporal resolution. We do not expect the wind direction to always be accurate at city scales and at the time of the overpass, and the automatic filtering simply ensures that the measurements are downwind of the source, and that the wind rotation is not too large. We have added the following text:

"Errors in wind direction can be caused by the inability of the coarse model resolution to resolve local topography, or if the 50-m winds are not representative of the winds at the local plume height. The wind rotation we perform should at least partially correct for both these errors."

> *Figure 1 – How are the red points assigned to the city? Without more information or geographical details about the city and location of the overpass, it is hard to interpret.*

The new figure we have added shows the city and OCO-2 ground track, which we hope makes the assignment of the plume (red points) clearer. In what was previously Figure 1 (Figure 2 in the updated manuscript), we added clarifying text to the figure caption:

"Red points indicate the enhancement and are where the Gaussian plume intersects with the OCO-2 ground track."

> *Line 273 – What is allowed to change in the bootstrap? Are the anomalies of each species within each overpass resampled?*

Within the bootstrap, *pairs* of anomalies are randomly selected and the slope is computed from those anomaly pairs. This selection process is repeated to compute the average slope and variability of the slope. This has been clarified in the paper in this sentence:

"Bootstrapping is a re-sampling technique in which random pairs of anomalies are drawn with replacement and fit independently, and has been used in previous enhancement ratio studies..."

> *Lines 284-285 – "As we are correcting the enhancement ratios, the effect of dispersion cancels out in the ratio." This sentence is unclear to me. What dispersion?*

We were referring to the dispersion of the plume as it travels away from the city toward the OCO-2 ground track, but upon further reflection, this sentence is unnecessary in the manuscript and we have removed it.

> *Figure 2 – The color scale in the 'Background' plot is hard to interpret using the color bar. I understand this the same color bar as the observed data which has a larger spread of values, but I cannot tell if it is all one value for the background or not. This might be an issue with my screen, but it is something to think about.*

We have used a different set of colour scales for the plots in this figure.

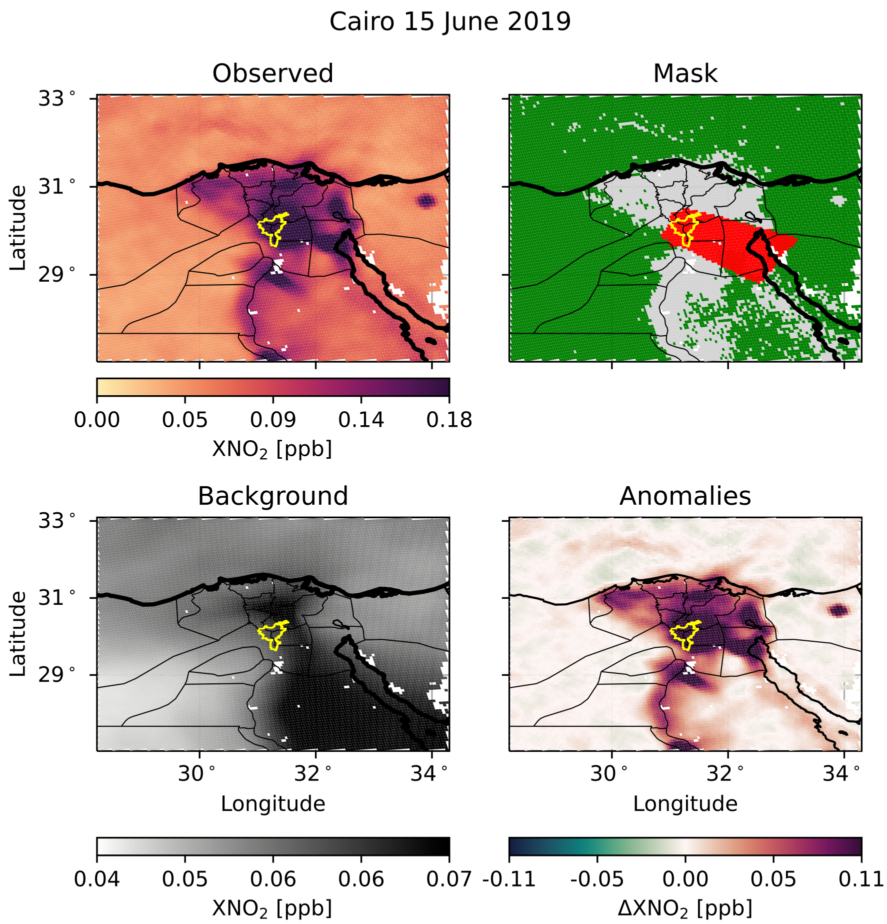


Figure 4 & Section 3.4 – It was not until I saw Figure 4 that I realized that a single regression is calculated for all anomalies across multiple days. I would suggest explicitly stating this in the text since, in contrast, Figures 1 & 2 are for single days. In this analysis, are the number of anomaly data points that same for each overpass? If not, is there potential for a subset of overpasses with higher density of data (i.e. more points) to drive the combined regression?

This is now explicitly stated in the text in §3.4. The number of points is not the same for each overpass, and this will generally bias the number of points to summer days or days that are less cloudy. We have added a brief discussion of representativeness error in the Discussion section.

"To determine enhancement ratios, we aggregate all overpasses for a given city and regress one set of anomalies onto the other using a reduced major axis regression..."

"The overpasses that successfully pass our filtering criteria are biased toward sunnier conditions, and are most often collected in summertime, and some sites have very few overpasses (e.g., Toronto). If the enhancement ratios change seasonally, as expected, this type of analysis could cause a representativeness error, in which the comparisons between the measured enhancement ratios and the reported annual inventory ratios are systematically biased. Currently, the EDGAR and MACCity inventories, which provide CO and NO_2 emissions, do not report sub-annual emissions, so comparing to seasonal inventory ratios is not possible. With longer satellite time series providing more opportunities for wintertime enhancement ratios, we will be able to compute robust annual enhancement ratios to compare with the annual inventories."

Figure 5-7 – The number of overpasses that each measurement ratio is based upon should be included in the figure. It seems that this information is at least partially available in the appendix, so another option would be to include a reference to those tables in the caption.

We have update Tables 2 & 3 to include the number of overpasses used in each ratio.

Table 2. All enhancement ratios derived using OCO-2/3 and TROPOMI. Overpasses must individually have sufficient linear dependence ($R > 0.2$). Cities without any such overpasses are marked by a dash.

City	No. of OCO-2 / 3 Overpasses	$CO:CO_2$ Enhancement Ratio [ppb:ppm]	No. of OCO-2 / 3 Overpasses	$NO_2:CO_2$ Enhancement Ratio [ppb:ppm]	No. of TROPOMI Overpasses	$NO_2:CO$ Enhancement Ratio [ppb:ppb]
Ahmadabad	3	12.7 ± 1.1	-	-	-	-
Alexandria	-	-	5	0.39 ± 0.04	30	0.018 ± 0.002
Baghdad	2 / 2	6.3 ± 0.3	6 / 2	0.89 ± 0.04	67	0.176 ± 0.013
Buenos Aires	6 / 4	7.9 ± 0.3	6 / 4	0.57 ± 0.02	39	0.048 ± 0.004
Cairo	12 / 2	14.0 ± 0.9	12 / 2	0.49 ± 0.02	84	0.037 ± 0.001
Chicago	5	13.3 ± 0.7	5	0.61 ± 0.03	44	0.038 ± 0.005
Dallas	3 / 1	3.7 ± 0.2	3 / 1	0.18 ± 0.01	57	0.032 ± 0.002
Delhi	3 / 1	13.0 ± 0.5	4 / 1	0.79 ± 0.04	46	0.017 ± 0.001
Guangzhou	3	13.2 ± 1.6	-	-	23	0.014 ± 0.002
Houston	-	-	5 / 1	0.36 ± 0.02	54	0.015 ± 0.001
Indianapolis	7	13.2 ± 0.9	7	0.41 ± 0.02	32	0.018 ± 0.003
Johannesburg	11	15.4 ± 0.5	11	2.54 ± 0.16	62	0.180 ± 0.012
Lahore	-	-	-	-	42	0.021 ± 0.002
Las Vegas	10 / 1	8.1 ± 0.5	10 / 1	0.41 ± 0.02	58	0.021 ± 0.001
London	1 / 1	5.7 ± 0.3	1 / 1	0.50 ± 0.01	47	0.071 ± 0.006
Los Angeles	7 / 2	8.2 ± 0.3	7 / 2	0.32 ± 0.01	84	0.031 ± 0.001
Madrid	-	-	9 / 1	0.92 ± 0.04	49	0.097 ± 0.007
Mexico City	7 / 5	19.1 ± 0.7	7 / 5	1.24 ± 0.05	50	0.041 ± 0.003
Moscow	5	12.5 ± 0.7	5	1.13 ± 0.06	53	0.082 ± 0.007
Paris	4 / 2	12.4 ± 1.0	4 / 2	0.70 ± 0.04	39	0.052 ± 0.007
Phoenix	14 / 6	6.6 ± 0.2	14 / 6	0.30 ± 0.01	74	0.028 ± 0.001
Riyadh	8	11.2 ± 0.5	-	-	52	0.359 ± 0.030
San Francisco	-	-	7	0.18 ± 0.01	62	0.009 ± 0.001
Seoul	-	-	4	0.698 ± 0.04	45	0.066 ± 0.008
Tehran	10 / 3	23.9 ± 1.2	10 / 3	1.78 ± 0.16	86	0.051 ± 0.001
Toronto	1	10.6 ± 1.2	1	0.80 ± 0.07	39	0.026 ± 0.005
Vienna	2	7.2 ± 0.6	-	-	-	-

Table 3. All $NO_2:CO_2$ and $NO_2:CO$ enhancement ratios derived using OCO-2/3 and TROPOMI, including NO_2 lifetime correction. Overpasses must individually have sufficient linear dependence ($R > 0.2$). Cities without any such overpasses are marked by a dash.

City	No. of OCO-2 / 3 Overpasses	Average Wind Speed for Correction (m/s)	$NO_2:CO_2$ Enhancement Ratio [ppb:ppm]	No. of TROPOMI Overpasses	Average Wind Speed for Correction (m/s)	$NO_2:CO$ Enhancement Ratio [ppb:ppb]
Alexandria	5	5.13	0.44 ± 0.10	30	6.30	0.019 ± 0.005
Baghdad	6 / 2	5.79	1.12 ± 0.23	67	6.91	0.210 ± 0.044
Buenos Aires	6 / 4	4.33	1.26 ± 0.25	39	6.58	0.074 ± 0.015
Cairo	12 / 2	4.39	0.85 ± 0.17	84	5.02	0.092 ± 0.018
Chicago	5	4.49	1.20 ± 0.24	44	4.75	0.064 ± 0.014
Dallas	3 / 1	4.41	0.33 ± 0.07	57	4.37	0.119 ± 0.024
Delhi	4 / 1	2.15	5.83 ± 1.17	46	4.65	0.047 ± 0.010
Guangzhou	-	-	-	23	3.88	0.286 ± 0.057
Houston	5 / 1	4.94	0.74 ± 0.15	56	3.84	0.058 ± 0.012
Indianapolis	7	4.45	0.55 ± 0.11	32	4.29	0.032 ± 0.007
Johannesburg	11	5.20	2.77 ± 0.58	62	5.55	0.193 ± 0.041
Lahore	-	-	-	42	2.73	0.072 ± 0.014
Las Vegas	10 / 1	4.44	0.89 ± 0.18	58	3.87	0.052 ± 0.011
London	1 / 1	6.86	0.63 ± 0.13	47	4.38	0.118 ± 0.024
Los Angeles	7 / 2	3.59	0.68 ± 0.14	84	4.34	0.058 ± 0.012
Madrid	9 / 1	5.22	1.71 ± 0.34	49	3.92	0.208 ± 0.042
Mexico City	7 / 5	2.80	3.79 ± 0.76	50	2.56	0.145 ± 0.029
Moscow	5	5.92	2.08 ± 0.42	53	5.42	0.159 ± 0.033
Paris	4 / 2	5.44	1.29 ± 0.26	39	3.51	0.188 ± 0.038
Phoenix	14 / 6	4.17	2.01 ± 0.41	74	3.59	0.386 ± 0.078
Riyadh	-	-	-	52	7.30	0.505 ± 0.105
San Francisco	7	4.73	0.22 ± 0.05	62	4.80	0.010 ± 0.002
Seoul	4	7.57	1.42 ± 0.29	45	3.55	0.303 ± 0.061
Tehran	10 / 3	3.26	3.89 ± 0.79	86	5.28	0.079 ± 0.016
Toronto	1	2.88	2.47 ± 0.50	39	4.29	0.056 ± 0.012

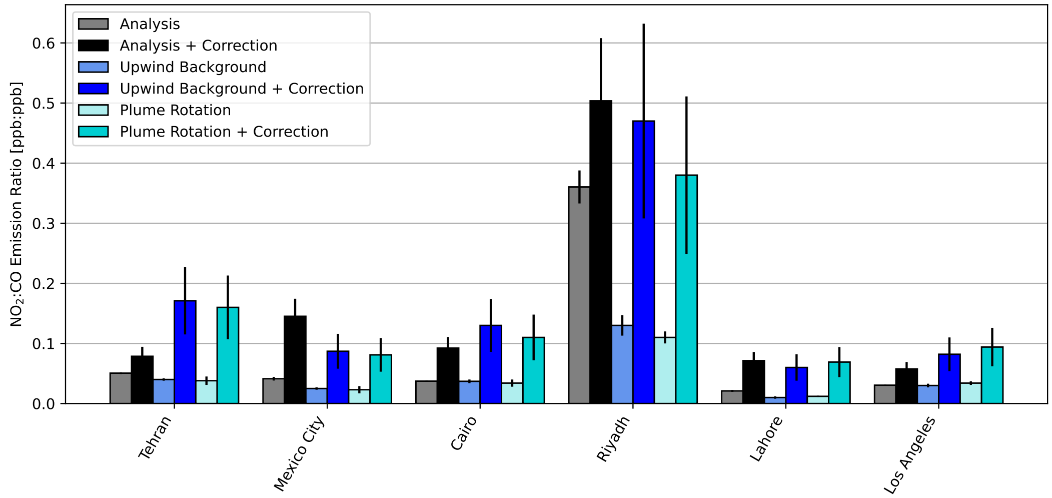
Figure 5 – I cannot see where the "light blue areas" are on top of the measured ratios. I see black error bars, so perhaps this is a typo.

This is indeed a typo that refers to an older version of the figure. It has been fixed in the manuscript.

Lines 355-358 – Is the comparison for cities both in this work and Lama et al. shown anywhere, in something like a figure or table? If not, I believe this should be added, possibly to the appendix. If this comparison is buried into one of the figures, a reference to it should be added to the text.

We have added the following figure to the paper to show the comparison between Lama et al. (2020) and our work. We have also modified our discussion in section 4.2. Our previous discussion was with reference to incorrect values in the appendix of Lama et al. (2020). The authors have been contacted and are in communication with the journal to fix this issue. We include their corrected numbers in our paper.

With the corrected numbers from Lama et al., our results show good agreement, within the uncertainties, of corrected $NO_2:CO$ enhancement ratios at all cities except Tehran. This has also been added to our discussion.



In the introduction, you mention the TIMES scaling of CO2 by Nassar et al. Do you apply those scale factors to account for the diurnal variability of CO2, since your measurements are based on afternoon overpasses? You mention this correction (the magnitude of which is not included) in section 5.2 for LA and Indianapolis, however, it is not clear if these corrections are applied to the ratios shown in Figures 5 &6.

Yes, the TIMES scaling is added to the ratios computed from the inventories for the reasons stated above. We have added clarification in the text in §2.5.

Lines 425-429 – You present the NO2 emission estimates based on ratios with and without the NO2 lifetime correction. The reader should not assume the un-corrected emission rate is accurate, right? If this is the case, I would suggest you state that because as written it is not clear which value for NO2 is the one the reader should remember.

That is correct. We have rephrased the sentence:

"... we estimate emissions of NO_2 within the SoCAB to be 89 $\pm$ 17 GgNO_2/yr (after the NO_2 lifetime correction is applied), which agrees with the CARB estimate for 2015 (105 GgNO_2/yr). However, it is smaller than the annual EDGAR estimate of 132 GgNO_2/yr, and larger than the MACCity estimate of 43.3 GgNO_2/yr..."

Line 455 – underestimations of which species?

We are referring to CO. This has been clarified in the text.

Table 2 – What metric is used to discriminate a "poor" linear relationship? Low correlation coefficient, low R?

We use $R < 0.2$ as the metric. We've added this to the Table 2 & 3 captions.

Tables 2, 3 – You should include how many TROPOMI overpasses are used to generate the NO2:CO enhancement ratios for each city.

These values have been added to tables 2 & 3.

References

Wu, D., Liu, J., Wennberg, P. O., Palmer, P. I., Nelson, R. R., Kiel, M., and Eldering, A.: Towards sector-based attribution using intra-city variations in satellite-based emission ratios between CO2 and CO, Atmos. Chem. Phys., 22, 14547–14570, https://doi.org/10.5194/acp-22-14547-2022, 2022.

Plant, G., Kort, E. A., Murray, L. T., Maasakkers, J. D., Aben, I.: Evaluating urban methane emissions from space using TROPOMI methane and carbon monoxide observations, Remote Sensing of Environment, Volume 268, 2022, 112756, ISSN 0034-4257, https://doi.org/10.1016/j.rse.2021.112756.