TROPOMI NO_2 emissions from mining and other industrial activities in the Copperbelt (DRC and Zambia)

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SUPPLEMENT

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S1 Effect on TROPOMI Emissions of ERA5 Wind Data Uncertainty

We have quantified the error in TROPOMI-derived NO_2 raw emissions (i.e., emissions before background removal) due to uncertainties in the ERA5 wind data. Wind uncertainty was estimated from the spread of the 10-member ensemble of ERA5 data assimilation. We recalculated annual TROPOMI emissions for 2020 over the Copperbelt study area (-10.5^{o}N to -13.5^{o}N, 24.5^{o}E to 29.5^{o}E) after perturbing the wind data by first adding and then subtracting the wind spread. These two sets of perturbed results were then compared to unperturbed TROPOMI emissions for the same year and region. Results are shown in Fig. S1 and summarized in Table S1. Differences between unperturbed and perturbed emissions are in all cases below 4 %.

S2 Annual TROPOMI Maps of VCD and Emissions

Annual maps of TROPOMI NO_2 VCD (Vertical Column Density) were produced for the years between 2019 and 2022 from daily TROPOMI VCD data (Fig. S2). The maps show six distinct emission point sources which coincide with copper or copper-cobalt mines (points a to d) and cities (points e and f). The VCD maps show inter-annual variability in VCD among the point sources, while background VCD values remain close-to-constant between 2019 and 2021. Background values appear to be higher in 2022.

Figure S3 shows annual maps of raw TROPOMI emissions. These maps show that the magnitude of the emissions released from each point source changes from year to year. Background emissions do not show strong changes.

S3 Are Lubumbashi and Ndola Emissions from Urban Activity Alone?

To test the hypothesis that background-removed TROPOMI emissions from Lubumbashi and Ndola were not produced by urban activity alone, we identified two nearby cities of similar population: Mbuji-Mayi and Kitwe, respectively. We calculated annual

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emissions for these two cities using the methodology described in Section 3. Results are summarized in Fig. S4 and Table S2.

Background-removed NO_2 emissions from Lubumbashi are higher than those from Mbuji-Mayi by $\sim$ 90 %, even though the population of the former is lower by $\sim$2 %. Similarly, while its population is lower by $\sim$27 %, emissions from Ndola surpass those from Kitwe by 40-80 %, depending on the year, and display inter-annual variations which do not correlate with changes in population.

These findings are consistent with our hypothesis that emissions from Lubumbashi and Ndola are not the result of urban activity alone.

S4 TROPOMI Emissions versus CAMS-GLOB-ANT v5 Inventory Emissions

We compared CAMS-GLOB-ANT v5 inventory data to TROPOMI-derived NO_2 emissions. Only 2019-2021 inventory data were analyzed because those from 2022 were unavailable at the time of writing. The inventory includes emissions from the following sectors: power generation, industrial processes (including mining), road and non-road transportation, residential, fugitive fuel emissions, solvents application and production, solid waste and wastewater handling, agriculture livestock, agriculture soils, and agriculture waste burning. In order to compare inventory emissions to background-removed TROPOMI emission, we aggregated the emissions from all sectors excluding agriculture livestock, agriculture soils, and agriculture waste burning, since those would be part of the background in mines and cities. We could not compare inventory to raw (i.e., non background-removed) TROPOMI emissions because the inventory does not include other possible background emissions (from wildfires, non-agriculture soils, and lightning).

We aggregated monthly inventory emission values as explained above and converted them from NO_x (the original) into NO_2 ($NO_2 = NO_x / 1.32$; Beirle et al. (2019), Dix et al. (2022)). The highest inventory value among the nine data points coinciding spatially with each point source was selected. The selected inventory values were used to calculate annual means for each point source. Results are presented in Table 1 in the main body of this paper and summarized in Fig. S5.

Emissions from the Copperbelt mines analyzed are not well represented in the inventory (Table 1 and panels a to d in Fig. S5). Inventory values are consistently lower than the TROPOMI emissions (by between 61 and 96 %) and do not show the annual trends identified by TROPOMI. Differences between inventory and TROPOMI emissions for cities (Table 1 and panels e to h in Fig. S5) are between -62 to +1869 %. The inventory overpredicts emissions for Mbuji-Mayi and Kitwe, two cities that we infer have no additional industrial emissions, and most years underpredicts emissions for Lubumbashi and Ndola, where we hypothesize that highly-emittant industrial processes take place.

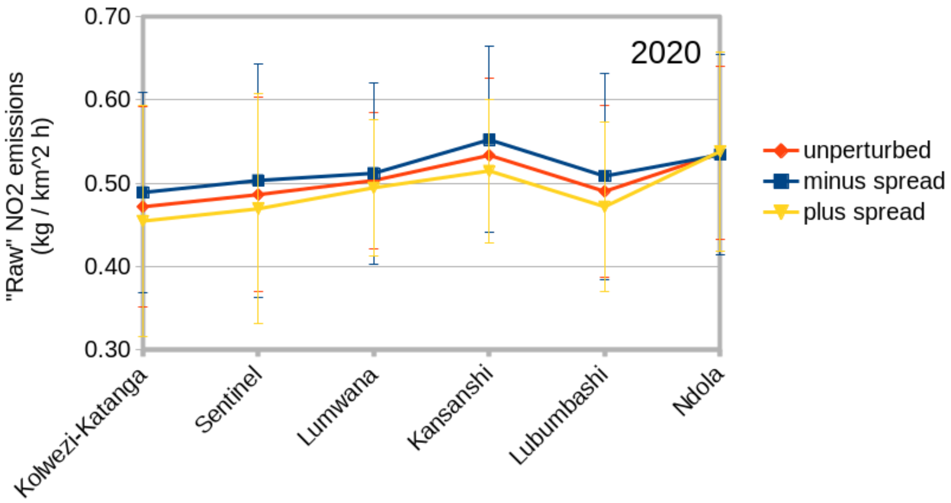

Figure S1. Effect of wind uncertainty (i.e., the spread of the ERA5 10-member wind ensemble) on raw TROPOMI-derived NO_2 emissions for the six point sources analyzed. For 2020 only. Vertical bars show standard deviation of emissions.

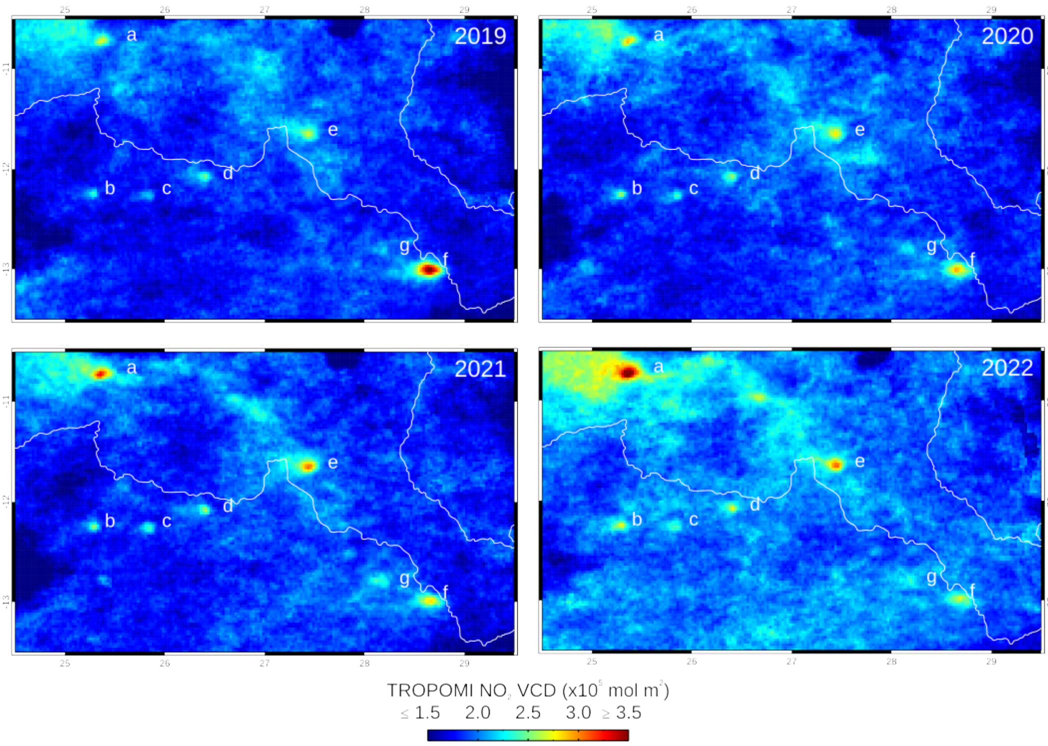

Figure S2. Annual means of TROPOMI NO_2 VCD for the Copperbelt study region. Labels a through f show the six point sources analyzed. a: Kolwezi-Katanga Mines, b: Sentinel Mine, c: Lumwana Mine, d: Kansanshi Mine, e: Lubumbashi City, f: Ndola City. Label g shows location of Kitwe City. White line indicates border between the DRC and Zambia.

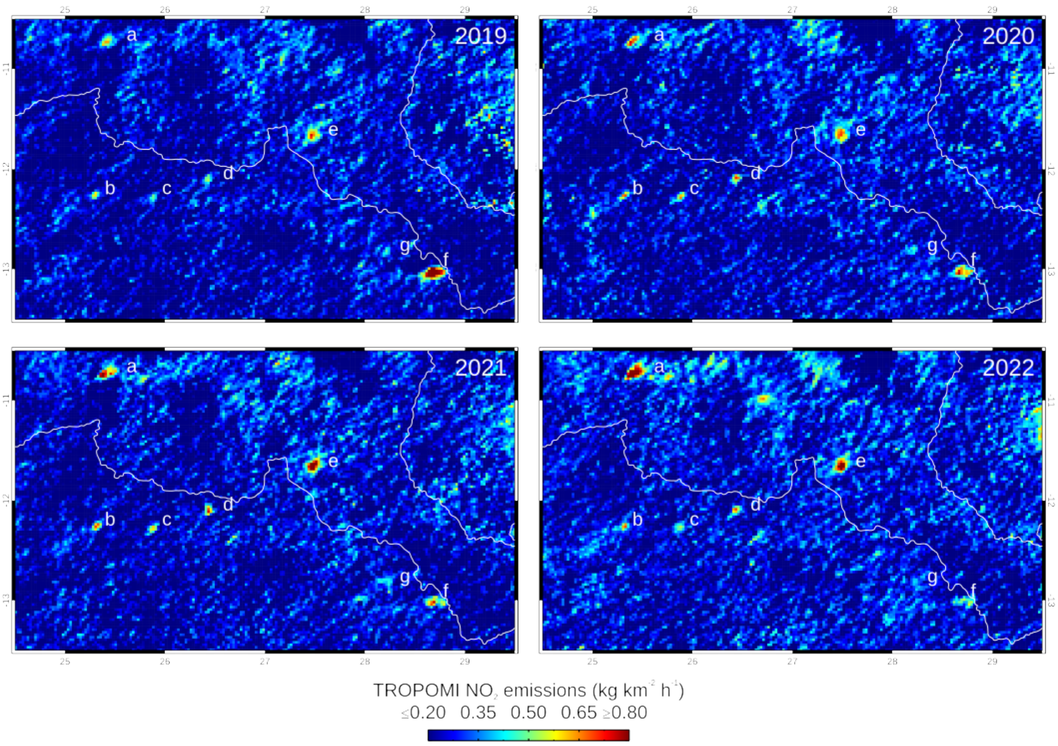

Figure S3. Annual means of TROPOMI-derived NO_2 raw emissions for the Copperbelt study region. Labels a through f show the six point sources analyzed. a: Kolwezi-Katanga Mines, b: Sentinel Mine, c: Lumwana Mine, d: Kansanshi Mine, e: Lubumbashi City, f: Ndola City. Label g shows location of Kitwe City. White lines indicate border between the DRC and Zambia.

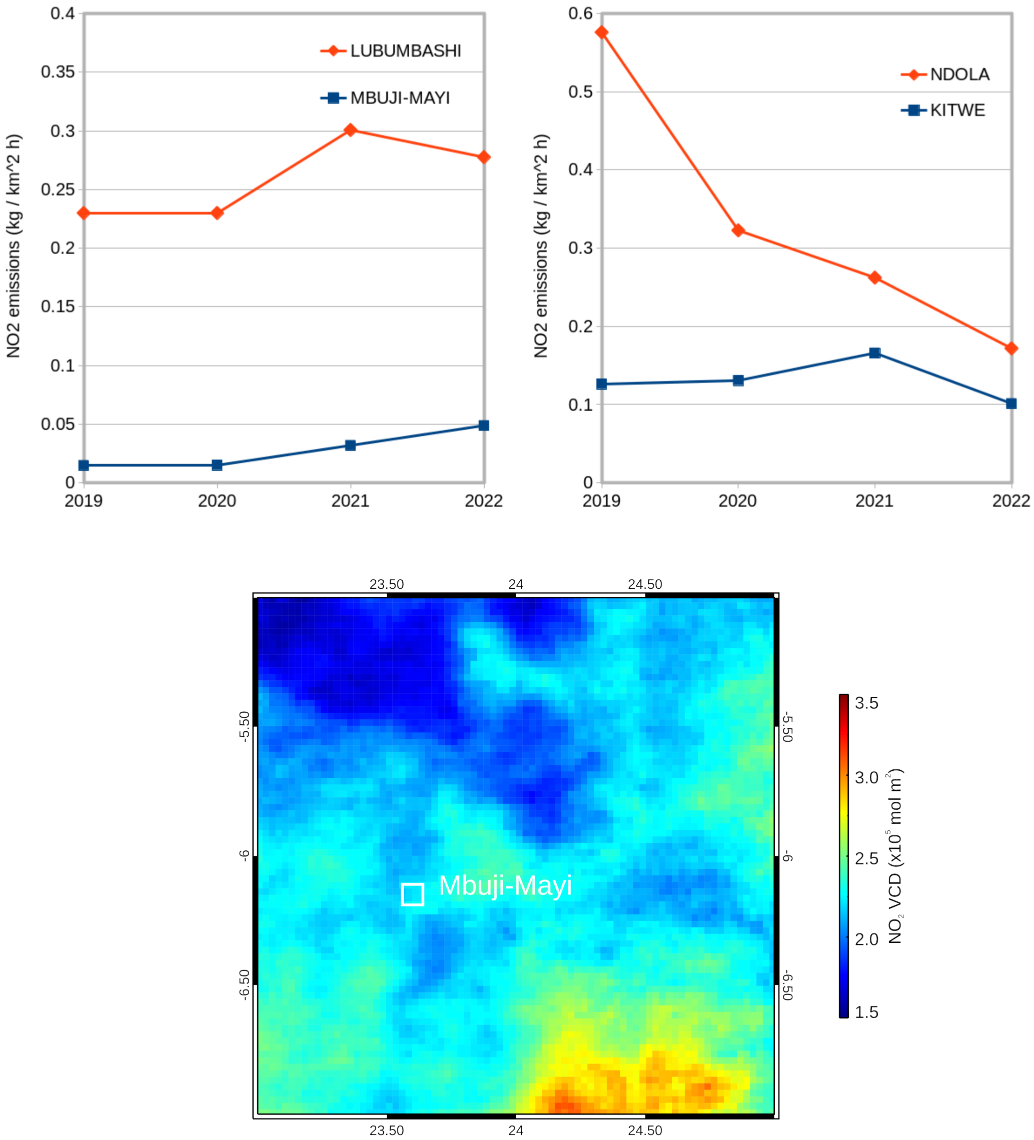

Figure S4. (Top) Time series of annual TROPOMI-derived, background-removed NO_2 emissions for two Copperbelt point sources corresponding to cities (Lubumbashi and Ndola) paired with cities of similar population (Mbuji-Mayi and Kitwe, respectively). The magnitude of the emissions from the two point sources amply surpasses that of the reference cities. The inter-annual trends in emissions from the two point sources cannot be explained by population changes (Table S2). (Bottom) TROPOMI NO_2 VCD shows no plume or enhancement at Mbuji-Mayi which, despite its large population, is a remote and isolated enclave.

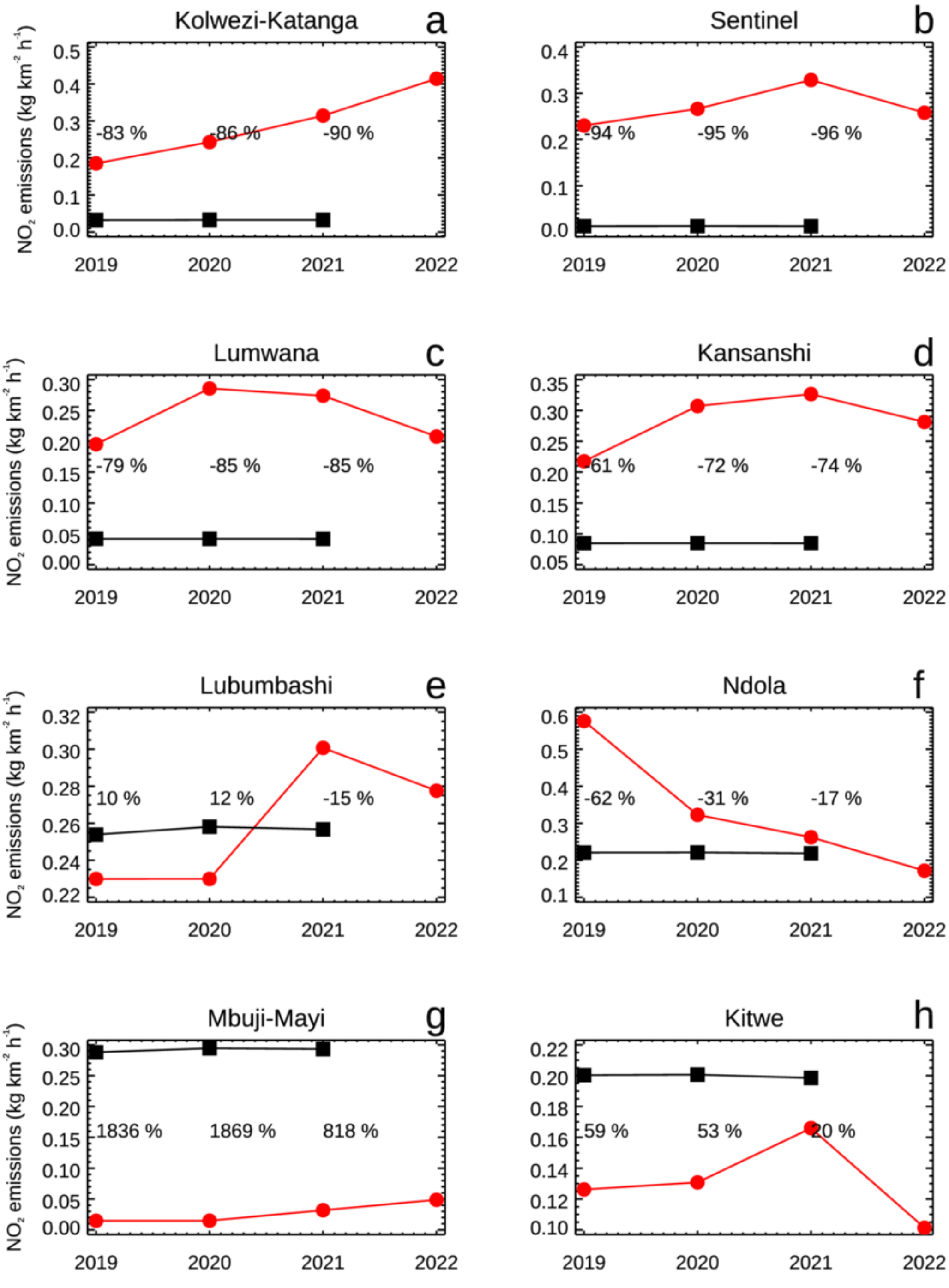

Figure S5. Annual means of NO_2 emissions from TROPOMI (red circles) and the inventory (black squares) for mines (panels a to d) and cities (panels e to h). Percent differences between the two datasets are shown in each panel. TROPOMI emissions have been background-removed. Inventory emissions include all anthropogenic sectors except for agriculture livestock, agriculture soils, and agriculture waste burning, which would be part of the background emissions. Inventory data unavailable for 2022.

Table S1. Effect of wind uncertainty (i.e., ERA5 10 member ensemble spread) on TROPOMI-derived mean NO_2 raw emissions for 2020. Emission values and their standard deviation (in parenthesis) are in kg km^2 h^{-1}.

	Wind - spread	No perturbation	Wind + spread
Kolwezi-Katanga	0.49 (0.12)	0.47 (0.12)	0.45 (0.14)
Sentinel	0.50 (0.14)	0.49 (0.12)	0.47 (0.14)
Lumwana	0.51 (0.11)	0.50 (0.08)	0.49 (0.08)
Kansanshi	0.55 (0.11)	0.53 (0.09)	0.51 (0.09)
Lubumbashi	0.51 (0.12)	0.49 (0.10)	0.47 (0.10)
Ndola	0.53 (0.12)	0.54 (0.10)	0.54 (0.12)

Table S2. TROPOMI-derived NO_2 emission data for two Copperbelt city point sources (Lubumbashi and Ndola) and two additional cities of similar population to each of them (Mbuji-Mayi and Kitwe, respectively). TROPOMI emissions (background-removed, background) as well as their standard deviation values (in parenthesis) are in kg km^2 h^{-1}. Population in million inhabitants.

		2019	2020	2021	2022
Lubumbashi	TROPOMI	0.23 (0.10)	0.23 (0.10)	0.30 (0.16)	0.28 (0.17)
	Background	0.25 (0.08)	0.26 (0.08)	0.25 (0.08)	0.26 (0.08)
	Population	2.377	2.478	2.584	2.695
Mbuji-Mayi	TROPOMI	0.01 (0.05)	0.01 (0.04)	0.03 (0.04)	0.05 (0.05)
	Background	0.28 (0.06)	0.29 (0.06)	0.30 (0.06)	0.32 (0.07)
	Population	2.413	2.525	2.643	2.765
Ndola	TROPOMI	0.58 (0.30)	0.32 (0.10)	0.26 (0.13)	0.17 (0.10)
	Background	0.20 (0.09)	0.21 (0.09)	0.21 (0.08)	0.23 (0.08)
	Population	0.531	0.542	0.556	0.571
Kitwe	TROPOMI	0.13 (0.07)	0.13 (0.07)	0.17 (0.04)	0.10 (0.06)
	Background	0.23 (0.09)	0.24 (0.08)	0.24 (0.08)	0.25 (0.08)
	Population	0.663	0.686	0.710	0.735

References

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