Supplement

S1 Land cover change analysis for dataset and study site choice

Initially both the MODIS and the ESA-CCI (ESA, 2017) land cover data were considered due to their global and long-term coverage. Both datasets record a region of high deforestation rates in the southern Amazon based on total forest cover loss between 2001 and 2019. However, a comparison with the PRODES (Programa de Monitoramento da Floresta Amazônica Brasileira por Satélite, INPE (2022)) deforestation dataset for the Legal Amazon highlights an unusual temporal distribution of deforestation in the ESA data. The ESA land cover data has a spike of deforestation in 2003, which accounts for around 78% of the deforestation in the Legal Brazilian Amazon between 2001 and 2019. This is despite the ESA data recording less deforestation (138,616 km^2) than either the PRODES (215,784 km^2) or MODIS (211,780 km^2) datasets. Consequently, the MODIS dataset was used for the analysis.

Using the MODIS dataset, rates of deforestation were calculated by fitting a linear trend to the annual forest cover values for each $1° \times 1°$ grid cell. To test the sensitivity of the trend to the choice of start and end period, this result was compared to trends fitted for 2001-2018, 2002-2019, as well as the difference between the 2015-2019 and 2001-2005 mean values. The areas of major deforestation, such as the southern Amazon, were consistent for all the above.

The change in other land cover types was studied in more detail for the southern Amazon. The land cover types were grouped into broader categories (forest or broadleaf forest, savannas, grasslands, shrubs etc.). The 21st century trends indicate the forest cover in the region is decreasing in favour of the savanna and grassland land cover categories (Fig. S1). Forest cover reduced substantially from 2001 to 2013 with a smaller decline thereafter. Over the same period, grasslands expanded from 19.3% to 22%, with a relatively stable period between 2007 and 2013, while savannas initially increased in area (from 27.4% to 30% in 2013), but decreased back to 28.6% of the study region area by 2019. The asymmetry in the savanna and grassland trends (an increase in one land cover tends to correspond to a decrease in the other) could be related to known issues of underrepresentation of agriculture in the tropics, as well as potential for some grasslands being miss-classified as savannas in the land cover data (Sulla-Menashe and Friedl,

2018). Further, recently deforested regions may first be reclassified as savannas, due to some remaining tree cover, before passing the grassland threshold, when the tree cover decreases further.

S2 Detrending HCHO data

30   To isolate regional variations in HCHO from the positive drift probably due to instrument degradation and background concentration changes, the long-term trend in the data was removed. Theil - Sen regression was used to calculate the trend in the background HCHO over a reference region in the Pacific Ocean (140° W to 100° W longitude, 30° S to 0° latitude). The calculated background trend was removed from the monthly southern Amazon HCHO (Fig. S2). The difference between the original and detrended

35   data peaks in 2018 at 1×10^{15} molecules cm^{-2}. The data for 2019 was removed from analysis and not included when calculating the background trend due to the substantial quantity of anomalously large data values.

S3 Sensitivity of the results to the choice of burned area boundary

The 0.04% grid cell area boundary was chosen to isolate grid cells strongly affected by fire emissions,

40   while ensuring sample sizes were sufficient in all categories in section 3.4.1. The sensitivity to the boundary choice was tested by comparing regional-mean isoprene and NO_2 column densities in the high and low burned area categories for boundary values 25% lower and greater than 0.04%, that is 0.03% and 0.05% (Table S1). As the boundary was decreased or increased by 25%, the mean isoprene and NO_2 column densities remained within 5% of the original values, but the high burned area sample size

45   increased by around 20% (for the 0.03% boundary) and decreased by around 15% (for the 0.05% boundary). Therefore, the mean column densities of the trace gases are not significantly influenced by the choice of burned area boundary, but the sample size in the high burned area category decreases rapidly when the boundary is increased. Therefore, the chosen threshold does not affect the results substantially, and further increasing the boundary to isolate more extreme fires would result in sample size issues.

S4 Sensitivity of the results to the size of land cover bins

To study the general trends in atmospheric composition with changes in land cover the data for each atmospheric constituent was separated based on the percentage of broadleaf forest cover (sections 3.4.2 and 3.5). The sensitivity of the result to the choice of bin size (10 percentage points for land cover) was tested by repeating the analysis for bins of sizes equal to 5, 20 and 25 percentage points. The results remained consistent with peak values for the atmospheric variables occurring at equivalent land cover percentages.
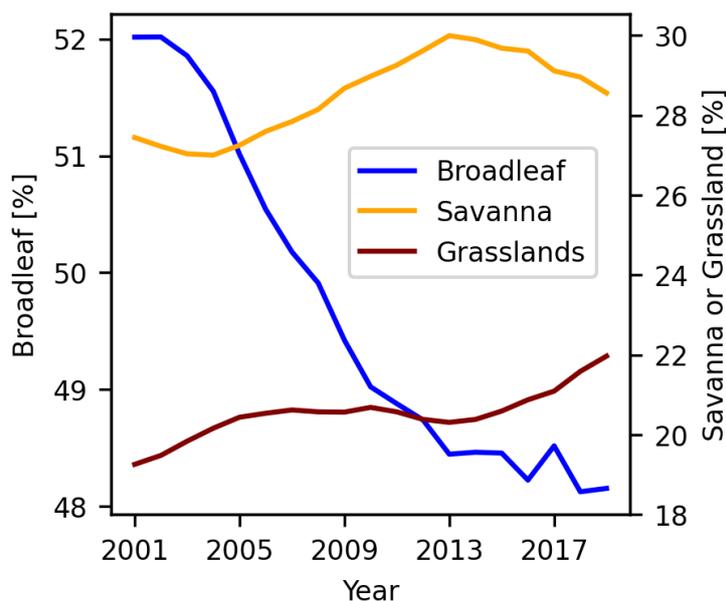
Figure S1: Change in the areas of broadleaf forests, savannas, and grasslands (as % of the study region area) from 2001 to 2019.
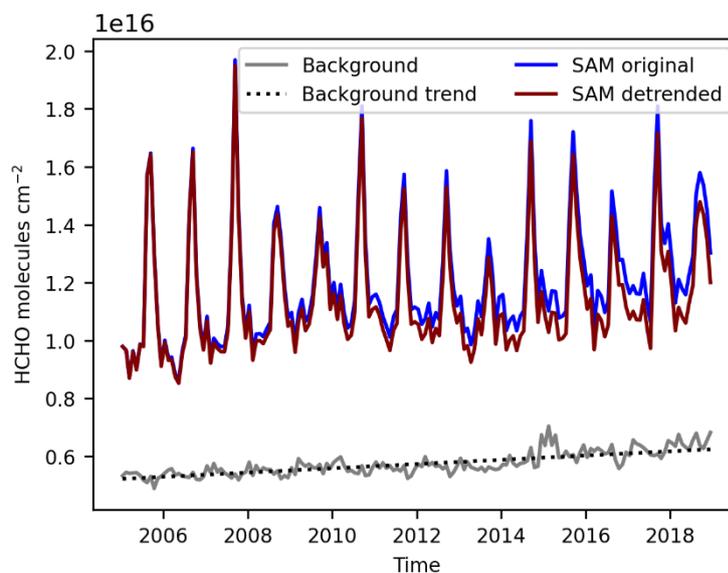
Figure S2: Background reference remote Pacific regional mean monthly HCHO (grey line) and calculated trend (grey dashed line), original monthly mean HCHO for the southern Amazon region (blue line) and detrended monthly mean HCHO for the southern Amazon region (red line).

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Table S1: Percentage change in isoprene and NO₂ column amounts and sample sizes when the burned area boundary value is changed by 25%.

	Decreased	Current	Increased
Burned area boundary	-25%	0.04%	+25%
Mean high fire isoprene	+1.44%	6.93×10^{15} mol cm^{-2}	+0.60%
Mean low fire isoprene	-0.08%	7.02×10^{15} mol cm^{-2}	+0.07%
High burned area isoprene count	+22%	837	-16%
Mean high fire NO$_2$	-4.49%	2.60×10^{15} mol cm^{-2}	+4.60%
Mean low fire NO$_2$	-1.67%	1.30×10^{15} mol cm^{-2}	+1.48%
High burned area NO$_2$ count	+19%	2462	-14%

References

ESA: Land Cover CCI Product User Guide version 2.0, maps.elie.ucl.ac.be/CCI/viewer/download/ESACCI-LC-Ph2-PUGv2_2.0.pdf, 2017.

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