

Increased estimates of air-pollution emissions from Brazilian sugar-cane ethanol

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Accelerating biofuel production has been promoted as an opportunity to enhance energy security, offset greenhouse-gas emissions and support rural economies. However, large uncertainties remain in the impacts of biofuels on air quality and climate^{1,2}. Sugar-cane ethanol is one of the most widely used biofuels, and Brazil is its largest producer³. Here we use a life-cycle approach to produce spatially and temporally explicit estimates of air-pollutant emissions over the whole life cycle of sugar-cane ethanol in Brazil. We show that even in regions where pre-harvest field burning has been eliminated on half the croplands, regional emissions of air pollutants continue to increase owing to the expansion of sugar-cane growing areas, and burning continues to be the dominant life-cycle stage for emissions. Comparison of our estimates of burning-phase emissions with satellite estimates of burning in São Paulo state suggests that sugar-cane field burning is not fully accounted for in satellite-based inventories, owing to the small spatial scale of individual fires. Accounting for this effect leads to revised regional estimates of burned area that are four times greater than some previous estimates. Our revised emissions maps thus suggest that biofuels may have larger impacts on regional climate forcing and human health than previously thought.

Air-pollutant emissions from biofuel production and combustion may have significant impacts on climate and air quality. The change in vehicle emissions that would result from a large-scale conversion from gasoline to E85 (a blend of up to 85% ethanol with gasoline or another hydrocarbon) in the United States could have significant health consequences, by increasing tropospheric ozone concentrations⁴, for example. Monetizing the health and climate impacts of US ethanol emissions (fuel production and vehicle emissions) suggests that the use of corn ethanol has higher health costs than gasoline, whereas cellulosic ethanol may reduce health costs compared with gasoline use⁵.

Although much has been learned about long-lived greenhouse-gas emissions from sugar-cane ethanol production^{6,7}, emissions of other gases and aerosols remain relatively uncertain. Brazilian sugar-cane ethanol has potentially large life-cycle air-pollution emissions because many sugar-cane croplands are burned before harvest^{8,9}. Epidemiological studies suggest that exposure to these emissions results in health impacts such as respiratory disease^{8,10,11}. The radiative forcing of the emissions may also have significant regional climate impacts¹². Although some regional governments have started to encourage farmers to gradually reduce field burning³, more than half of sugar-cane croplands continue to be burned¹³.

Air pollution, which depends on the spatial and temporal distribution of emissions, is thus a crucial issue for sugar-cane

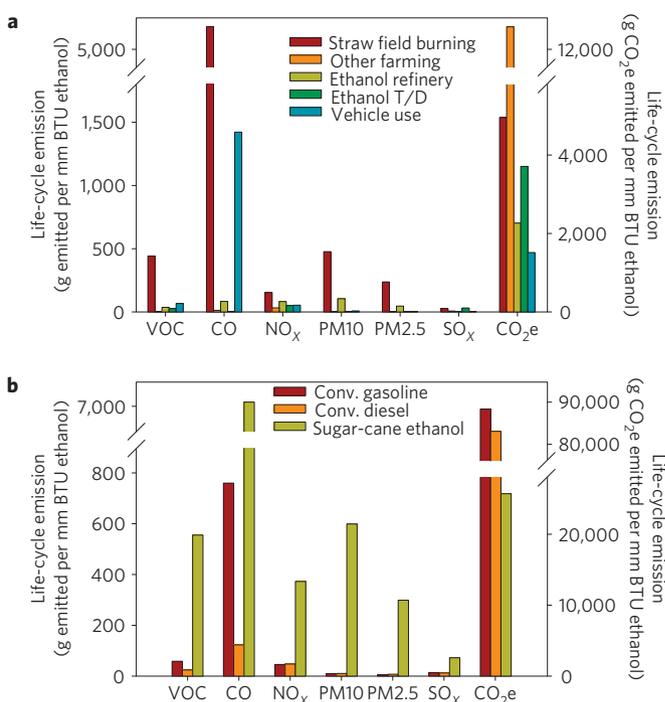


Figure 1 | Comparisons of life-cycle emissions for sugar-cane ethanol in Brazil and conventional liquid fuels. a, Life-cycle emissions per unit energy of sugar-cane ethanol produced within five life-cycle phases. Although our life-cycle emissions account for a mix of sugar-cane fields where the burning practice is used and not used, the burning-phase emissions shown here are for ethanol produced from croplands that are burned. T/D, transportation/distribution; BTU, British thermal units. **b**, Comparisons of life-cycle emissions for conventional gasoline, diesel and sugar-cane ethanol. Estimates from the GREET model include six air pollutants (VOC, CO, NO_x, PM₁₀, PM_{2.5}, and SO_x) and greenhouse gases (as CO₂ equivalent, CO₂e). Right axis is for greenhouse-gas emissions.

ethanol sustainability. In this paper, an emissions inventory for the life cycle of sugar-cane ethanol produced and used in Brazil is estimated using agriculture survey data, emissions factors and life-cycle assessment. Emissions factors from the Greenhouse Gases, Regulated Emissions, and Energy Use in Transportation (GREET) model for the farming, field burning, refinery, transportation and distribution, and vehicle-use phases are combined with temporally and spatially explicit activity rates (Methods). The results of the open-burning phase are found to be of particular importance and are compared with estimates obtained using alternative,

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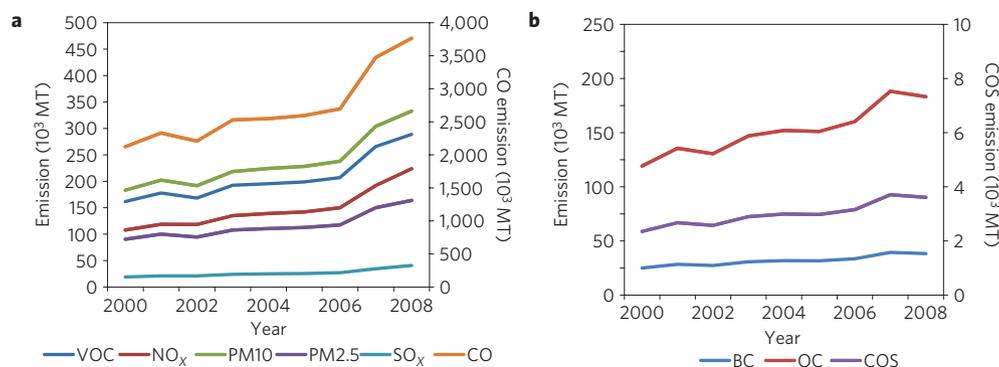


Figure 2 | Estimated life-cycle emissions of ethanol in Brazil from crop year 2000 to 2008 (crop year is from April to January the following year). **a**, Emissions of VOC, NO_x, PM₁₀, PM_{2.5}, SO_x and CO. **b**, Emissions of black carbon (BC), organic carbon (OC) and carbonyl sulphide (COS), which are estimated only for the field-burning phase.

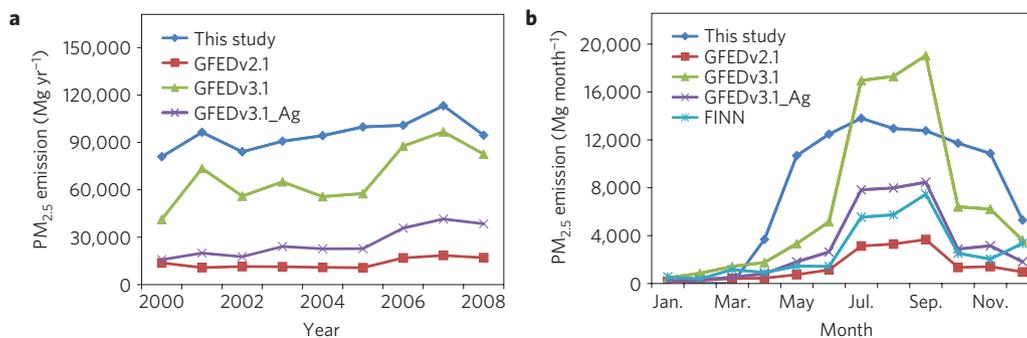


Figure 3 | Temporal variations of PM_{2.5} emission from biomass burning. **a**, Comparison of annual (a) and monthly (in 2008) (b) emissions from GFEDv2.1, GFEDv3.1, GFEDv3.1_Ag, FINN, and our approach.

remote-sensing approaches. The new emissions data are available online (<http://faculty.ucmerced.edu/ecampbell3/>).

The estimated life-cycle emissions per unit energy of sugar-cane ethanol produced are shown in Fig. 1, together with life-cycle emissions from conventional gasoline and diesel. The field-burning phase is the dominant source of emissions for all species except SO_x. The occurrence of burning is thought to have been reduced to half of the sugar-cane cropland area in São Paulo state³, resulting in NO_x and SO_x emissions from field burning that are the same order of magnitude as the emissions from the other stages of the life cycle, but the field-burning phase remains dominant for the other emitted species. In contrast to air-pollution emissions, there has been extensive work on estimating life-cycle CO₂ emissions from Brazilian ethanol and the GREET results for CO₂ emissions are shown in Fig. 1 for comparison.

The life-cycle emissions of ethanol in Brazil for crop years 2000 to 2008 are shown in Fig. 2. Despite reductions in the fraction of sugar-cane croplands burned, the total emissions increase over time because of increased production. More than 40% of the total emissions from biofuels in Brazil arise from biofuels produced in São Paulo state (Supplementary Table S1). Emissions estimates for each life-cycle phase for São Paulo state are shown in Supplementary Table S2. The spatial and seasonal variations of these emissions are shown in Supplementary Figs S1 and S2. Concentrated emissions appear near the central region and northern border of São Paulo state owing to the high utilization of land for sugar-cane crops in these areas. Seasonal variation in emissions from burning is associated with sugar-cane cultivation and harvest, with the peak occurring in July (Fig. 3b).

We compare our life-cycle-based estimates of emissions from sugar-cane field burning with emissions estimates from the Global Fire Emissions Database (GFED) and Fire INventory

from NCAR (FINN) satellite fire-detection approaches^{14,15}. This comparison is done for São Paulo state because the GFED and FINN approaches integrate burning from all land-cover sources, which requires that we focus on a region where sugar-cane field burning is the dominant source of biomass burning. The emissions of particulate matter less than 2.5 μm (PM_{2.5}) for GFEDv2.1, GFEDv3.1, GFEDv3.1_Ag (calculated from the burning fraction of agricultural land only), FINN, and our life-cycle approach are shown in Fig. 3 (see also Supplementary Figs S3 and S4). The satellite-based approaches might be expected to lead to larger emissions because they include all sources of biomass burning (for example forest, cropland, savannah, pastures and so on) in addition to sugar-cane burning. However, our burning emissions are on average 7.3 times greater than the GFEDv2.1 emissions, 1.5 times greater than the GFEDv3.1 total emissions, 3.6 times greater than the GFEDv3.1_Ag emissions, and 3.1 times greater than the FINN emissions.

The low emissions for the satellite-based approaches are largely due to low estimates of burned area (Table 1, Fig. 4). One limitation of the GFEDv2.1 approach is that the burned area was estimated indirectly by a regression tree relationship between fire hotspots and burned area¹⁵. The larger areas in GFEDv3.1 relative to GFEDv2.1 represent an improvement wherein 90% of the areas were mapped directly from 500 m Moderate Resolution Imaging Spectroradiometer (MODIS) burned area data rather than the regression tree relationship. The GFEDv3.1 burned areas are still only 33% of our life-cycle estimates in 2008. Although the subset of GFEDv3.1 burned areas that are on agriculture lands are not reported, these areas would be even smaller. The satellite-based underestimate of burned areas in the region may be due to the small size of the individual sugar-cane fires relative to the 500 m resolution for MODIS burned areas¹⁶.

Table 1 | Comparison of PM_{2.5} emissions, burned areas, fuel loading and emission factors in São Paulo state for the year 2008 among GFEDv2.1, GFEDv3.1, GFEDv3.1_Ag, FINN and our approach.

Item	This study	GFEDv2.1	GFEDv3.1	GFEDv3.1_Ag	FINN
Emission (Mg yr ⁻¹)	100,857	16,894	87,703	38,595	32,457
Burned area (ha)	2,222,638	341,682	746,316	NA	NA
Fuel load (Mg dm yr ⁻¹)	25,859,274	3,428,936	12,697,708	4,678,169	NA
Avg. EF (g kg ⁻¹ dm ⁻¹)*	3.9	4.9	6.9	8.3	NA
Avg. fuel load (Mg dm ha ⁻¹ yr ⁻¹)†	11.0	10.0	17.0	NA	NA

*The average emission factor is calculated by dividing emission by fuel load. †The average fuel load is calculated by dividing fuel load by burned area. dm stands for dried matter.

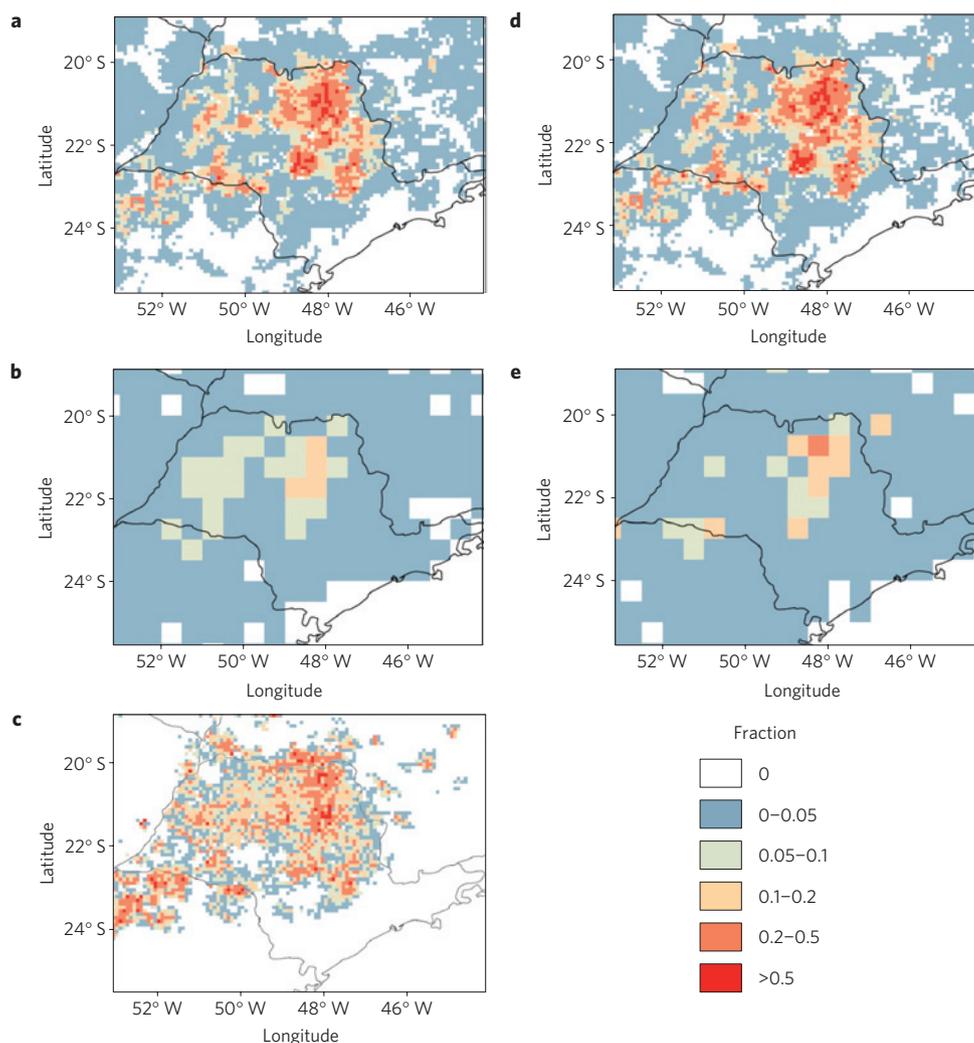


Figure 4 | Fraction of each grid cell burned by three approaches. a-c. Results for year 2008 using the approach in this study (resolution: 5 arcmin × 5 arcmin) (a), GFEDv3.1 (resolution: 30 arcmin × 30 arcmin) (b) and Landsat-based Canasat fire data (re-gridded to 5 arcmin × 5 arcmin) (c). **d-e.** Results for year 2006 using the approach in this study (resolution: 5 arcmin × 5 arcmin) (d) and GFEDv3.1 (resolution: 30 arcmin × 30 arcmin) (e).

High-resolution Landsat data are often used to validate MODIS-based products. Recent work has used Landsat data to produce estimates of burned areas for Brazilian sugar-cane croplands¹³. Our approach results in estimates of burned area that are within 13% of the Landsat-based estimate for São Paulo state. Comparison of the gridded (Fig. 4) and municipality-level (Supplementary Table S3) burned areas also provide similar estimates for our results and the Landsat-based approach. Although our approach and Landsat data yield similar burned areas, the MODIS burned areas markedly underestimate these burned areas.

Although the MODIS-based emissions estimates are driven by relatively small burned areas, these estimates are driven by average fuel loading and average emissions factors that are relatively high compared with our life-cycle approach (Table 1). The GFED fuel loadings are probably higher because the GFEDv3.1 land-cover map indicates that only 38% of the burned area in São Paulo state is on agricultural land, with the rest of the burning occurring on other lands, such as forests, that have higher fuel loadings¹⁷. The emission factors assumed in the MODIS-based approaches for agricultural biomass burning are also higher than the GREET

emission factors we applied for PM_{2.5} and are similar or larger for other species (Table 1, Supplementary Table S4). Thus, the emissions factors we applied are at the low end of the range, making our emissions approach conservative with respect to the MODIS-based estimates. Had we used the larger emissions factors assumed in the MODIS-based approaches, the gap between our emissions estimates and the GFED and FINN estimates would have been even larger.

Some similarities are observed between the temporal and spatial variability of the satellite and life-cycle approaches, suggesting a relationship of drivers between the sugar-cane burning and the large-scale burning from the satellite-based approaches. The interannual variability of the GFEDv3.1 and our approach is somewhat consistent, as suggested by an r-squared value of 0.67 between the annual PM_{2.5} emissions for the GFEDv3.1 and our emissions. This correlation suggests that crop area, which is the primary driver of the variability in our field-burning emissions estimates, is also a possible driver of the MODIS-based estimates. All emissions estimates in Fig. 3a show positive trends over the 2000–2008 period, with regression slopes that are 3, 6, and 7% growth for our approach, GFEDv2.1, and GFEDv3.1, respectively. The seasonal variation for all methods has emissions concentrated during the harvesting season, although our emissions have a longer burning season that extends from May to November (Fig. 3b). These peak emissions during the May to November harvest season are consistent with analysis of Advance Very High Resolution Radiometer (AVHRR) fire spot detection for the region and field air-quality measurements¹⁸. The spatial variability is shown in Fig. 4 for burned areas and in Supplementary Fig. S3 for PM_{2.5} emissions. The burned area maps all show widespread burning in the north-central region where sugar cane is concentrated.

Improvements in the GFED and FINN methods may be achieved by integrating information from the inventory-based approach applied here or through the use of higher-resolution satellite data where available¹⁶. Because the GFED and FINN data may not account for sugar-cane burning, the total burning emissions for the region may be the sum of our estimates and the satellite-based emissions. This would increase total PM_{2.5} emissions, relative to only using the satellite-based estimates, by a factor of 2.1 and 4.1 for the GFEDv3.1 and FINN data, respectively. The burned area for the sum of our estimates and the GFEDv3.1 estimates would be four times greater than the GFEDv3.1 burned area alone.

It is not possible to validate emissions from the sugar-cane life cycle because of a lack of direct measurements. However, we have explored the uncertainties in our emissions inventory, particularly with respect to the parameters associated with the dominant open-burning phase, including fuel loadings, burned area and emission factors. Our fuel loadings for the open-burning phase are based on a fixed ratio of burned biomass to yields that has been applied in previous work^{3,19}. Agronomic assessment is needed to provide uncertainty estimates for this ratio.

The estimates of burned area are driven by agronomic data for yield and production as well as remote sensing. The scale of our estimates (5 arcmin × 5 arcmin, ~9.3 km) differs markedly from the MODIS-based approaches (500 m–1 km) as well as the validation Landsat data (30 m). The coarse resolution of MODIS-based approaches may have resulted in detection problems of sugar-cane burning. However, even without these detection problems, the coarse scale could result in challenges in applying these emissions to high-resolution atmospheric models that are used to simulate atmospheric composition and climate forcing.

A range of emission factors for open burning have been reported in the literature, introducing significant uncertainty into the modelling approach. The standard deviations for the emissions factors vary widely on the basis of the emission factors applied in a range of studies (Supplementary Table S3; refs 15,17,20,21).

The uncertainty is particularly large for PM_{2.5} and CO, at 32% and 46%, respectively. Incorporating crop-specific emissions factors as opposed to those used in GREET and MODIS-based approaches could lead to improved estimates. Top-down studies of atmospheric composition could provide one approach to validating these emission factors.

The rapid expansion of sugar-cane ethanol and changing production methods result in air-pollution emissions that are changing in space and time. The impacts of air pollution on health and regional climate forcing are perhaps some of the most uncertain impacts of sugar-cane ethanol, but are critical for assessing its sustainability. Further work coupling the spatially and temporally resolved maps from this work with satellite-based methods and atmospheric chemical transport models is needed to better understand these impacts on climate and health.

Methods

Spatially and temporally explicit emissions from Brazilian sugar-cane ethanol were estimated by integrating emission factors and activity rates associated with emissions in different life-cycle phases. The emissions, $E_{i,s,m}$, of pollutant i (that is, volatile organic compounds (VOCs), nitrogen oxides (NO_x), particulate matter less than 10 and 2.5 μm (PM₁₀; PM_{2.5}), sulphur oxides (SO_x), and carbon monoxide (CO)) at location s during the month m is determined by aggregating the emissions from p phases, where p represents farming ($p=f$), field burning ($p=b$), refinery ($p=r$) and transportation/distribution (T/D) ($p=t$) as follows:

$$E_{i,s,m} = \sum_p E_p = \sum_p (A_{s,p} \times \text{MAF}_{m,p} \times \text{AR} \times \text{EF}_{i,p}) \quad \forall p=f, b, r, t \quad (1)$$

where $A_{s,p}$ is the activity rate associated with emissions at location s in phase p , including sugar-cane production and ethanol production. $\text{MAF}_{m,p}$ represents the monthly allocation factor, which converts annual emissions to a monthly basis. It is derived from the monthly intensity of emission activities for each phase, that is, the monthly ratio of cultivation, harvest, ethanol production, distributed ethanol, or ethanol consumption (Supplementary Methods). AR is the fraction of sugar-cane production used for producing ethanol, as not all sugar-cane crops in Brazil contribute to ethanol production (for example, around 60% of sugar-cane crops were converted into ethanol products in 2008)²². $\text{EF}_{i,p}$ is the emission factor of pollutant i in phase p .

As activity rates in the above four phases are associated with spatially explicit sugar-cane production, the estimation of life-cycle emissions can be further expressed by the following formula:

$$E_{i,s,m} = \sum_p [(Y_s \times \text{CA}_s \times \text{CF}_p) \times \text{MAF}_{m,p} \times \text{AR} \times \text{EF}_{i,p}] \quad \forall p=f, b, r, t \quad (2)$$

where Y_s represents the spatial dataset of annual sugar-cane yield (Mg/ha yr), CA_s denotes the sugar-cane crop area (ha) at location s , and CF_p denotes conversion factors (24 gal ethanol per Mg sugar cane). The sugar-cane crop area, CA_s , is determined by the fraction of cropland in a grid cell (ha cropland per ha grid cell) and the area of a grid cell. Monfreda *et al.*²³ produced datasets of global cropland in 2000, including yield (Mg sugar cane per ha cropland per year) and the fraction of cropland (ha cropland per ha grid cell), by spatially disaggregating agricultural inventory data at the political unit level and redistributing to grid cells in relation to remotely sensed land cover (resolution 5 arcmin × 5 arcmin). Given the expansion of sugar-cane production from 2000 to 2008, we scale the production data for the year 2000 to the more recent production year, based on state-level data. Sugar-cane production in São Paulo state increased by 81% between 2000 to 2008 because of an increase in area and yield²². We evaluate this assumption by comparing our map of sugar-cane production with a map of sugar-cane production based on 2008 Landsat data¹⁵. At the time of this study the year 2008 was the most recent year for which GFED and FINN data were available.

Although the focus of this study is on production-phase emissions, we also develop a simple vehicle-phase emission estimate. It is assumed that the spatial distribution of ethanol consumption for vehicles follows the distribution of population in Brazil, and ethanol is evenly consumed over the twelve months in a year. Although distributing vehicle emissions by population is a rough first approximation, this approach has been found to be useful in previous air quality studies of the region²⁴. The vehicle emission factors are taken from GREET and the spatial distribution of population is obtained from LandScan, a global population database with a 30 arcsec × 30 arcsec resolution that is available for 2008 (ref. 25).

We compare the field burning emissions estimated from our approach with global inventories of biomass burning emissions. The Global Fire Emissions Database version (GFEDv2.1) provides open biomass burning emissions by combining 500 m MODIS burned areas, the Carnegie–Ames–Stanford Approach (CASA) biogeochemical model, and emissions factors²⁶. The 1° × 1°-gridded, 8-day timestep fire emissions are available from 1999 to 2008 (GFEDv2.1)^{26,27} and

at a monthly time step in a more recent release (GFEDv3.1)¹⁵. Wiedinmyer *et al.* applied a related approach of integrating fuel loadings and MODIS fire observations to estimate fire emission in the FINN (refs 14,21). FINN uses 1-km daily fire data identified by the MODIS Thermal Anomalies product. Except burned area data, another important difference between those two studies is the data sources used for fuel loadings and combustion completeness. The GFED approach estimates fuel loadings as a function of the net primary production modelled by the CASA biogeochemical model and the value of combustion completeness is determined by fuel types. Wiedinmyer *et al.* assign the value of fuel loading for each grid cell by combining the Global Land Cover Dataset for 2000 (GLC2000), the MODIS Vegetation Continuous Field (VCF), and the Fuel Characteristic Classification System (FCCS).

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Author contributions

C.-C.T. and J.E.C. developed the emissions model. All authors contributed to the analysis of results and writing of the manuscript.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/natureclimatechange. Reprints and permissions information is available online at <http://www.nature.com/reprints>. Correspondence and requests for materials should be addressed to J.E.C.