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Air Quality Impact of Diffuse and Inefficient Combustion Emissions in Africa (DICE-Africa)

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Supporting Information

ABSTRACT: Anthropogenic pollution in Africa is dominated by diffuse and inefficient combustion sources, as electricity access is low and motorcycles and outdated cars proliferate. These sources are missing, out-of-date, or misrepresented in state-of-the-science emission inventories. We address these deficiencies with a detailed inventory of Diffuse and Inefficient Combustion Emissions in Africa (DICE-Africa) for 2006 and 2013. Fuelwood for energy is the largest emission source in DICE-Africa, but grows from 2006 to 2013 at a slower rate than charcoal production and use, and gasoline and diesel for motorcycles, cars, and generators. Only kerosene use and gas flaring decline. Increase in emissions from 2006 to 2013 in this work is consistent with trends in satellite observations of formaldehyde and NO_2, but much slower than the explosive growth projected with a fuel consumption model. Seasonal biomass burning is considered a large pollution source in Africa, but we estimate comparable emissions of black carbon and higher emissions of nonmethane volatile organic compounds from DICE-Africa. Nitrogen oxide ($NO_x \equiv NO + NO_2$) emissions are much lower than from biomass burning. We use GEOS-Chem to estimate that the largest contribution of DICE-Africa to annual mean surface fine particulate matter ($PM_{2.5}$) is >5 $\mu g\ m^{-3}$ in populous Nigeria.

INTRODUCTION

Air pollution in Africa is dominated by seasonal open field burning (biomass burning);[1] however, anthropogenic activity may surpass biomass burning as the largest pollution source by 2030.[2] Africa is experiencing rapid economic and population growth that will inevitably degrade air quality. Sustained economic growth requires access to reliable and affordable electricity. Currently the majority of Africans depend on inefficient sources of combustion for their energy and transport needs.[3] Inefficient combustion results in direct emission of fine particles ($PM_{2.5}$, aerosols with diameter less than 2.5 μm) like black carbon (BC) and organic carbon (OC), and formation of secondary organic aerosol (SOA) and ozone from photo-oxidation of nonmethane volatile organic compounds (NMVOCs). $PM_{2.5}$ and ozone are hazardous to human health. Here we develop a detailed inventory of anthropogenic Diffuse and Inefficient Combustion Emissions in Africa (DICE-Africa) to determine the impact of these sources on ambient air quality.

The population in Africa has doubled since the mid 1980s,[4] but average electricity access has only increased by 12%.[5] Currently access to reliable electricity is less than 50% in most African countries. In some countries (Chad, Burundi, Malawi, and Liberia) access is below 10%.[5] Most people rely on crop residue, fuelwood, charcoal, kerosene, and backup generators for energy. The mix of fuel varies for urban and rural populations, determined largely by access to fuel and affluence. Rural households typically use fuelwood and crop residue, whereas urban households and businesses use a mix of fuelwood, charcoal, and backup generators.[6,7] Kerosene is used in urban and rural homes for cooking and lighting.[8,9] Rapid urbanization and economic growth are contributing to expansion in backup generator use and charcoal production.[10,11] Charcoal is formed by slow-burning wood under anoxic conditions and occurs predominantly in rural areas close to forests.[12]

Proliferation of motorcycles, a vehicle fleet dominated by outdated cars, poor road management, and low-quality fuel exacerbate pollution in dense urban centers.[13,14] The problem will worsen with increased demand for cars and motorcycles.[15] Regular fuel shortages in Nigeria, where one in five Africans reside, has energized an informal industry of ad hoc oil refining in the Niger Delta. As much as 37 500 barrels of crude oil is refined daily,[16] nearly 2% of crude oil production in Nigeria.[17]

A standard practice at conventional oil fields in Africa, in particular Nigeria, is to flare natural gas, as oil fields were established when natural gas was not marketable.[17] Flaring is a major source of pollution, since flares in Africa operate at low

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combustion efficiency (<70%).[18] In Nigeria as much as 15 billion m^3 (BCM) natural gas was flared in 2008,[19] equivalent to 2.4% of end-user consumption of natural gas in the US for the same year.[20]

Emission inventories of pollutants in Africa are highly uncertain and likely underestimated, in particular in urban centers.[21] Global inventories such as the Emissions Database for Global Atmospheric Research (EDGAR),[22] Reanalysis of the Tropospheric Chemical Composition (RETRO),[23] Emissions for Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP),[24] and Bond et al.[25,26] are typically used in chemical transport models (CTMs) to simulate air quality in Africa. Important sources in Africa are missing from these inventories, namely motorcycles, kerosene use, open waste burning, and ad hoc oil refining. Pollution from cars and flaring of natural gas are included, but the emission factors to convert fuel burned to pollution released are likely not representative of the inefficient combustion conditions in Africa. The Yevich and Logan[27] global inventory of solid biofuel use is for 1986, but is still used to represent present-day emissions.[28] Liousse et al.[2] developed a regional inventory of anthropogenic emissions in Africa, but many emission factors in that inventory are representative of efficient combustion conditions typical of developed countries.

Here we construct an updated inventory of pollution from diffuse and inefficient combustion sources in Africa at high spatial resolution ($0.1° \times 0.1°$) to distinguish rural and urban pollution sources. We construct our inventory for 2006 and 2013. Over this period the African population grew at a rate of 2.7% a^{-1}; faster than China (0.44% a^{-1}) and India (1.2% a^{-1}) for the same time period. We evaluate the trend in emissions associated with rapid population growth and compare this to trends reported for other regional and global emission inventories and from satellite observations of formaldehyde (a high-yield oxidation product of nonmethane volatile organic compounds, NMVOCs) and nitrogen dioxide (NO_2). We incorporate our emission inventory in the GEOS-Chem CTM to assess the impact of DICE-Africa on ambient air quality.

■ METHODS

DICE-Africa includes emissions from the following sources of pollution: solid biofuels (wood, crop residue, charcoal), charcoal production, kerosene, backup generators, cars, motorcycles, gas flaring, and ad hoc oil refining. Calculation of pollution from these sources in Africa follows a standard approach: emissions E are calculated as the product of the quantity of fuel i burned in each country—the activity factor (AF)—and the mass of each pollutant j emitted per unit of fuel i burned—the emission factor (EF):

$$E_{i,j} = \mathrm{CE} \times \mathrm{AF}_i \times \mathrm{EF}_{i,j} \quad (1)$$

CE is the conversion efficiency of fuel carbon to CO_2 and depends on the relative amount of flaming (CE = 0.99) and smoldering (CE = 0.80) combustion.[29] Crop residue burning is predominantly flaming (CE = 0.95), charcoal and fuelwood is a mix of smoldering and flaming (CE = 0.90), and charcoal is produced by smoldering wood (CE = 0.80).[29−31] We apply an additional scaling factor to kerosene to account for ventilation of 89% of emissions to the outdoors.[32] CE = 1 for natural gas flaring and for gasoline and diesel for cars, motorcycles, and backup generators. Where available we use emission factors representative of inefficient combustion (Supporting Information Table S6). The approach used to estimate emissions $E_{i,j}$ is summarized below. Additional details are in the Supporting Information.

Activity factors for individual countries (Tables S2−S3) are from the UN energy statistics database for fuelwood use, charcoal use, kerosene use, charcoal production, and gasoline and diesel use for transport.[33] Crop residue used for energy in each country is determined with agro-climate data[34] and reported amounts of crop residue reserved for fuel in each agro-climatic zone.[27] We use a 20% wood-to-charcoal conversion efficiency to estimate the mass of wood used to generate charcoal.[27]

Diesel and gasoline for backup generators is either not available in the case of gasoline or only available for five countries in the case of diesel.[33] Instead we calculate electricity generated with backup generators for each country (Table S4) using reports of the contribution of generators to installed capacity,[6,10] total installed capacity in each country,[35] and an energy conversion efficiency of 25%.[36] The amount of gasoline and diesel consumed by motorcycles and cars follows the approach of Assamoi and Liousse;[14] that is, the amount of fuel consumed by motorcycles is estimated with car ownership data, reported motorcycle-to-car ratios (Table S5), and 0.5 L day^{-1} fuel consumption. Fuel used by cars is then the difference between total transport fuel reported by the UN and our estimate of fuel used for motorcycles.

The volume of natural gas flared (in BCM) in individual countries is derived using satellite observations of gas flare hotspots.[19,37] Pollution from ad hoc oil refining includes volatilization of hydrocarbons and burning of crude oil to sustain fires during distillation. In the absence of data on the amount of crude oil used to sustain wood fires, we assume 2.5% (940 bpd) of acquired crude oil is burned.

Country-level $E_{i,j}$ values are gridded to the LandScan 2006[38] (Figure S1), and 2013[39] population data, except for natural gas flares and ad hoc oil refining. Flaring emissions are gridded to gas flare hotspots obtained with the Defense Meteorological Satellite Program nighttime lights for 2006 and 2013,[19] and pollution from ad hoc oil refining is uniformly distributed across land area of the Niger Delta. Fuelwood, kerosene, car, and motorcycle emissions are assigned uniformly to total population; backup generator and charcoal use to urban centers; and charcoal production and crop residue use to rural populations. We distinguish rural regions from urban centers (Figure S1) with a population density threshold of 1000 people km^{-2}.

We incorporate DICE-Africa in the GEOS-Chem CTM (version 10−01; http://acmg.seas.harvard.edu/geos/) using the HEMCO emissions package[28] and from there determine the impact of year 2006 DICE-Africa emissions on air quality in Africa and on global tropospheric ozone. GEOS-Chem is driven by GEOS-5 assimilated meteorology from the NASA Global Modeling and Assimilation Office and includes detailed oxidants and aerosol chemistry as described for example by Mao et al.[40,41] Emissions from DICE-Africa are regridded to the global model horizontal resolution (2° latitude × 2.5° longitude). Pollution sources other than those in DICE-Africa include on-grid electricity and formal industry (excluding gas flares) from EDGAR v4.2 for NO_x ($NO_x \equiv NO + NO_2$), SO_2, and CO,[22] RETRO v2.0 for NMVOCs,[23] and Bond et al.[26] for OC and BC. Emissions from open trash burning, another prevalent inefficient combustion source in Africa, are from Wiedinmyer et al.[42] Other emissions include biomass burning (open field fires) from GFED3,[43] biogenic emissions from

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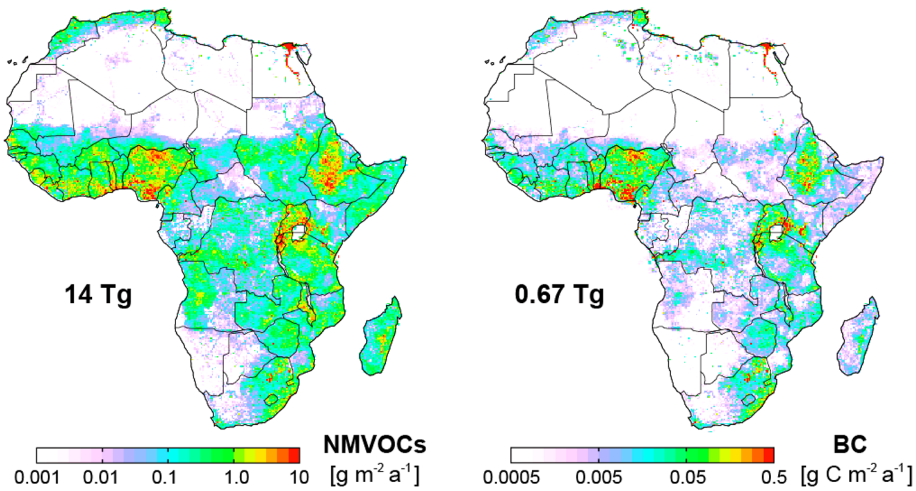

Figure 1. Spatial distribution of surface sources of nonmethane volatile organic compounds (NMVOCs) (left) and black carbon (BC) (right) for 2006. Maps show emissions on a 0.1° × 0.1° grid. Total emissions are given. Surface sources of pollution include solid biofuel use (wood, crop residue, charcoal), charcoal production, kerosene, backup generators, cars, motorcycles, gas flaring, and ad hoc oil refining.

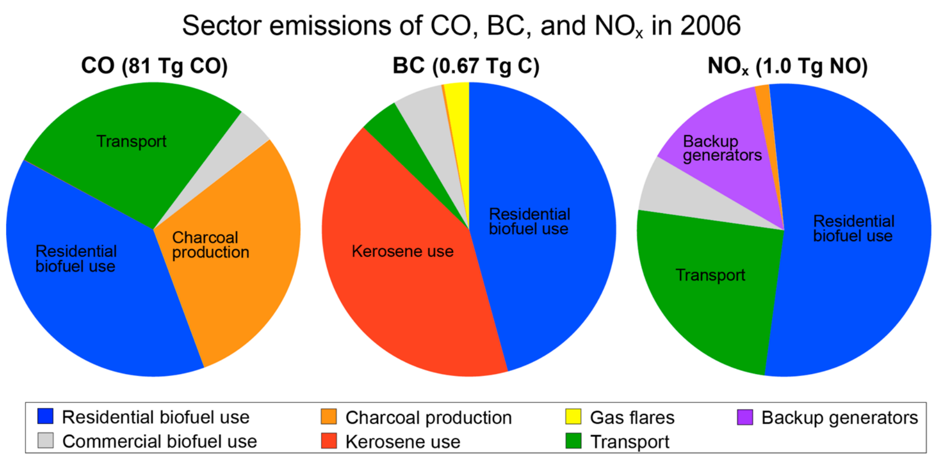

Figure 2. Sources of CO, BC, and NO_x by sector in 2006. Labels are shown for sectors that make at least 10% contribution to total emissions. Residential biofuel use includes crop residue, fuelwood, and charcoal, transport includes gasoline (petrol) and diesel consumption by cars and motorcycles, commercial biofuel is fuelwood use only.

MEGAN2.1,[44] and soil NO_x as described in Hudman et al.[45] Model results are for year 2006 following one year of spinup for chemical initialization.

RESULTS AND DISCUSSION

Figure 1 shows the distribution of NMVOCs (14 Tg) and BC (0.67 Tg C) emissions from DICE-Africa for 2006, which follow closely the spatial distribution of population (Figure S1). Emissions are high in dense rural and urban areas along the Nile River, throughout northern Ethiopia, in northern Nigeria, the Niger Delta, and surrounding Lake Victoria in Kenya, Uganda, Rwanda, and Burundi. BC emissions in the Sahara Desert and the Gulf of Guinea are from gas flaring. Liousse et al.[2] forecast that anthropogenic emissions will surpass biomass burning pollution by 2030, but total NMVOC emissions in Figure 1 already exceed estimates from biomass burning for the same year: 5.9 Tg and 6.2 Tg from the FINN[46] and GFED3[43] inventories, respectively. Our estimate of BC emissions is similar to the range from biomass burning: 1.1 (FINN) and 0.67 (GFED3) Tg C.

Figure 2 shows the contribution of sectors to emissions of CO, BC, and NO_x in 2006. Sector emissions of other species (NMVOCs, OC, SO_2, and NH_3) are in the Supporting Information (Figure S2). Residential biofuel, dominated by fuelwood use, makes a substantial contribution to all pollutants in 2006. Growth in fuelwood use from 2006 to 2013 (1.1% a^{-1}) is slower than growth in charcoal use (3.5% a^{-1}), charcoal production (3.8% a^{-1}), and transport fuel (3.8% a^{-1}). As a result the contribution of fuelwood to CO emissions is declining (31% in 2006 and 28% in 2013). Backup generators only contribute significantly to NO_x emissions (13%). Yearly data on backup generator use in Africa is lacking, so backup generator use scales with growth in installed capacity that increases by 1.9% a^{-1}. This is likely a conservative estimate, as backup generators are used in the absence of on-grid electricity and should increase with economic growth, due to increased affluence and an expanding business sector.[10] Kerosene use is a large source of BC (41% of emissions in 2006). Its contribution to NMVOCs and NO_x is unknown, as there are no emission factors reported for these species. The transport sector emits 27% of CO and 25% of NO_x, almost all (>90%) from cars. In general, cars are a larger source of trace gases than motorcycles, whereas cars and motorcycles make near-equivalent contributions to BC and OC.

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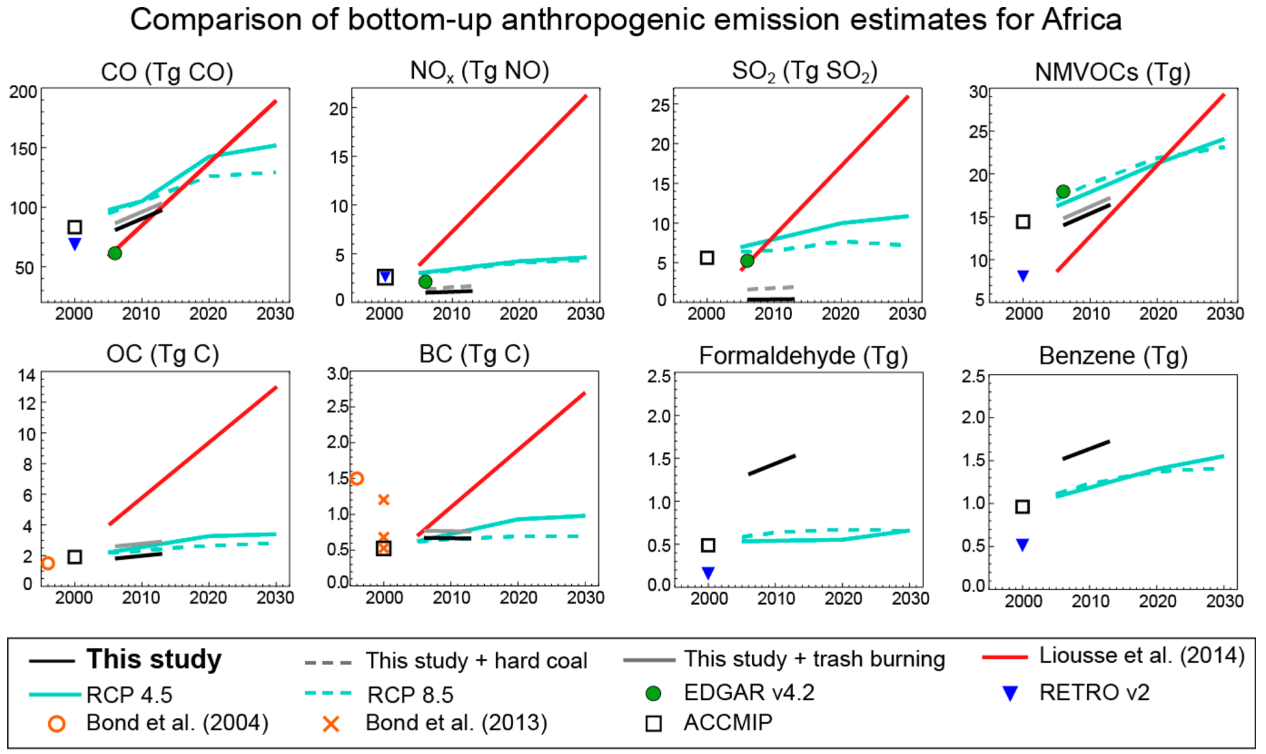

Figure 3. Comparison of emission inventory estimates and trends for Africa. Results from this study (solid black line) are compared to emission totals and trends from the regional Liousse et al. inventory, and global inventories: EDGAR v4.2, RETRO v2.0, ACCMIP, Bond et al. inventories, and the RCP 4.5 and 8.5 scenarios. Liousse et al. values for 2030 are their business-as-usual "REF" scenario. Gray lines show emissions from this study with hard coal emissions of NO_x and SO_2 from EDGAR v4.2 (dashed gray lines) and trash burning emissions of CO, NMVOCs, OC, and BC from Wiedinmyer et al. (solid gray lines).

Figure 3 compares emission totals and trends from this study with values from global and regional inventories. BC emissions in this work for 2006 are consistent with values from the Liousse et al.[2] regional inventory (henceforth referred to as "Liousse et al.") for 2005, but we obtain lower NO_x and higher NMVOC and CO emissions than the Liousse et al. inventory. They estimate NO_x, NMVOC, and CO emission factors for semideveloped and developing nations by scaling developed country NO_x, CO, and NMVOC emission factors each by the same amount, so that NMVOC-to-NO_x and CO-to-NO_x emission ratios are the same for different development categories. Reliance on inefficient combustion sources for energy in semideveloped and developing nations should lead to an increase in NMVOC-to-NO_x and CO-to-NO_x emission ratios. SO_2 emissions in this work are more than an order of magnitude lower than Liousse et al. SO_2 emission factors for diesel and gasoline for cars and motorcycles in this work (Table S6) are much lower than emission factors in Liousse et al., as we assume that the majority of diesel and gasoline is refined from low-sulfur crude oil (2.5 wt % sulfur) that is typical of African oil fields.[47] Liousse et al. include pollution from hard coal in their inventory, but adding EDGAR v4.2 hard coal SO_2 emissions to SO_2 emissions estimated in this work does not resolve the discrepancy (Figure 3; gray dashed line).

Total NMVOC emissions from the global inventories EDGAR v4.2 in 2006, ACCMIP in 2000,[24] and the Representative Concentration Pathways (RCP) 4.5[48] and 8.5[49] in 2005 are similar to this study, but with very different species contributions. Formaldehyde and benzene, for example, are higher in this work than the global inventories by at least a factor of 2.3 for formaldehyde and 1.4 for benzene. We also estimate higher emissions of the other aromatics toluene and xylenes (not shown). Aromatics (benzene, toluene, and xylenes) are SOA precursors, so their contribution will substantially alter secondary sources of $PM_{2.5}$.[50] Ozone production is also sensitive to the mix of NMVOC species.[51,52]

All emissions, except BC, increase from 2006 to 2013 in this work. Decline in BC of 0.2% a^{-1}, not seen in the other emission inventories, is due in large part to reduced kerosene use. Kerosene is prohibitively expensive, and cheaper alternatives, like solar power, are becoming more readily available.[9] Flaring of natural gas is a smaller source of BC than kerosene (Figure 2), but is declining rapidly (5.0% a^{-1}). The reason for the decline in flaring is speculative, but proposed causes include increased utilization of natural gas for energy generation, reinjection of natural gas into oil wells to increase crude oil production, and disposal of natural gas by venting instead of flaring.[19]

The rate of change in emissions in this study is very different to Liousse et al. projected emissions for 2030. Liousse et al. forecast 2030 emissions with a fuel consumption model and assume no environmental regulation. Liousse et al. project steep increases in NMVOCs and NO_x emissions from 2005 to 2030 of 9.6% a^{-1} and 19% a^{-1}, respectively. By comparison, increase in pollution from 2006 to 2013 in this work is 2.2% a^{-1} for NMVOCs and 2.5% a^{-1} for NO_x. Positive trends in satellite observations of total column formaldehyde (a high-yield oxidation product of NMVOCs) are only detected over three cities in Africa and range from 1.1 to 2.7% a^{-1}.[53] These compare more favorably with NMVOC emission trends in this work (2.2% a^{-1}) than in Liousse et al. (9.6% a^{-1}). Increase in tropospheric columns of NO_2 observed from space range from

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1.5 to 13% a^{-1} and are detected in most large African cities.[54,55] This can be compared to 2.5% a^{-1} in this work and 19% a^{-1} in Liousse et al.

Emissions from the RCP scenarios in Figure 3 are determined assuming strict environmental policy, and so both RCP scenarios predict similar 2030 emissions of NO_x, NMVOCs, and OC. Consistent DICE-Africa and RCP trends in CO, NO_x, NMVOC, and OC emissions may imply delay in the rapid increase in pollution projected by Liousse et al., as strict environmental policies imposed by the RCP scenarios seem unrealistic for Africa. The continent is yet to experience rapid industrialization and air quality degradation that precede implementation of air quality policy.

We use GEOS-Chem to determine the impact of diffuse and inefficient combustion emissions on air quality in 2006. Figure 4 shows maps of the contribution of diffuse and inefficient combustion emissions to annual mean surface $PM_{2.5}$ and ozone, obtained as the difference between simulations with and without DICE-Africa emissions. Contribution to surface concentrations of pollutants is highest in populous areas (Figure 1 and Supporting Information, Figure S1). The largest enhancement in pollutant concentrations is in southern Nigeria. There diffuse and inefficient sources of combustion contribute 5 $\mu g\ m^{-3}$ to annual mean surface $PM_{2.5}$. Enhancements in the cancer-causing pollutants benzene and formaldehyde, shown in the Supporting Information (Figure S3), exhibit similar spatial patterns to $PM_{2.5}$.

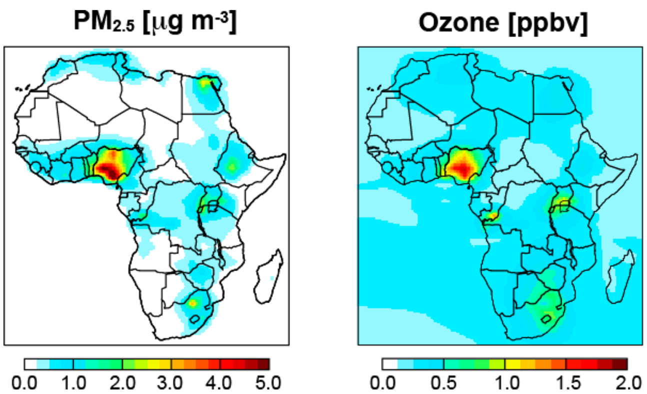

Figure 4. Air quality impact of diffuse and inefficient sources of combustion. Maps show annual mean surface $PM_{2.5}$ and ozone for 2006 obtained as the difference between GEOS-Chem simulations with and without pollution sources calculated in this work (see text for details).

The increase in annual mean surface ozone is small (<2 ppbv in southern Nigeria) and consistent with a modeling study that found anthropogenic ozone precursors in Africa to be much smaller contributors to surface ozone than intense seasonal biomass burning (>40 ppbv).[1] Biomass burning inventories estimate much larger NO_x emissions in 2006 (4.0 Tg NO from FINN and 7.1 Tg NO from GFED3) than DICE-Africa (1.0 Tg NO). Backup generators and cars are a growing source of NO_x in Africa (Figure 2) and so increased use of both will lead to very high ozone production efficiency, due to sensitivity of ozone production to the high NMVOC-to-NO_x emission ratios typical of inefficient combustion.[56] Long-lived NMVOCs and CO contribute 0.29 Dobson Units (DU; 1 DU = 2.69 × 10^{16} molecules cm^{-2}) to tropospheric ozone averaged across 50°N–50°S (Figure S4), offsetting the Earth's radiative balance in a region that is very sensitive to changes in ozone.[57]

There are large data gaps in Africa that contribute to uncertainties in the inventory developed in this work. Country-level fuel use data are often based on limited field measurements that are then extrapolated to the rest of the country. Data are lacking on the location of paved and unpaved roads to map car and motorcycle use and estimate fugitive road dust emissions. There is also insufficient data to distinguish diesel/gasoline use by passenger and nonpassenger vehicles. Information on the extent and magnitude of electronic waste burning and the associated emission factors is also limited. Emission factors are missing for important pollution sources in Africa, in particular NMVOCs and NO_x from kerosene. Persistent organic pollutant (POPs) emission factors are rarely reported for the sources considered here. Concentrations of the POPs polycyclic aromatics hydrocarbons (PAHs) north of the Niger Delta are among the highest in the world,[58] necessitating better understanding of the contributing sources.

We have developed the DICE-Africa inventory to address missing, outdated, and misrepresented data in global and regional inventories and assess the effect of these sources on ambient air quality. We obtain NMVOC emissions in Africa that are comparable with estimates from global inventories, but with a very different distribution of contributing compounds. Increases in emissions from 2006 to 2013 are consistent with the RCP scenarios, but weaker than projections from a fuel consumption model. The satellite record shows slow growth in pollution in urban centers in Africa, similar to the trends in this work. We estimate with GEOS-Chem that diffuse and inefficient combustion sources contribute 5 $\mu g\ m^{-3}$ to annual mean surface $PM_{2.5}$ in populous Nigeria. The contribution of anthropogenic diffuse and inefficient combustion to surface ozone is small compared to that from biomass burning, but this may change as NO_x emissions increase with increased use of backup generators and cars. Africa is 20% of the Earth's land surface, and is projected to be 40% of Earth's population by 2100.[59] There is pressing need for comprehensive country-level activity data and sustained in situ measurements to better constrain diffuse pollution sources in Africa, and assess the impact on global atmospheric composition and local air quality.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.6b02602.

Detailed methodology to derive emissions and figures of population distribution, sector-specific emissions, and simulated contribution of diffuse and inefficient combustions sources to annual mean surface formaldehyde and benzene concentrations and tropospheric ozone (PDF)

Tables of country-level population, activity data, and emission factors (XLSX)

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Notes

The authors declare no competing financial interest.

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