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Natural and human-related sources of ozone-forming trace gases in southern Africa

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Nitric oxide (NO) reacts with hydrocarbons and carbon monoxide (CO) in the presence of sunlight to form ozone in the lower atmosphere. Tropospheric ozone can be harmful to plants and animals, and is usually regarded as a symptom of industrial or vehicular pollution. The cloud of tropospheric ozone which forms over southern Africa every spring probably has its main origin in natural emissions of the ozone-forming trace gases, including CO from vegetation fires, emissions of NO from soils, and hydrocarbons from plants. The observed levels of ozone are not dangerous to humans, but form a high background level to which pollutants of human origin are added.

When nitric oxide, carbon monoxide and hydrocarbons, including methane (CH_4) and non-methane volatile organic compounds (NMVOC), are present in air in concentrations of a few parts per billion (ppb) and are exposed to sunlight, ozone (O_3) is formed.[1,2] Ozone in the stratosphere has the well-known beneficial effect of shielding the Earth's surface from ultraviolet radiation, but when present in the troposphere (the lower atmosphere, between the ground and the tropopause at about 12-km altitude) in concentrations above about 60 ppb, it can cause direct damage to plants and animals, or can react further with other pollutants to form toxic gases such as peroxyacetyl nitrate (PAN).[3,4] Ozone in the troposphere is also a potent greenhouse gas.[5]

The observation, inferred from the Total Ozone Monitoring satellite (TOMS), of a cloud of tropospheric ozone over the southern Atlantic Ocean each year around September, stimulated interest in southern Africa as a source of ozone-forming trace gases.[6] The first hypothesis was that they were produced by vegetation fires, which were known to be frequent and widespread in Africa during the southern hemisphere winter. Ozone sampling using sondes, aircraft and ground stations during the Southern African Fire– Atmosphere Research Initiative (SAFARI)[7] campaign in 1992 confirmed that the tropospheric ozone concentrations over remote, sparsely populated parts of southern Africa approached levels typically found in heavily industrialised regions. High concentrations of the ozone-precursor trace gases were also observed (G.W. Harris *et al.*, in prep.)

Rough calculations had suggested that biomass burning might be responsible for a large fraction of the global production of ozone-precursor gases.[1] However, when the vegetation fire ('pyrogenic') emissions were recalculated for southern Africa[8] using the improved data, arising from SAFARI, on area burned, fuel load and emission factors, the emission estimates for the subcontinent were revised downward nearly tenfold. Was this quantity consistent with the magnitude and extent of the observed ozone cloud? A second difficulty was in the timing of the emissions and the ozone cloud. The ozone peaks in about September, but the occurrence of fires, as observed by the Advanced Very High Resolution Radiometer carried on the NOAA weather satellite, is in July.[9] Due to its high reactivity, the longevity of ozone in the troposphere is only a few weeks.

This paper approximately quantifies the known sources of ozone-forming trace gases in southern Africa and examines their characteristic seasonal emission patterns, in order to suggest which sources may be involved in producing the seasonal ozone cloud.

'Pyrogenic' emissions from vegetation fires

Many of the vegetation types of southern Africa are fire-prone, due to the existence of a long, hot dry season over most of the subcontinent.[9] It is inferred from the evolutionary characteristics of the species they contain that fires have been a frequent feature of these ecosystems for tens of millions of years. It is not known whether fires have become overall more or less prevalent in the colonial and post-colonial period relative to the precolonial period. There is reason to believe that they may have been less frequent, but more extensive in the precolonial past, but little solid evidence exists for extrapolation to the continental scale.[10]

The 3 million km^2 of moist savannas on infertile soils (in particular, the miombo woodlands of south central Africa) are the main location of fires, and the largest pyrogenic source of CO, CH_4 and NMVOC. The protein content of the grasses in these savannas is too low during the dry winter months to support a substantial biomass of large grazing animals. As a result, the accumulated dry grass is set alight in the early dry season (beginning, in the north, in about May) by pastoralists seeking to stimulate more nutritious new grass, and accidentally by farmers and charcoal-burners. It was previously believed that virtually the entire area

burned every year, but high-resolution satellite images show that at most one third of the core moist savanna area burns in a given year, decreasing to a tenth or less on the arid fringes. There is less fuel to burn in the savannas on fertile soils, since it is mostly consumed by the large biomass of grazers which they support. The high-elevation grasslands of South Africa burn once every three to five years, usually in late winter or early spring, but are increasingly planted to maize or wheat, which is not burned. The fynbos, a sclerophyllous thicket which occurs mainly in the winter-rainfall areas of the southern Cape, burns approximately once every 20 years, in summer. The large desert areas of the west coast of southern Africa hardly burn at all.[10]

The only crop which is routinely burned in southern Africa is sugar cane, prior to harvest, and this is a declining practice. Other residues are seldom burned, due to their high value as cattle fodder in subsistence societies. About 0.5% of the 1.4 million ha of plantation forest in South Africa burns accidentally every year, typically in late winter or early spring.[10]

The amount of a given trace gas released per unit mass of fuel consumed (known as the 'emission factor') for southern African savannas have been revised downward for all the products of incomplete combustion, such as CO, CH_4 and NMVOC. Fires in savannas mostly consume fine, dry, well-oxygenated fuels such as dry grass (although a significant part of the fuel consists not of grass but dead fallen tree leaves and twigs and litter), leading to flaming combustion rather than the smouldering combustion which dominates forest fires.[11] Emission factors for NO and soot from savanna fires have been increased, since they are primarily products of flaming combustion. Nevertheless, the total emission estimates for southern Africa have decreased due to the lower estimates of area burned and fuel load per unit area.

'Biogenic' emissions from soils, living vegetation and animals

Soil microorganisms are known to produce large quantities of methane and nitrous oxide (N_2O) when the soil is saturated for long enough to become anaerobic.[12] These circumstances are relatively uncommon in southern Africa, since the rainfall over most of the area is less than the potential evaporation, and the soils are generally well drained. Suitable anaerobic conditions are found in the seasonally flooded swamps (known as dambos or vleis) which form in the valleys of some landscapes. In the miombo landscape, up to one fifth of the surface area can be occupied by dambos, although they are individually too small to appear on regional vegetation maps. The duration and magnitude of methane emissions from these systems is at present unknown, but under investigation. The seasonal pattern is likely to be a peak in mid- to late summer, a few weeks after the low-lying areas flood.

The upland soils are seldom saturated for a sufficient period to generate methane and nitrous oxide, even following rain. Most of the time they oxidise methane at a low rate.[13–15] Termites, which are common in southern Africa, produce methane in their guts, but a large fraction is oxidised by the soil before escaping the mound. Calculated at the landscape scale, termites are thought to be a minor methane source.[16]

On the other hand, the aerobic upland soils are an important source of NO. The biochemistry of this source is not well understood. It is thought that one of the intermediates in the process of conversion of ammonium to nitrate (known as nitrification) can also be converted to NO, which occurs spontaneously and results in a nitrogen 'leak' equivalent to about 5% of the annual nitrogen turnover flux.[17–19] Some plant communities, such as undisturbed highveld grasslands in South Africa, block the nitrification step, absorbing their nitrogen in the form of NH_4^+.[20] This could be an adaptation which conserves nitrogen in the ecosystem. In heavy textured soils, the highly reactive NO is converted mostly to nitrate before it can diffuse out of the soil, and in dense vegetation the remainder is absorbed by the plant canopy. However, in deep, sandy soils, such as those of the Kalahari sand basin which covers about a third of southern Africa, most of the soil nitrogen cycle passes through the nitrification step, and the NO produced escapes from the soil and the sparse vegetation. These are major NO sources, where the measured emission rates are among the highest observed anywhere.[17–19]

The seasonal pattern of NO emission from the soil basically follows that of the rainfall. The emission of NO is predominantly a function of soil water content, peaking around 35% water-filled pore space (typically just below field capacity), and declining at higher water contents.[18] The first rainfall event following a prolonged dry spell produces an exceptionally large pulse of NO, which can last for several days. Our working hypothesis is that the process of mineralisation of organic nitrogen in the soil continues at a slow rate during the dry spell, but the ammonium it produces is not taken up by plants. The first wetting results in rapid nitrification until this accumulated substrate is used up. This 'first rains' NO pulse has been found to last for only one wetting, and to produce NO at a rate about twice that of subsequent wettings. It accounts for about 6% of the annual NO emission, but since the rate of ozone production is non-linearly related to NO concentration, could be disproportionately important for springtime ozone generation.[17]

All live plants release hydrocarbons through their leaves, at a rate which varies greatly between species, but which can amount to several per cent of the carbon assimilated by the plant.[21] Grasses, in general, are low hydrocarbon emitters but data to support this are extremely few. About half of the emission from trees is in the form of isoprene, and the remainder can be in a wide variety of monoterpenes. The rate of emission is correlated with intercepted solar radiation and the rate of photosynthesis, and for isoprene is apparently highest when the leaf temperature is high and the plant is water-stressed.[22,23] These are conditions which are prevalent in Southern Africa. A global model of NMVOC emissions, based on light interception and leaf area, but with very scanty species-specific data outside of the temperate areas of the northern hemisphere, suggested that savannas could be the largest biogenic source of hydrocarbons.[24] A screening survey of savanna trees at two locations in South Africa (the *Burkea africana* woodland at Nylsvley, and *Colophospermum mopane* shrublands south of Phalaborwa) revealed many species which were high emitters of isoprene or monoterpenes. More detailed measurement and modelling of the NMVOC emissions from the dominant species at these two locations suggested a landscape level emission of 0.65–12 mg C m^{-2} h^{-1}. Assuming 12-hour days and a tree leaf duration of 170 days per year, this suggests annualised emissions of 1.3–24.5 g C m^{-2} yr^{-1} (refs 24, 25). Extrapolating these values to the area of savannas in southern Africa (6.7×10^{12} m^2) gives a first approximation of the NMVOC emissions from the region of 8.7–164 Tg C yr^{-1}). The seasonal pattern of NMVOC emissions is anticipated to parallel that of the woody-plant leaf area, rising rapidly in spring and declining in winter when the trees shed their leaves. The emission-promoting conditions of high irradiance, high leaf area and probable water stress are most likely in late winter or early spring, since over most of the area the woody plants expand their leaves just before the first rains.

Methane is produced by the anaerobic bacteria which inhabit

the digestive systems of mammals. Methane production is especially high in ruminants, amounting to about 15% of the energy ingested by the animal, and increases as the quality of the forage decreases.[26] Southern Africa was historically the habitat of large herds of wild ungulates. The biomass per square kilometre of these ungulates under natural conditions can be estimated from the mean annual precipitation and the soil type. Some further assumptions about mean body size allows the energy intake to be calculated, and thus the probable precolonial methane production. For all of the rangeland areas of Africa south of the equator, this estimate is about 323×10^9 g CH_4 yr^{-1}.[27] At the present time, about 90% of the wild ungulates have been replaced by domestic cattle. In some circumstances the ungulate biomass density may have increased (possibly by a factor of 2) as a consequence, but in others it is probably a simple substitution.[27] The seasonal pattern of methane emissions from ungulates is thought to be fairly constant, with a slight peak in late winter when the forage quality is at its lowest.

Carbon monoxide is emitted when dead leaves ('litter') are exposed to the ultraviolet radiation in sunlight.[28] The rate of emission is relatively constant across a wide variety of litter types, and is positively related to the intensity of the sunlight and the air temperature. Extrapolating from the few available small-patch estimates to the whole subcontinent provides a rough estimate of 0.5×10^{12} g CO yr^{-1}.

Emissions from burning fossil fuels ('industrial')

South Africa dominates the industrial economy of southern Africa, and produces about 88% of the 'industrial' emissions.[29] South African emission data have therefore been used in this survey as proxies for the entire region.[27] The largest fraction of the fossil fuel combustion in the region is associated with the generation of electrical power in coal-burning power stations. This is a year-round activity, which peaks slightly in the winter months due to increased demand for domestic and office heating. Coal is also used in the metallurgical industries, at a fairly steady year-round rate, and for domestic cooking and heating, with a strong winter peak. Oil is burned largely in the form of petrol and diesel, in internal combustion engines. The rate of emission is assumed to be approximately constant throughout the year. The uncertainties associated with industrial emissions are small (about 20%) relative to those associated with biogenic and pyrogenic emissions.

Other human-induced emissions

Most of the population of southern Africa depends on wood collected from natural woodlands, savannas and shrublands for domestic cooking and heating. The combustion of wood in open hearths, as is the norm in rural southern Africa, generates large quantities of CO and hydrocarbons relative to burning it in enclosed stoves, but small amounts of NO_x, due to the smouldering nature of the combustion. The annual per capita use of fuelwood ranges from 250–1200 kg per person, as a result of differences in fuelwood availability, climate and cultural factors.[30] Assuming a rural population of 61 million people,[31] this translates to an annual emission of 1100–5400 Gg CO yr^{-1}, 30.5–142 Gg CH_4 yr^{-1}, 24–115 Gg NO yr^{-1} and 30–146 Gg NMVOC yr^{-1}.

The urban population in South Africa, Namibia and Zimbabwe mostly burn coal or use electricity. Elsewhere, the wood is typically first converted to charcoal. In this form it has a much higher energy density, and can thus be transported to urban areas more economically. Charcoal is typically burned in semi-closed braziers, where combustion is relatively efficient. The process of conversion of wood to charcoal in traditional kilns, however, is very inefficient, and generates large quantities of CO and CH_4. Assuming an annual per capita consumption of 100 kg (range 26–742 kg) of charcoal, and an urban population of 113 million,[31] the total emissions (including during the charcoal manufacture) are 500 Gg CO yr^{-1}, 60 Gg CH_4 yr^{-1}, 0.25 Gg NO yr^{-1} and 11.4 Gg NMVOC yr^{-1}.

What causes the ozone cloud?

The way in which tropospheric ozone is calculated from the TOMS observations has been improved, allowing the technique to be applied equally well over land as over the ocean.[32] As a result, it is now apparent that the August to October southern African tropospheric ozone maximum extends over most of south central Africa, as well as over the adjacent Atlantic Ocean. Ozone production may be promoted over this region during the winter months by the formation of an extremely stable anticyclonic circulation. These gyres confine the precursor gases in a relatively thin (0–6 km) near-ground layer, which accumulates emissions for periods of two to three weeks before venting them over the Atlantic and Indian oceans.[33]

Table 1 shows that pyrogenic, biogenic and industrial (including domestic wood-burning) sources all contribute significant amounts of ozone precursors to the atmosphere over southern Africa, but that each gas is dominated by one source. In the case of methane, the major uncertainty relates to the as yet unquantified source due to seasonal swamps in south central Africa, which may bring the biogenic sources more in line with the industrial sources, and would tend to lead to a greater summer peak in emissions.

Even allowing for the huge uncertainties associated with the emissions of NMVOC from plants, it seems that this is by far the dominant source. The timing of the emissions, inferred from what is known of the ecophysiology of the species, is consistent with the springtime ozone maximum, but has not been verified by a rigorous year-round observation campaign. This must be a high priority in the future.

Vegetation fires are the dominant source of carbon monoxide. The timing of the pyrogenic emission peak (Fig. 1) is about a month too early to be clearly responsible for the peak in ozone observations, unless some peculiarity of the southern African circulation slows down ozone formation or extends its life in the atmosphere. If the estimated pyrogenic emissions from all the savannas of the world are reduced by a factor similar to recent revised calculations for southern African savannas, there could be a problem accounting for the observed concentration of CO in the global atmosphere. The gap in the budget can be partly satisfied by sources such as the photolysis of litter and domestic wood combustion, which are poorly quantified at present. Modelling of CO concentrations over Africa could be one way of testing the validity of the revised pyrogenic emissions.

Nitric oxide was formerly thought to originate mostly from

Table 1. Summary of the emissions of ozone-forming trace gases ($\times 10^{12}$ g yr^{-1}) from broad categories of sources in Africa south of the equator (9.6×10^{12} m^2).

	CH_4	NMVOC	CO	NO
Pyrogenic	0.5	0.54	14.9	1.04
Biogenic	>0.32	30–500	0.5	0.29
Industrial + domestic	>2.59	0.61	5.6	1.95
Total	4	100?	21	3.3

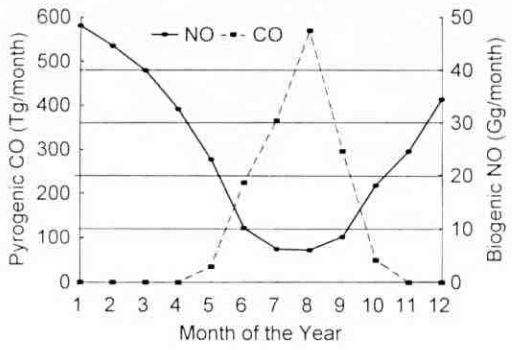

Fig. 1. The estimated seasonal emission patterns of key sources of ozone-forming trace gases in southern Africa. There is great uncertainty associated with the absolute magnitude of these emissions (50% or more, in some cases), but the seasonal timing is determined by observable drivers such as rainfall events and the occurrence of fires, and is therefore thought to be quite reliable. The peak of the tropospheric ozone maximum is centred on October. Month 1 = January; month 12 = December.

combustion, either of fossil fuels, or vegetation. The high NO emission rates from soil measured in South Africa[17–19] are supported by data from savannas in Brazil,[34] and from aircraft observations during the SAFARI campaign, of NO clouds far from any pyrogenic sources, soon after the first rains (G.W. Harris *et al.*, in prep). While the modelling of NO emissions at a subcontinental scale is still at an early stage, it is clear that soils represent a major NO source. The timing of the ozone cloud is not consistent with the peak of biogenic NO emissions, which models suggest occurs in January, but the period of production (October to June) is broad enough that soils could be significant contributors to the phenomenon, especially in the early spring.

The combination of NO, CO and NMVOC is the starting point for the photochemical smog which is often blamed on motor vehicles. It is clear that biogenic sources of NO and NMVOC, and pyrogenic sourcesof CO, can contribute to high background levels of these pollution precursors, and will need to be considered when environmental receiving capacity limits are set for the industrial emissions of these gases.

In conclusion, none of the major sources can yet be clearly eliminated as not contributing to the springtime ozone maximum, either on the basis of quantitative insufficiency or of timing. It seems probable that the phenomenon results from a combination of the tail end of the pyrogenic emissions and the start of the biogenic emissions. If this is the case, it has been a feature of the atmospheric chemistry of southern Africa for millions of years. At their maximum, the concentrations of ozone currently measured at ground level in rural locations are not toxic to plants or animals. When an increasing pollution load of human origin is added to this background, damage thresholds could be exceeded in the late winter and early spring.

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