Land Use Change Impacts on Air Quality and Climate

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1. Introduction

Historical land use and land cover change (referred to herein as "land use change") has dramatically altered the Earth's landscape, perturbing energy, moisture, and chemical fluxes and impacting the Earth's climate.[1-3] Land use change (LUC) in the next century has been projected to have profound impacts on regional climate.[4] These changes connect to critical issues of food security, energy supply, and biodiversity. Large-scale perturbation of the biosphere will also play a major role in determining atmospheric composition, with implications for both air quality and climate. Our goal here is to review current understanding of the interplay between land use change and atmospheric chemistry, with a focus on short-lived atmospheric pollutants.

Particulate matter (PM, or aerosols) and ozone in the atmosphere impact human and environmental health, visibility, and climate. Exposure to outdoor particulate matter is a major environmental cause of mortality world-wide, responsible for over 3.2 million premature deaths per year.[5] Ozone exposure contributes to chronic respiratory and cardiovascular illness.[6] PM at the surface also degrades visibility, a concern for pristine wilderness and recreation areas. In addition, aerosols and ozone can alter the Earth's climate, via both direct effects (absorption or scattering of radiation) and the indirect effects of aerosols (changes to the albedo or lifetime of clouds). Further, particle deposition is an important mechanism by which nutrients and/or toxins can be introduced into ecosystems, and thus may have important biogeochemical consequences.[7] In addition, elevated surface ozone concentrations damage plants,[8, 9] with potential implications for forest growth[10] and crop yields.[11, 12]

The terrestrial biosphere is a major source of natural aerosols, including dust, smoke, and primary biological aerosol particles (PBAP), as well as volatile organic compounds (VOCs), and nitrogen oxides (NO_x) which are both aerosol precursors and play a role in ozone formation. The role of natural emissions in controlling global air quality and climate, both independently and synergistically with anthropogenic pollution, is poorly understood. In addition, deposition to the surface, and specifically uptake by

vegetation is an important sink for trace gases and particles, particularly for ozone.[13] Indirect climate forcing from pollution aerosol interactions with clouds is particularly sensitive to the natural aerosol background.[14, 15] In turn, recent work has emphasized the critical role that climate plays in controlling the level of natural aerosols.[16] Dynamically changing land cover, responding to both human forcing and natural factors, is also a key factor in modulating air quality and climate. Recent work by Ward et al.[17] suggests that anthropogenic land use change has resulted in a warming that is equivalent to up to 45% of the net warming estimated as present-day anthropogenic radiative forcing.

Human-driven land use change includes agricultural development, urbanization[18] as well as forestry practices[1] and agricultural waste burning or clearing. These changes have greatly altered the Earth's surface, with between one-third and one-half of the land surface now modified by humans.[3] Cropland and pasture now cover 34% of the Earth's ice-free land surface,[19] with agricultural lands replacing natural forests and grasslands. Anthropogenic induced land use change may have started as long as 10 000 years ago, [21] although the significance of changes prior to the industrial period are highly uncertain. [21-23] By the year AD 100, between 2% [23] and 10% [21]of the global land surface may already have been under anthropogenic land use. By AD 1850, modification may have increased to between 10% [23] and 21% [21]of global land surface, meaning substantial LUC had already occurred by the start of the industrial period.

Deforestation[24] is often associated with the establishment of food crops and pasturelands for meat production[25] as well as biofuel crop development[26-28] including plantation development.[29] Forests covered approximately 50% of the Earth's land surface 8000 years ago compared to 30% today.[20] Between 1980 and 2000, 83% of new agricultural land across the tropics was established on previously forest land.[30] Forestry management practices include fire management,[31] afforestation and reforestation,[32] and selective logging[33] as an approach to mitigate deforestation. Humans use fire as a land-management tool to clear forests and vegetation for agriculture, maintain pasture and to burn crop residues and agricultural waste. Human activity can reduce fire through fire suppression, as well as through land use change, grazing and wood harvest that reduce and fragment fuel loads limiting fire spread.[34, 35] It is possible that humans have

been altering fire extent for as long as 1.5 My, with impacts likely to have increased as human population expanded and land-use change progressed.[36]

Natural land cover change includes wildfires,[37] biome shifts,[38, 39] CO_2 fertilization,[40] and insect infestation,[41] often in response to climate change. Projecting these land cover changes therefore relies on regional estimation of future climate and biospheric response. Insect infestations can affect the widespread health of forest ecosystem. The recent bark beetle outbreak in western North America destroyed over 100,000 km^2 of forest over the decade of 2000-2010.[42] Fire is a natural component of many ecosystems, controlling vegetation structure and functioning. Fires are a major cause of landscape change. The frequency and extent of fire is impacted both by both climate and human activity.[43] Increases in atmospheric CO_2 concentrations decrease the stomatal conductance of plants, thereby enhancing their water, light, and nutrient efficiency.[40] This so-called "CO_2 fertilization" effect increases vegetation productivity and density. This effect is typically viewed as "natural land use change" in response to an altered atmospheric state, however throughout the Anthropocene the major driver of increasing atmospheric CO_2 concentrations has been anthropogenic emissions. Conversely, afforestation, which we describe above as anthropogenic land-use change, may also occur naturally through ecological succession.

Table 1 contrasts the historical extent and timescales of typical types of land use change. Conversion of natural ecosystems to agriculture has impacted an average of 0.3 million km^2 annually from preindustrial to present day. [19] Fires impact an even greater area, with the global area burned by fire estimated to be 3-6 million km^2 annually.[45-47] Biome shifts and CO_2 fertilisation may result in subtler shifts to vegetation over even more extensive areas. Large changes in global forest cover continued over the last decade (2000 to 2012) when there was 0.2 million km^2 $yr^{-1}$of forest loss and 0.07 million km^2 yr^{-1} of forest gain due to a combination of fire and anthropogenic induced LUC.[44]

Figure 1 shows global historical trends (1850 to present-day) and future projections (out to 2100) for land use area in 3 major categories.[48] This illustrates the major historical trend through expansion of agricultural croplands at the expense of grasses and forests. Figure 1 also highlights the uncertainty in

future land use conversion as described in the RCP scenarios. Hurtt et al.[49] project anywhere from a 13% decrease to a 24% increase in croplands over the 21st century, reflecting either intenstification or extensification of croplands to meet rising global food demand. These changes likely had, and will continue to have, a large impact on air quality and climate. Our objective is to review the literature concerning the impacts of land use change on short-lived pollutants, and provide an integrated perspective on the resulting radiative impacts. Land use change largely modulates natural emissions, however in this review, we also consider agricultural sources of ammonia, which is a precursor to inorganic aerosol formation and is tied to land use conversion for agriculture. The impacts of LUC on greenhouse gas emissions is outside the scope of this review.

2. Land Use Change Impacts on Sources and Sinks of Short-Lived Atmospheric Pollutants

Here we review the literature on land use change and its impacts on emissions, deposition, and air quality. This section is broken down by key specific chemical species. The micrometeorological feedbacks of vegetation changes associated with land use change (e.g. radiation, Bowen ratio, boundary layer heights, etc.) on air quality are beyond the scope of this review, however these factors may also play a critical role, as discussed for example by Ganzeveld et al.[50] Throughout this review we employ the terms radiative effect (RE) and radiative forcing (RF). The RE is the instantaneous radiative flux imbalance resulting from the presence of a constituent of the Earth's atmosphere. The RF is defined by the IPCC as the externally-imposed perturbation to the radiative balance of the planet (excluding climate feedbacks), and is often approximated as the anthropogenic change between pre-industrial and present-day. Thus, RE is often the best metric to characterize the baseline impact of short-lived atmospheric constituents (e.g. ozone and aerosols), and RF represents the policy-relevant metric for human perturbation.[51] For aerosols, we restrict our analysis to the direct radiative effect (DRE) and direct radiative forcing (DRF) and exclude aerosol indirect effects.

The beginning of substantial human influence on the Earth and climate system is usually assumed to be the start of the industrial period, variously defined in the studies reviewed here as AD 1750 to AD 1850. Humans started altering land cover and fire extent well before the beginning of the industrial period, so in this context the definition of pre-industrial may be less useful. However, very few studies have explored how land use change before the preindustrial altered short-lived atmospheric pollutants, so here we confine our discussion to changes after the pre-industrial.

Table 2 summarizes the past and projected emission changes related to land use change discussed in this section.

2.1 Terrestrial Biogenic Volatile Organic Compounds

2.1.1 Introduction

Biogenic volatile organic compounds (BVOCs) are among the key reactive atmospheric constituents emitted from the biosphere. Terrestrial emissions of isoprene (~500 TgC yr^{-1}) rival methane sources as the largest flux of reactive organic carbon to the atmosphere. Isoprene and monoterpenes play an important role in ozone[52] and secondary organic aerosol (SOA) formation[53, 54] in the troposphere. Measurements of organic aerosol (OA) suggest that these particles make up an important, sometimes dominant, fraction of fine particle mass in the atmosphere[55] and originate primarily from secondary sources,[56] however their formation and processing in the atmosphere remain highly uncertain, and therefore poorly characterized in models.[57] The global source of biogenic SOA (BSOA) is estimated at ~17-52 $Tgyr^{-1}$, with an associated global mean burden of ~0.5-0.77 Tg.[58-61] However model underestimates of OA by a factor of two or more based on both satellite and in situ observations[62-64] may include an underestimate of this biogenic secondary source. The DRE of BSOA from monoterpenes and isoprene are estimated at -0.01 to -0.13 Wm^{-2} [65-67] and -0.08 Wm^{-2},[67] respectively. An additional -0.02 to -0.19 Wm^{-2} of cooling is attributed to the indirect radiative effect of monoterpene BSOA.[65, 66] A larger indirect effect from BSOA is possible if BVOC oxidation products participate in new particle formation.[67] In the presence of nitrogen oxides (NO_x) as in urban environments, isoprene contributes to ozone

formation. In remote clean conditions (low NO_x) isoprene oxidation can consume ozone. Isoprene also plays an important role in controlling the oxidizing capacity of the troposphere via OH concentrations, and thus indirectly, the lifetime of methane (an important greenhouse gas).[68] However, there are significant uncertainties regarding the degree to which OH is consumed by isoprene oxidation,[69-71] which limits our ability to link isoprene emissions changes with impacts on methane.

BVOC emissions are typically described using empirical (e.g. [72]) or process-based models tied to photosynthetic activity (e.g. [73]). These emissions are highly sensitive to temperature (e.g. [74, 75]), and are therefore expected to increase with climate change.[76, 77] Underlying basal emission rates also vary substantially across plant functional type (PFT) categories (e.g. emissions of isoprene from deciduous broadleaf temperate trees are typically an order of magnitude greater than from crops or grasses).[72] Therefore emission estimates are highly dependent on the description of the (evolving) underlying vegetation class, as well as phenology and climate. In addition, the variability in emissions between vegetation species within a PFT class is substantial, poorly understood, and can introduce considerable uncertainty in emissions at a local scale.[78] This may be of particular importance when considering the evolution of BVOC emissions through ecological succession. Both Lathière et al.[79] and Heald et al.[76] find that global isoprene emissions differ by ~30% when gridded emission factors (accounting for sub-PFT vegetation speciation from Guenther et al.[80]) were used compared to fixed PFT emission factors. Further uncertainty may be introduced by the description of land cover itself. Guenther et al.[80] suggest that this uncertainty may be larger than the impact of climate change on isoprene emissions. Gulden et al.[81] show how two different land cover datasets over Texas result in mean statewide emissions of isoprene that differ by a factor of three. Oderbolz et al.[82] estimate a range of almost a factor of two in estimated isoprene and monoterpene emissions when using three different vegetation inventories for Europe.

Given the importance of BVOC emissions to air quality, models have been applied at various scales to investigate the impact of natural and anthropogenic land use change. Table 3 summarizes the modeling studies discussed below.

2.1.2 Natural Land Use Change Effects on BVOC Emissions and Air Quality

Rising atmospheric CO_2 concentrations lead to enhanced vegetation density through CO_2 fertilization, thereby increasing BVOC emissions. Tao and Jain[83] estimate that this effect has increased global isoprene emissions by 7-14% over recent decades (1981-2000). Isoprene emissions are projected to grow by a further 17-27% over the 21st century under rising CO_2 concentrations.[84-87] The range in response may be larger at a local scale or when considering multiple future scenarios, for example isoprene emissions over the same time horizon are projected to increase by 26-41% in Europe.[88] Several of these studies have projected isoprene emissions under simultaneous changes in climate and atmospheric CO_2 concentrations, and therefore do not specifically separate the role of CO_2 fertilization (see Table 3). The impact of CO_2 fertilization on future monoterpene emissions is less well studied; projections of global 21st century emission increases range from 10% to 51%. [86,85] Overall, changes in the tropospheric O_3 burden associated with CO_2 fertilization are estimated to be small,[87] but Sanderson et al.[84] and Wu et al.[86] suggest that local surface O_3 concentrations could drop by up to 10 ppb or increase up to 50 ppb as a result of CO_2 fertilization. A single study has examined the impact of CO_2 fertilization on SOA burden, projecting a 20% increase over the 21st century.[86]

Exposure to elevated atmospheric CO_2 has also been shown to directly inhibit isoprene production in plants.[89-91] While this is not a direct response to land use change, the suppression of isoprene at elevated CO_2 levels counteracts the concomitant CO_2 fertilization effect. Global modeling studies suggest that this effect decreases estimates of late 21st century isoprene emissions by 26-52%,[87, 92-96] which may entirely offset temperature-driven increases in emissions. Overall these decreases in isoprene emissions are estimated to produce a 4% decrease in the tropospheric O_3 burden,[87] with local surface O_3 changes between -10 ppb to +10 ppb.[95] Tai et al.[94] find that a 26% decrease in isoprene emissions due to CO_2 inhibition in 2050 leads to a corresponding 25% decrease in BSOA surface concentrations. Lathière et al.[79] estimate that the net effect of atmospheric CO_2 concentrations on global isoprene emissions (through fertilization and inhibition) was a 21% decrease over the 20th century; in contrast Unger [97] estimate that atmospheric CO_2 produced a 7% net increase in emissions over the same

time period. Over Europe, Arneth et al.[88] estimate that isoprene emissions will decrease by 12 to 36% over the 21st century due to the net effect of atmospheric CO_2. Monoterpene emissions have also been shown to be inhibited when exposed to elevated CO_2,[98, 99] but this effect and its impacts on air quality has not yet been investigated in models.

Insect infestations can damage forests, and can also trigger short-term increases in monoterpene emissions.[100-103] Berg et al.[104] suggest that the recent bark beetle infestation in Western North America, which may have been initiated by a warming climate, produced up to a 4-fold increase in monoterpene emissions and local SOA enhancements of up to 40%.

Natural succession will also alter BVOC emissions. Succession model simulations for southern Texas suggest that land cover change since the 1800s has produced a 3-fold increase in BVOC emissions, largely as a result of succession from C3 grasses to C4 shrubs and trees.[105] Klinger et al.[106] estimate almost an order of magnitude change in isoprene emissions through the successional trajectory of grassland savanna to primary rainforest in central Africa. Through impacts on vegetation, fire will also alter BVOC emissions. Fire consumes biomass likely reducing total BVOC emissions, although we found no studies quantifying this effect. Succession of vegetation after fire will alter species composition changing the composition of BVOC emissions. In boreal environments, the post-fire succession of vegetation typically results in the development of grasses, followed by deciduous shrubs and trees developing into coniferous trees over a 60-100 year time cycle.[107] This implies that fire is likely to shift boreal forest environments from monoterpene dominated emissions to isoprene dominated. Isoprene and monoterpene emissions in the United States increased rapidly from the 1980s to the 1990s due to increases in forest leaf area, ecological succession, harvesting, and plantation management.[108] However these changes had little impact on surface O_3, which was driven primarily by anthropogenic NO_x emission changes over this time period.

2.1.3 Anthropogenic Land Use Change Effects on BVOC Emissions and Air Quality

Anthropogenic land use change is dominated by the expansion of agricultural lands (often associated with deforestation) and urbanization. Over the 20th century model studies suggest that this land use conversion has decreased global isoprene emissions by 15% to 22%,[79, 97, 109] with a 2% reduction estimated for the last two decades of the 20th century.[83] A continuing trend of cropland expansion with increasing global food demand through the 21st century gives rise to estimates of further declines in global isoprene emissions by 12-40%.[76, 86, 87] Arneth et al. [88] and Chen et al. [111] project even larger decreases within Europe (-42%) and the United States (-52%) over the 21st century. Given the dominance of tropical forests as a global isoprene source, tropical deforestation in particular may lead to large decreases in global isoprene emissions. Ganzeveld and Lelieveld [112] suggest that Amazonian deforestation may lead to a 30% decrease in local isoprene emissions. Lathière et al. [113] show that a scenario wherein tropical broadleaf trees are replaced with crops and grasses implies a 29% decrease in global isoprene emissions. Wiedinmyer et al. [114] explore the effects of urbanization by converting 50% of natural forests into urban or pasturelands in the United States and Amazon basin; global isoprene emissions decline by 9% as a result. Unger[110] suggests that emissions of monoterpenes have declined by 28% since 1850 due to anthropogenic land use change; however Acosta-Navarro[109] suggest that historical LUC has led to no net change in monoterpene emissions. Given that boreal forests are less impacted by anthropogenic land use change, monoterpene emissions (largely from coniferous trees) are projected to be less sensitive to these changes over the 21st century with two estimates suggesting either an increase of emissions by 2% [86] or a decline by 10%.[76] However Chen et al. [111] suggest larger local decreases of 32% over the United States. Anthropogenic land use change is projected to decrease the global burden of SOA by 14-20%,[76, 86] and modulate surface O3 by ±5 ppb.[86, 87, 111, 112, 114]

Afforestation may occur when managed lands are abandoned or selected for conservation. A study by Lathière et al. [113] investigates the impact of European afforestation and finds that complete replacement of crops with forests in this region implies a large local increase in isoprene emissions (+54%) but only a 4% global increase (with a negligible change in monoterpene emissions).

In addition to the development of agricultural lands to support food production, substantial land is converted to support biofuel production.[27] Typical fast-growing bio-energy crops, such as poplar, willow, and eucalyptus have relatively high isoprene emission capacities compared with native tree species. The same is true for oil palm, an important commodity crop in Southeast Asia, where oil palm plantations now occupying over 13% of the land area of Malaysia.[115] Measurements from Borneo, suggest that isoprene emissions from oil palm exceed emission rates from surrounding native tropical forests by a factor of 7.[116] However, no appreciable difference in surface O_3 was measured over these two landscapes.[115] Nevertheless, oil palm plantations continue to expand in Asia and several model studies have investigated the potential impacts of increasing isoprene emissions from oil palm on air quality. Interestingly, Hardacre et al. [117] suggest that in the future these plantation-driven increases may be offset by intensified food production (with forests replaced by low-emitting crops). Extreme scenarios of complete conversion of Borneo land to oil palm plantation, produce local changes in surface ozone ranging from -5 ppb to +18 ppb.[118, 119] Ashworth et al. [120] explore a realistic scenario where globally 1% of fossil fuel demand in 2020 is supplied by biofuel supplied by replacing natural rainforest with oil palm. Global mean air quality impacts are small (< 1% change in surface O_3 and BSOA) however local scale changes are more significant, with increases of up to 0.5 μgm^{-3} in BSOA in Borneo in July. When only increases in isoprene emissions are considered, surface ozone decreases by up to 3 ppb; however the addition of attendant processing plant emissions of NO_x reverses the sign of surface O_3 changes with local increases of up to 7 ppb. The same study showed that an equivalent planting scenario undertaken with short rotation coppice (SRC) at mid-latitudes shows similar, but more modest increases in BSOA and that sufficient NO_x concentrations ensure that enhanced isoprene emission from SRC lead to increases in O_3 surface concentrations. Similarly, Porter et al. [121] show how the air quality impact of replacing a coal power plant with biochar from locally grown *arundo donax* biofuel crops is almost negligible in the low NO_x environment of Oregon, but is substantial in Texas or Illinois with an average increase of 2-4 ppb in surface O_3 and 44% in surface BSOA concentrations due to enhanced isoprene emissions. Beltman et al. [122] replace 5% of current European croplands with poplar plantations, and estimate that a 15-20%

decrease in NO_x emissions would be required to compensate for the resulting increases in surface O_3. A US and Amazonia tree plantation scenario produces mean 7 ppb decreases in surface O_3,[114] whereas O_3 increases by up to 12 ppb are projected under a global SRC scenario.[117] Overall, conversion of lands for biofuel production generally elevates BSOA concentrations, but the impact on surface O_3 depends on the NO_x environment, both in terms of the location of the plantation crop and any local associated emissions. For this reason, Hewitt et al. [115] argue that nitrogen management is a key factor in managing the impacts of biofuel plantations on air quality.

While anthropogenic land use change, including cropland expansion and biofuel scenarios, can substantially perturb local OH concentrations, model studies have suggested that the global oxidizing capacity of the atmosphere, and hence methane lifetime, are relatively unaffected.[50, 86, 119, 120]

2.2 Ozone Dry Deposition

Dry deposition accounts for approximately ~20% of the O_3 loss in the troposphere.[123] This removal is highest over vegetated surfaces where O_3 is directly taken up by stomatal pores of plants.[13] Under elevated CO_2, reduced stomatal conductance can decrease O_3 deposition.[124] Dry deposition of ozone is also strongly influenced by vegetation cover. McDonald-Buller et al. [125] demonstrate this sensitivity by showing how two different land use datasets can produce differences in surface O_3 concentrations of up to +11 ppb and -6 ppb locally in Eastern Texas. Similarly, Li et al. [126] show that alternate land use descriptions over China can locally increase or decrease O_3 deposition velocities, producing differences of up to 6 ppb in simulated surface ozone. Incorrectly describing vegetation phenology can lead to surface ozone biases of more than 15 ppb.[127]

Land use change may therefore control air quality via changes in direct removal of surface O_3. Simulated dry deposition changes in response to urbanization and biofuel plantation scenarios have been shown to be small, particularly when compared to related changes in BVOC emissions (see Section 2.1.3).[120, 122, 128] Ward et al. [17] suggest that historical land use change has led to a only a small (6%) increase in the dry deposition of O_3. Ganzeveld and Lelieveld [112] use a single column model to estimate

that deforestation may reduce daytime ozone deposition velocities by 60-70%, but lead to only small net changes in surface O_3 (< 2 ppb), given compensating increases in dry deposition at night. A reduction in BVOC emissions over converted agricultural regions, can compensate for direct decreases in O_3 dry deposition velocities (due to decreases in LAI and turbulent transport), producing little change in O_3 deposition flux.[50] Yet, Sigler et al. [129] measure substantially higher surface O_3 concentrations (20 ppb) over a pasture site in the Brazilian Amazon compared to a local forest site (6 ppb), consistent with their measured 3-fold decrease in O_3 deposition velocity at the pasture. They suggest that conversion of the Amazonian rainforests may lead to a substantial net reduction in the ozone sink, and rising surface O_3. Alternatively, Wu et al.[86] show how the incursion of broadleaf trees into the temperate and boreal forests (as well as CO_2 fertilization) through the 21st century can lead to large increases in ozone deposition, reducing mid-latitude summer surface ozone by up to 10 ppbv. Thus, while the sensitivity of O_3 to the changing vegetative landscape is primarily driven by BVOC emission changes (see Section 2.1), large deforestation or afforestation activities may also play a secondary role in controlling surface O_3 through dry deposition.

2.3 Ammonia

2.3.1 Introduction

Ammonia (NH_3) is the most important base in the atmosphere and contributes to the formation of inorganic particulate matter. In particular, atmospheric ammonia drives the conversion of gas-phase nitric acid to particle-phase nitrate, which has an estimated present-day radiative forcing of -0.05 to -0.12 Wm^{-2}.[51, 130-132] Reducing ammonia has been shown to be an effective air quality strategy for reducing particulate matter concentrations, particularly in winter.[133-135] Global ammonia emissions are dominated by agricultural activities, with livestock waste providing 39% of the source, and agricultural fields and fertilizer use representing a further 24%.[136] Application of fertilizer is an extremely cost effective means of increasing crop yields.[137, 138] It is estimated that half of the world's population is currently supported by the industrial production of synthetic ammonia fertilizer via the Haber-Bosch process.[139] However, losses

of nitrogen through leaching, volatilization and nitrification and denitrification, reduces the amount of nitrogen converted to plant matter and accelerates the delivery of nitrogen to the atmosphere and water bodies.[140] These losses vary with time of day and season, and are influenced by climatic conditions.[137, 141, 142] Erisman et al. [138] estimate that only 5-15% of fertilizer nitrogen reaches humans, with the majority of the remainder lost to the environment.

Recent decades have seen a rapid increase in synthetic nitrogen fertilizer production,[138] with a 5-fold increase in ammonia emissions since pre-industrial times.[139, 143] Modest reductions in ammonia emissions can be achieved via agricultural management; for example switching from urea fertilization (10-20% nitrogen losses) to ammonium nitrate fertilizer which has higher nitrogen use efficiency (1-2% losses).[138] Furthermore, excess application of fertilizer may be widespread, with several studies suggesting that in particular regions, nutritional requirements could be met by reductions in fertilizer use ranging from 20 to 60%, with little to no change in yields.[144, 145] Cowell and Apsimon [146] suggest an average 30% maximum feasible reduction for ammonia emissions in Europe. Successful agricultural management, largely aimed at reducing the evaporation of ammonia, led to a 9% reduction of ammonia emissions in Europe from 1990 to 2002.[147]

2.3.2 Land Use Change Effects on Ammonia

The trajectory of future ammonia emissions and the resulting air quality and climate impacts are tied to global fertilizer usage and the demand for meat. Global food demand is expected to increase by more than 50% from 2010 to 2050 as a result of population growth and the "westernization" of diets in developing countries.[148] This escalating pressure on food production may trigger the expansion of croplands and pasturelands, particularly in developing countries.[149] However, increased demand can also be met through agricultural intensification, as seen in the United States where total farmland decreased by 20% in the latter half of the 20th century, while nitrogen inputs more than doubled.[145] Erisman et al. [139] estimate that global improved nitrogen use efficiency (up to 50%) will compensate somewhat for increasing food demand, and project an overall 50% increase in ammonia emissions over the 21st century.

However they also suggest that increasing demand for bioenergy crops may lead to a doubling of fertilizer ammonia emissions. Bouwman et al.[150] confirm that that even the most optimistic agricultural efficiency scenarios will not lead to ammonia emissions reductions in the coming decades. As a result future RCP emissions scenarios project a 28 to 105% increase in agricultural ammonia emissions from 2000 to 2100,[151] although these scenarios may not capture the full uncertainty range.[152]

The rise in agricultural ammonia emissions is projected to offset reductions in anthropogenic NOx emissions, producing little change in overall nitrogen deposition.[153, 154] Similarly, Pye et al. [155] find that some of the projected decrease in nitrate concentrations due to domestic NOx emissions reductions over the United States is offset by ammonia emissions increases. Bauer et al. [130] suggest that a 46% increase in anthropogenic ammonia emissions alone in 2030 would lead to a 7% increase in the global nitrate burden. Finally, nitrogen application has been shown to decrease biodiversity,[138] and an increasingly fertilized biosphere may therefore be characterized by altered vegetation, with implications for BVOC emissions (see Section 2.1).

2.4 Nitrogen Oxides

2.4.1 Introduction

Both fire and land use perturbation of soils can alter nitrogen oxide (NO_x) emissions to the atmosphere, with implications for the secondary formation of O_3 and aerosols.

Fires contribute 15% of the global NO_x emissions to the atmosphere,[156, 157] enhancing tropospheric ozone concentrations.[158-160] NO_x emissions depend on the amount of biomass consumed by the fire as well as fuel burn conditions and nitrogen content of fuel.[161]

Microbial transformations in soils are responsible for 10-20% of the global source of nitrogen oxides to the atmosphere, with above-canopy source estimates ranging from 5-13 $TgNyr^{-1}$ (see Hudman et al. [162] and references therein). Nitrification and denitrification processes produce nitric oxide (NO) which diffuses through soils into the atmosphere. The emission of NO is governed by nitrogen availability, as well as soil properties including temperature, moisture and porosity.[163] "Pulses" of NO emissions can

occur after rain events re-activate nitrifying bacteria in dry soils where inorganic nitrogen has accumulated.[164, 165] For example, soil-atmosphere exchange of NO in semi-arid Australia was shown to increase by a factor of 5-9 after watering.[166] Croplands exhibit some of the highest NO emission rates,[167, 168] where low basal emissions are compensated by additional N availability through fertilizer use.[169] Veldkamp et al. [170] find a more than 5-fold increase in NO emissions when pasturelands are fertilized. Over a growing season, 1-10% of fertilizer nitrogen is lost as NO to the atmosphere.[168] Globally fertilizer use is estimated to contribute up to a third of NO soil emissions,[168] with more than half of the global fertilizer nitrogen applied to crops in Asia.[171] Veldkamp et al [170] find that NO emissions are more sensitive to soil water content than fertilizer composition. Viable agricultural management strategies could reduce global soil NO emissions by 4%,[172] however Smith et al. [173] suggest that these gains will be insignificant compared to the increases expected through global increases in fertilizer use (see Section 2.3). Soil NO emissions are also projected to increase under a warming climate.[155, 174, 175]

The NO emitted from soils is rapidly converted to NO_2 which can absorb to leaves, with roughly 50% of soil NO_x lost to the vegetation canopy.[168] Once in the atmosphere this NO_x participates in O_3 formation, often with high efficiency given the lack of anthropogenic NO_x sources over rural agricultural or forested landscapes. Soils may be the dominant natural source of NO_x contributing to background ozone,[176] the effect of which may have been underestimated according to satellite observations. [157, 177] Using a source of 9.7 $TgNyr^{-1}$, Steinkamp et al. [178] estimate that 5% of global lower tropospheric ozone can be attributed to soil NOx emissions. Williams et al. [179] estimate that 3% of the tropospheric ozone burden over the African continent can be attributed to soil NO_x emissions. Soil NO_x can also be a precursor to aerosol nitrate. Thus soil NOx emissions can contribute to air quality degradation through both O_3 and PM formation, and may contribute to climate forcing via these same pollutants. Shindell et al. [180] show how decreasing pre-industrial soil NO_x emissions (to account for the lack of fertilizer use), increases the estimate of present-day O_3 radiative forcing. In addition, soil NO_x leads to substantial increases in global OH, decreasing the methane lifetime by ~10%.[178]

2.4.2 Land Use Change Effects on Nitrogen Oxide Emissions

Land use change will alter fire NO_x emissions primarily through changing the amount of fire. We review the impacts of humans and land-use change on fire amount in section 2.5.1. Additionally, fire NO_x emissions are sensitive to the nitrogen content of fuel and fuel burn conditions [161] both of which may be altered by LUC. Castellanos, et al. [181] used satellite observations to show variability of NO_x emission factor according to fire type, ranging from low NO_x emission factors for deforestation to high emission factors for agricultural fires.

Land use change is likely to alter global soil NO_x emissions through either changes in vegetation types (deforestation, afforestation, succession, etc), or fertilizer use. Yienger and Levy [168] predicted a 25% increase in soil NO_x emissions over 3 decades (1995-2025) due to increased fertilizer use. Oil palm plantations in Borneo emit 5 times more NO_x than neighboring rainforest, although this includes contributions from vehicle emissions and processing facilities as well as fertilizer use.[115] Weitz et al. [182] report a 4-fold increase in NO emissions when forests were cleared to unfertilized agricultural areas, with short-term increases of more than a factor of 10. Ganzeveld and Lelieveld [112] estimate that conversion of tropical forests to pasturelands leads to a doubling of soil NO_x emissions over deforested regions in the Amazon, due to both increases in fertilizer application, and decreases in canopy uptake. Similarly Hardacre et al.[117] show that vegetation reductions through deforestation lead to larger soil release of NO in South America. Overall, cropland expansion, the intensification of agricultural activities, and climate change are projected to increase global soil NO_x emissions by 9% by 2050.[50]

2.5 Smoke

2.5.1 Introduction

Fire causes substantial perturbations to the Earth system through changes to vegetation, atmospheric composition and climate.[43] Fire is the largest source of carbonaceous aerosol to the atmosphere [143, 183] with global emissions of organic carbon (OC) and black carbon (BC) estimated as 23-31 Tg C yr^{-1} and 2.2-3.1 Tg C yr^{-1} respectively.[47, 184] At the global scale, emissions are dominated by

Africa which constitutes >50% of global carbon emissions from fire.[184] Smoke from fires can dominate PM variability in both remote [185, 186] and polluted urban regions [187] and fire PM has been linked to poor air quality and increased human mortality.[188, 189] At the global scale smoke emissions from fires are estimated to result in 260 000 to 600 000 premature deaths each year.[190]

Fire emissions are typically estimated using satellite data of fire occurrence or area burned combined with estimates of fuel load and laboratory emission factors.[184, 191] Satellite sensors have issues spotting small fires, implying that emissions in some regions may be underestimated.[192] Furthermore, atmospheric ageing may increase PM and aerosol number concentration after emission,[193, 194] although some studies report little SOA formation in smoke plumes.[195] Both underprediction of small fires and atmospheric formation of PM may explain why some studies scale fire emissions by a factor ~2-5 to match observed aerosol optical depth.[196-199]

Globally, the top-of-atmosphere (TOA) direct radiative effect from fire aerosol is estimated to be close to zero since warming effects from BC are cancelled by cooling effects of OC.[200] The DRE from smoke aerosol is sensitive to physical [201] and optical [202] properties, which have been previously reviewed. Estimates of the global TOA DRE of fires (present day fire compared to no fire) range from -0.01 Wm^{-2} [203] to -0.19 Wm^{-2}.[51] Calculating the DRF due to fire requires estimation of the amount of fire in the pre-industrial, with TOA DRF (1750-2100) estimated to be 0 Wm^{-2} (-0.2 to 0.2 Wm^{-2}).[200] The OC component of fire aerosol, which was typically thought to only scatter light, may also have important absorption properties,[204, 205] increasing the warming potential of fire aerosol. There are fewer studies of the global aerosol indirect effect from fire, with estimates from -0.09 Wm^{-2} [203] to -1.64 Wm^{-2}.[199] Deposition of aerosol from fires onto snow and ice can enhance melting rates.[206] The regional impacts of fire aerosol on climate and weather can be substantial, causing large regional TOA DRE [207, 208] and reductions in surface radiation, and potentially altering cloud formation, [207, 209] dry season duration,[210] and precipitation patterns.[211]

2.5.2 Land Use Change Effects on Smoke Emissions

The impact of human activity and land-use change on fire is complex. Human activity increases fire through accidental and intentional fire ignition, but decreases fire directly through fire suppression and indirectly through altering vegetation through land-use change and wood harvest. Regional studies have shown humans increase fire in Russian boreal forests [212] and moist tropical forests,[213, 214] but decrease fire in arid regions.[34] At the global scale, Knorr, et al. [215] used a multi-year satellite record of fire to show that increasing human population decreases fire frequency, except at very low population densities (<0.1 persons km^2) where fire is increased by 10-20% compared to a situation with no humans. A number of historical fire projections have accounted for human impacts, often applying empirical relationships between population density and fire.[216-221] Kloster, et al. [220] used a global fire model to estimate that humans have decreased global fire carbon emissions from 1850 to present day by 11%; this was the net effect of increased emissions due to deforestation fires combined with decreased emissions due to LUC and wood harvest reducing above ground biomass. Knorr, et al. [215] estimated that global area burned has declined by 14% since 1800. Yang, et al. [217] predicted that humans have had a larger impact, reducing area burned by fire by ~30% from the 1900s to present day.

Long-term variability of fire can be explored through sedimentary charcoal records and from air trapped in ice, providing insight to fire emission drivers.[222-224] Marlon et al. [223] used analysis of sedimentary charcoal to suggest that global biomass burning declined from AD1 to ~1750, potentially driven by a global cooling trend, increased sharply between 1750 and 1870 due to deforestation fires, before declining from 1870 to present day due to fire management and agricultural intensification. Wang et al. [224] used a 650-year Antarctic ice core record to suggest a 70% decline in fire emissions from 1800s to present day, although the magnitude of this decline has been questioned.[225] In contrast, observations of CH_4 in ice suggest a minimum on fire activity around 1800 with a peak in fire activity in the present day.[226, 227]

Whilst the overall impact of humans on global fire emissions is not clear, particulate fire emission inventories typically assume greater fire emissions in the present day compared to the preindustrial.[143, 183] This is because these studies assume a positive relationship between fire emissions and population

density. Dentener et al. [183] estimated preindustrial fire emissions by scaling present day emissions according to human population (except in boreal forest regions where present day fire was assumed to be less than natural due to fire suppression). Using these assumptions they predict that emissions in the year 2000 were a factor ~3 of those in year 1750. Lamarque et al. [143] constructed decadal mean fire emissions from 1850 to 2000, predicting that fire emissions declined from 1900 to 1950 before increasing to the year 2000, with an overall 30% increase from 1850 to 2000. A number of global fire emissions datasets also predict that fire emissions have increased over the 20th Century, primarily due to increased deforestation in tropical regions.[156, 228, 229] Schultz et al. [156] estimate that global BC and OC fire emissions have increased by 60% and 68% respectively over the period 1960 to 2000. Mieville et al. [229] estimate that global BC emissions from fire declined slightly from 1900 to 1960, before increasing until the 2000s, with emissions in the 1990s 18% greater than in 1900. Ito and Penner [228] estimate that BC emissions have increased by ~60% and OC emissions by ~160% from 1870 to 2000. Historical fire PM emission estimates are therefore not consistent with paleo-records or recent understanding of the impact of humans on fire.

In regions of deforestation, humans have led to increased fire emissions [47, 156] resulting in greatly increased aerosol concentrations.[230, 231] In the present day, deforestation and degradation fires are largely confined to the tropics and are estimated to be responsible for ~20% of global fire carbon emissions.[184] In both the Amazon [213] and SE Asia,[232] the occurrence of fire is increased in regions of deforestation and amplified in years of drought. Nepstad et al. [233] showed that over the Brazilian Amazon fire was 4-9 times more likely outside government protected areas compared to inside. Koren et al. [234] reported increases in deforestation, fire counts and AOD over the Amazon during the period 1998 to 2005. However, recent declines in the rate of deforestation in Brazil have not resulted in commensurate declines in fire, likely due to the increased use of fire in agricultural land management.[235] In Africa, land-use change within savanna regions has led to a reduction in burnt area of 8×10^5 ha year^{-1}, equivalent to 0.4% of area burned.[236]

In regions of high fire risk, forest management and prescribed burns are used to reduce wildfire.[237] Prescribed burning involves the use of controlled fires to reduce surface fuel loads and the potential for future large and uncontrollable wildfires. Wiedinmyer and Hurteau [238] found that prescribed fire can reduce regional carbon fire emissions for the western US by 18-25% with reductions of up to 60% in specific ecosystems. The PM emissions from prescribed fire may also differ from wildfire [239] and the overall impacts of widespread use of prescribed burning on regional PM is not well known. Complex interactions between insect infestation and fire are likely,[240] but the overall impact on fire amount is uncertain and likely to be dependent on ecosystem type and time since pest outbreak.[241, 242]

Many projections of future fire simulate the impacts of climate change but do not include the impacts of anthropogenic land-use change.[243] Future climate change is expected to lead to an overall increase in fire during the 21st Century, although large regional variations are likely.[243] Regional reductions in fire are predicted in forested regions where precipitation is projected to increase.[218, 222, 243] In contrast, over savannah regions of Africa, increased precipitation in the future may lead to increased vegetation growth, greater fuel loads and increased fire.[219] Recently, a few studies have attempted to simulate the combined impacts of anthropogenic fire ignition, suppression and land-use change in addition to climate change on global fire amount. Pechony and Shindell [218] predict that climate change during the 21st Century will lead to increased fire emissions, partially offset by reductions in fire due to increased human population driving fire suppression and land-use change reducing vegetation cover. Kloster et al. [244] used a coupled fire model, including description of the impacts of climate, demography, and land-use change on fire, to predict that global fire carbon emissions will increase by 17-62% in 2075-2099 relative to 1985-2009. They predict changes in climate will increase fire emissions by 22-66%, whereas increased human population will decrease fire emissions by 6% (accounting for both anthropogenic fire emission and suppression) and forest harvest and land-use change will decrease fire emissions by 5-35%. Emissions from deforestation fire are lower in all RCP scenarios during 2075-2100 compared to the present day due to projections of reduced rates of LUC. In addition, projections of

increased wood harvest lead to a reduction in biomass available for burning and hence reduced emissions.[244]

To date there have been no studies of how both climate and land-use change will alter aerosol through changes in fire. A few studies have assessed the impacts of climate-driven changes to fire on atmospheric aerosol.[224, 245] Spracklen et al. [245] predicted that climate change (A1B scenario) will cause a 54% increase in wildfire area burned in the western United States by 2050 relative to present day (1996-2005), increasing summertime OC by 40% and summertime BC concentrations by 20%. Wang et al. [224] used ensemble projections from 15 climate models to predict a 46-70% increase in OC and a 20-27% increase in BC by 2050 relative to present day. Carvalho et al. [246] predict that climate change (A2 scenario) will increase July PM10 concentrations in 2100 over parts of Portugal by up to 40% (20 μg m^{-3}) relative to 2005, but with only a small contribution due to increased fire emissions.

2.6 Dust

2.6.1 Introduction

Mineral dust represents the largest terrestrial source of particulate matter to the atmosphere. Once in the atmosphere, dust degrades air quality and visibility,[247] supplies a source of nutrients to remote oceans and ecosystems,[248, 249] provides a surface for the heterogeneous reaction of trace gases,[250, 251] and impacts the Earth's radiative budget.[252] The global mean all-sky direct radiative effect of dust has been estimated at -0.26 Wm^{-2}; assuming that 20% of present-day is of anthropogenic origin, the associated direct radiative forcing of dust is -0.05 Wm^{-2}. [51] The most recent IPCC estimates of the radiative forcing from mineral dust is -0.10 Wm^{-2}, with a range of -0.30 to +0.10 Wm^{-2}, but which is not entirely of anthropogenic origin.[200]

Most major dust sources are topographic depressions in arid or semi-arid regions. Many such regions are linked to historic hydrological features (e.g. ephemeral or desiccated lakes),[253] with over 90% of the global dust source is located in the Northern Hemisphere.[254]

2.6.2 Land Use Change Effects on Dust

Dust fluxes can be modulated by both climate (by altering dust generation through wind speed and soil moisture) and land use. Vegetation typically increases both the stability and moisture level of soils, reducing dust flux.[255, 256] Reductions in vegetation due to both climate change and land use (e.g. excessive grazing) can increase dust emissions. Human activity can play a dramatic role in the generation of new dust regions; for example the anthropogenically dessicated playa of Owens Lake in Southern California which resulted from water diversions in the early 20th century to support urban growth in the region.[257, 258] Similarly, the "dust bowl" of the 1930s and 1950s in the United States occurred in a region where the soil was disturbed by cultivation, which has been shown to increase the erodibility of soils.[259] In general, cultivation, overgrazing, water diversion, and deforestation can all lead to the development or intensification of dust sources. This type of anthropogenic land degradation has been termed "desertification" by UNEP.[252]

A key question is what fraction of global dust sources are the result of human activities? Natural and anthropogenically generated dust are chemically and physically identical, therefore efforts to assess this split rely on geographical features. For example, anthropogenic dust sources are typically located at the semi-arid desert edge.[260] Tegen and Fung [260] provided the first estimate of this fraction when they found that model simulations where human disturbed sources accounted for 30-50% of total atmospheric dust loading provided the best match with AVHRR observations of aerosol optical thickness. Based on this, Tegen et al.[261] estimated that anthropogenic dust contributes a surface forcing of -1 Wm^{-2}, but a top-of-atmosphere (TOA) forcing of only +0.09 Wm^{-2}, due to the offsetting effects of dust on solar and thermal radiation. Sokolik and Toon [252] estimated that a smaller fraction of global dust is anthropogenic (20-30%). Prospero et al [262] also suggested that the Tegen and Fung estimate was too high, given that the strongest dust sources are in regions that cannot support agricultural and grazing activities and are largely uninhabitable. However Luo et al. [263] showed how currently available observations could not discriminate between models with 0 or 50% of anthropogenic dust, given the large uncertainties in meteorology and

dust parameterization. A related estimate that 14-60% of current dust emissions are anthropogenic highlights this increasing uncertainty.[264] In contrast, in a subsequent analysis of dust storm frequency observations, Tegen et al.[265] estimated that only 5-7% of dust comes from agricultural areas. However Mahowald et al. [266] showed that an alternate analysis of the same dataset could also be consistent with their previous estimates of 0-50%. Yoshioka et al. [267] find that a lower fraction of dust over North Africa is anthropogenic (0-25%). This is supported by the most comprehensive estimate to date from Ginoux et al. [253], who developed a high resolution mapping of dust sources based on MODIS AOD observations and land use datasets. Globally, they estimate that anthropogenic sources account for 25% of present-day dust, but demonstrate significant regional variation. In particular, while they find that only 8% of dust in North Africa is anthropogenic, over 75% of dust sources in Australia can be attributed to human activity. We refer the reader to Ginoux et al [253] for a thorough regional description of anthropogenic dust sources.

It remains challenging to assess the dust potential of currently stable soils and the formation of new dust source regions, and then to attribute these changes to climate or land use change. The doubling of dust over the 20th century observed in Antarctic ice cores has been linked to desertification over South America, possibly due to climate or alternatively to overgrazing and poor land use practices.[268] Mahowald et al. [269] find that paleoproxies support a doubling of dust over much of the globe during the 20th century. The increase in dust observed from North Africa over the 1960s-1980s has been attributed to both decreasing precipitation [270] and the intensification of human habitation in the Sahel.[271] Cowie et al. [272] suggest that the late 20th century down-turn in North African dust can be attributed to increasing vegetation in the Sahel; however Ridley et al. [273] show that these trends are consistent with a large-scale stilling of the winds, primarily over the Sahara. Ward et al. [17] suggest that land use change alone drove an 18% increase in dust emissions from pre-industrial to present-day, and predict a further 5-10% increase over the 21st century. Mahowald and Luo [264] predict that changes in vegetation and climate from pre-industrial to 2090 will decrease the global dust source by 40%. This decrease is tempered (17%) when including the opposing role of human cultivation. Finally, urbanization and the resulting co-location of

dust sources with anthropogenic pollution may alter the chemical composition, properties and impacts of dust.

2.7 Terrestrial Primary Biological Aerosol Particles

Primary biological aerosol particles (PBAP) are particles emitted from the biosphere including pollen, fungal spores, bacteria and vegetation debris. PBAP may contribute a substantial fraction of supermicron organic aerosol,[274] as well as impacting CCN [275] and ice nuclei (IN) [276] [277]concentrations. PBAP often dominates supermicron aerosol mass over forested regions,[230, 278] with concentrations as great as 7 μg m^{-3} measured during the wet season over the Amazon.[279] A recent review of PBAP was conducted by Despres et al. [280]

Global emissions of terrestrial PBAP of up to 1000 Tg yr^{-1} have been estimated,[274] consisting of 8-168 Tg yr^{-1} fungal spores, 0.4-28.1 Tg yr^{-1} bacteria and 47-84 Tg yr^{-1} of pollen.[280] Emission estimates are typically dependent on both ecosystem and meteorological variables. For example, Heald and Spracklen [281] parameterise fungal spore emissions based on LAI and atmospheric moisture.

To our knowledge there have been no studies of the impact of land-use change on PBAP emissions, with progress limited by our poor understanding of PBAP emission drivers. If emissions are related to vegetation density [281, 282] then deforestation is likely to reduce PBAP emissions. In contrast, Burrow et al. [283] report higher bacteria concentrations over grasslands and croplands compared to forests, suggesting that conversion of forest to cropland may increase bacteria emissions. MacKenzie et al. [284] suggest that conversion of tropical forest to oil palm would change PBAP emissions, but they did not observe PBAP over an oil palm landscape.

3. Global Radiative Effects of Land Use Change via Short Lived Pollutants

The IPCC estimates that the radiative forcing (RF) of short-lived pollutants from pre-industrial to present-day includes +0.4 Wm^{-2} of warming from tropospheric ozone (range given as +0.2 to +0.6 Wm^{-2}),

and –0.35 Wm^{-2} of cooling from the direct radiative forcing of particulate matter (range given as -0.85 to +0.15 Wm^{-2}).[200] An additional cooling of –0.9 Wm^{-2} (range of -1.9 to -0.1 Wm^{-2}) is associated with the aerosol indirect effects. The RF, by definition, incorporates the effect of anthropogenic land use change, but given the scarcity of studies which assess this affect, typically represents only the change in anthropogenic emissions (e.g. LUC was not included in the ACCMIP simulations [285]). Here we estimate how LUC (both natural and anthropogenic) modifies the climate impact of short-lived pollutants.

Table 4 details the radiative effects of LUC via short lived pollutants both for the pre-industrial (variously defined as1750 to 1850 in the literature) to present day and for the 21st century based on the information reviewed in Section 2. For aerosols we estimate the DRE due to land use change by combining present day DRE with projected changes in emissions due to LUC (see Table 2 and Table 4 footnotes for details). We neglect the contribution from PBAP given the lack of information on the global budget, radiative impacts, and projected change in this aerosol type. For fire PM we assume emissions scale with either area burned or fire carbon emission, since detailed projections of emissions are not available. For nitrate aerosol, we assume that DRE scales with emissions of ammonia, although this presumes that emissions of these are always limiting for the formation of ammonium nitrate (see footnotes for details). We restrict this assessment to the direct effect, since the DRE scales linearly with emissions and PM burden (e.g., [65]) which is not the case for the aerosol indirect effect. Given the non-linearities in ozone formation, we cannot follow a similar procedure of linking changes in precursor emissions with the radiative impacts. Instead we cite the one study by Unger[110] who estimate that anthropogenic LUC from 1850 implies a RE of -0.11 Wm^{-2} associated with tropospheric ozone, via changes in BVOC emissions alone. This estimate assumes present day anthropogenic pollution emissions in 1850, and also neglects the impact of changes to soil NO_x emissions and dry deposition to vegetation, but provides the best estimate to date from the literature.

Figure 2 shows our estimates of the RE changes in tropospheric O_3 and PM driven by LUC. Over the PI to PD, we estimate that LUC has caused a total cooling from short lived pollutants of -0.21 Wm^{-2} (range of -0.28 and -0.14 Wm^{-2}). Ward et al. [17] suggest that the LUC impact on tropospheric ozone is a

warming (+0.12 Wm^{-2}) primarily due to the effect of wildfire and changes in methane on O_3, effects that we do not include in our estimate; this is in contrast to the BVOC-driven O_3 cooling cited here from Unger[110] (-0.11 Wm^{-2}). The LUC aerosol DRE is -0.171 to -0.031 Wm^{-2}, equivalent to 10-50% of the IPCC estimate for net aerosol DRF cooling due to anthropogenic pollution. The LUC-driven aerosol cooling is driven by increases in ammonia and dust emissions partially offset by a warming due to decreases in BSOA and smoke aerosol. Ward et al. [17] estimate that the LUC aerosol DRE is nearly neutral (-0.03 Wm^{-2}), but do not provide speciated values to compare to our estimates. We note that our estimate of both the anthropogenic and natural LUC impacts on BSOA (+0.03 Wm^{-2}) is considerably more modest than suggested by Unger[110] for anthropogenic LUC alone (+0.09 Wm^{-2}). Their value is approximately half of the total DRE of BSOA estimated by Scott et al.,[67] suggesting that their simulations have substantially higher abundance or cooling efficiency of BSOA. The ranges shown in Figure 2 are based on the range of literature values and almost certainly underestimate the true uncertainties on these estimates. In particular, we do not include any estimate of the uncertainty on the historical LUC-driven cooling from O_3, as we base this value on only one study. We linearly scale global nitrate changes to ammonia emissions, an obvious oversimplification which may overstate the impact of ammonia on the PM DRE. Finally, the uncertainty in the DRE of fire aerosol, as well as contradictory attribution of human impact on the trajectory of emissions (see Section 2.5.2), translates to substantial uncertainty on the impact of smoke.

We are unable to estimate the aerosol indirect effect due to LUC because this effect is strongly non-linear.[286] Many estimates of the anthropogenic aerosol indirect effect assume present day sources of natural aerosol for their preindustrial simulations. This assumption may be violated by land use change. Different natural aerosol emissions in the preindustrial, will alter the natural aerosol baseline, altering the calculated aerosol indirect RF which is sensitive to this baseline.[14, 15]

Table 4 also shows the further change in DRE anticipated with 21st century LUC. It is critical to note that these results reflect existing literature estimates, and do not comprehensively characterize potential land use change effects suggested by Figure 1.Given a lack of literature on the topic, we do not include an estimate for future LUC-driven changes in O_3; Ward et al. [17] suggest that the next century will produce

relatively modest changes in O_3 forcing (-0.06 to +0.03 Wm^{-2}) compared to the 20th century. We estimate that while the potential impact of LUC on aerosol DRE from 2000 to 2100 remains a cooling, it is more modest (-0.042 Wm^{-2}) and more uncertain (-0.177 to +0.045 Wm^{-2}) than from PI to PD, with a larger possibility of warming included in our range. Ward et al. [17] project modest net decreases in aerosol cooling by 2100 (+0.01 to +0.04 Wm^{-2}).Our projections continue along the 20th century trajectories, but with the exception of smoke, are more muted. The 21st century projections are dominated by the offsetting effects of potential growth in agricultural emissions of ammonia (and the resulting nitrate cooling) and the decrease in smoke emissions driven by human fire suppression (and the resulting warming).

4. Local Land Use Change Scenarios

The estimated impacts of LUC on global mean DRE ignores the potential for more substantial regional changes in climate driven by LUC. Figure 3 illustrates the regional aerosol DRE from four idealized LUC scenarios: i) tropical deforestation and replacement by agriculture (soybean); ii) tropical deforestation and replacement by oil palm; (iii) boreal forest fire followed by natural succession, (iv) temperate hardwood deforestation and replacement by agriculture and pine plantation. We focus here on aerosols; the complex response of ozone to changes in emissions and deposition at various spatial scales is not easily captured in these idealized scenarios. We estimate changes in aerosol sources based on literature values for emission factors, biomass density and BSOA yields, assume a 5 day atmospheric lifetime, and apply median AeroCom radiative efficiencies from Myhre et al. [131] to convert these changes to aerosol DRE (see Tables S1 and S2 for details). LUC induced soil and fire sources of NO_x may impact nitrate levels, but these effects depend on the ammonium nitrate formation potential of the local chemical environment and are likely small. A full assessment of the DRE would require detailed numerical simulations; our scenarios are intended to illustrate the potential for LUC to cause regional radiative perturbations through short lived atmospheric pollutants on the 1-40 year time horizon.

We estimate that LUC can cause regional DRE changes as large as ±20 Wm^{-2}. Natural forest ecosystems typically induce a negative DRE, with aerosol cooling dominated by SOA from BVOCs. The magnitude of cooling depends on the assumed vegetation phenology, BVOC emission factor and SOA yields: with a DRE of -4 Wm^{-2} typical of broadleaf forests. In these scenarios, we assume deforestation is associated with/driven by fire and we simulate large DRE changes due to smoke emissions. The net DRE of fire emissions depends on assumptions about BC:OC ratios of the emissions. Pasture maintenance fires emit a higher BC:OC ratio compared to deforestation fires,[287] producing more relative warming as deforestation progresses and the fraction of pasture fires increase. At the same time, the amount of biomass burned per year declines as land is cleared and adapted for agriculture. Soybean crops consume almost no nitrogen fertilizer,[288] thus no additional ammonia source is implied in this scenario. Overall, we find that deforestation typically results in a net warming due to a reduction in BVOC emissions and BSOA, combined with an increase in warming from pasture maintenance fires (Figure 3a,b). Interestingly, for development of oil palm on SE Asian rainforests, we find the magnitude of warming depends crucially on the extent of crop residue burning, which we assume here is 50% of the biomass (Figure 3b). Increases in BSOA associated with oil palm (see Section 2) are less important, because isoprene emissions are thought to be lower in Asian tropical forests, compared to the Amazon for example. We do however neglect here the impact of NO_x emissions changes which accompany plantation developments and which may influence the rate and yield of BSOA formation. Unlike the warming from tropical fires, boreal forest fire results in a strong pulse of cooling due to high OC:BC emission ratios (Figure 3c). Over the longer term the net radiative effect of boreal fires is driven by changes in BVOC emissions due to vegetative succession, with a large reduction in emissions after the fire followed by a return to the pre-fire state over a period of 70+ years. We find that the combination of emissions and BSOA yields produce relatively similar BSOA source potentials over both deciduous and needleleaf boreal trees, suggesting that aerosol loading and radiative effects return to pre-fire levels ~20 years after the fire. The net DRE of deforestation over temperate regions is sensitive to agricultural practices, in particular the extent of fertilizer application and agricultural waste burning (Figure 3d). There is wide

variation in fertilizer application rates and the resulting ammonia emissions; for our mid-latitude clearing scenario we use an ammonia emission of 1 $gm^{-2}yr^{-1}$, and assume that 64% of this ammonia is converted to ammonium nitrate based on the particle to gas deposition ratios from Zhang et al. [289] The extent of agricultural waste burning likely varies substantially between regions; in countries where policies have been introduced to limit burning (e.g., U.K.) the net RE of this LUC scenario may be a cooling.

These scenarios describe the significant, and poorly understood potential for land use change to modulate the regional radiative balance via perturbations to aerosols. The net effect often represents offsetting cooling and warming effects, with high uncertainty and regional variation in emissions associated with different vegetation. In particular, we note that many of these scenarios result in a net positive DRE whereas the global estimates of Fig. 2 suggest a net cooling from LUC associated with aerosols.

5. Conclusions

Both natural and anthropogenic land use change may substantially impact global air quality, with significant radiative effects on global and local climate. Here we review the existing literature on how these landscape changes affect short-lived air pollutants (ozone and aerosols). In particular, we focus on how historical and projected land use change will alter emissions of BVOC, soil NO_x, dust, smoke, and bioaerosol, as well as the dry deposition of ozone. These constitute the key drivers of air quality changes related to a dynamic land surface.

While these impacts are not inconsiderable, there has been limited research to date on how land use change has or will impact overall air quality. Our synthesis of diverse studies suggests that land use change has led to an overall cooling over the 20th century, with aerosol affects alone equivalent to 10-50% of the aerosol direct radiative forcing due to anthropogenic emissions. This cooling is likely to continue through the 21st century, but is subject to large uncertainties associated with future agricultural practices (including: fertilizer usage, desertification, deforestation, biofuel development). Furthermore, the vast majority of the studies reviewed neglect the connections between different species or process responses to

land use change. For example, CO_2 fertilization likely increases fuel availability for wildfires, enhancing smoke emissions over time. Or alternatively, deforestation is likely to affect surface ozone through fire emissions, changes in BVOC and soil NO_x emissions, as well as a reduction in deposition to the forest canopy. Examining these effects individually likely presents a biased and incomplete assessment of the overall impact of land use change.

In an era of declining pollution emissions, anthropogenic land use change may become the dominant human fingerprint on ozone and aerosol climate forcing. In addition, natural land use change represents a critical climate feedback which impacts both air quality and our assessment of how anthropogenic pollution has affected cloud formation, and the associated climate cooling. This review serves as a call to further investigate and quantify these effects on both local and global air quality and climate.

Acknowledgements

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Supporting Information Available

Tables describing the parameters used for the scenarios described in Section 4 are available in supporting information. This information is available free of charge via the Internet at http://pubs.acs.org/.

List of Captions

Table 1: Illustrative changes in land cover due to various modes of land use change.

Table 2: Estimated change in emissions due to land use change, from pre-industrial (PI, 1850), to present-day (PD) to future (F, 2100), based on literature reviewed in Section 2. To date, there are no literature estimates for the effect of land use change on PBAP.

Table 3: Simulated changes in BVOC emissions and air quality due to land use xhange.

Table 4: Changes in global mean direct radiative effect due to historical and future (2100) LUC. Best estimate shown first, with range in brackets. Where not explicitly described in the footnotes, changes in direct radiative effect due to LUC are estimated by multiplying emission changes from Table 2 to the aerosol DRE. Similarly, unless otherwise stated, best estimates are mean of given ranges.

Figure 1: Historical (dashed) and projected (colored by RCP scenario) global land use area for 3 major categories. Data from Lawrence et al. [48]

Figure 2: Changes in global mean d radiative effect (RE) due to (a) Historical (pre-industrial to present day) and (b) Future (2100) LUC. For aerosol RE is restricted to the direct radiative effect. See Table 4 for details.

Figure 3: Local Land Use Change Scenarios (left) with Estimated Impacts on Aerosol Sources (center) and Radiative Effects (right). For each scenario land use change starts in year 10. Details on calculations provided in supplementary materials.

TABLES

Table 1: Illustrative changes in land cover due to various modes of land use change.

Land Use Change Process	**Change in Land Cover (normalized per year)**	**Reference**
Fire	3-6 million km^2 yr^{-1}	[45-47]
Agricultural Conversion	45 million km^2 from PI to PD ($300,000$ km^2yr^{-1})	[19]
Insect Infestations (e.g. bark beetle infestation of North America)	$100,000$ km^2 over 10 years ($10,000$ km^2yr^{-1})	[42]
Biofuel Plantation (e.g. oil palm in Malaysia)	$42,890$ km^2 since 1960s (~ 1000 km^2yr^{-1})	[115]

Table 2: Estimated change in emissions due to land use change, from pre-industrial (PI), to present-day (PD) to future (F, 2100), based on literature reviewed in Section 2. To date, there are no literature estimates for the effect of land use change on PBAP. In the studies synthesized here, the reference year for PI ranges from 1750 to 1850.

Species	PI to PD	PD to F
Isoprene	-15% to -36%[a]	-24% to -27%[b]
Monoterpenes	-28% to 0[c]	0% to +12%[d]
Ammonia	+170%[e]	+28% to +105%[f]
Soil NO_x	+50%[g]	+60%[h]
Smoke (and Fire NOx)	-11% to -30%[i]	-11% to -41%[j]
Dust	+18% to +25%[k]	+5% to +10%[l]

[a] We estimate the net effect of historical LUC on isoprene emissions as a decrease of 15-36%, the sum of the impacts from anthropogenic land use changes and CO_2 increases (fertilization moderated by inhabitation) given separately in Unger et al. (2013) and Lathiere et al (2010).
[b] We estimate the net effect of future LUC on isoprene emissions as a decrease of 24 -27%, the sum of -12% from CO_2 increases (lower range of Arneth et al. (2008) with an additional -12 to -15% from anthropogenic LUC (Heald et al., 2008; Wu et al., 2012). We neglect possible biofuel plantation effects.
[c] We estimate that LUC may result in 0 to -28% change in monoterpene emissions based on the estimates of Acosta Navarro (2014) and Unger (2014).
[d] The effect of future LUC on monoterpene emissions is estimated to range from 0% to +12%, the sum of +10% from the CO_2 fertilization effect (Wu et al., 2011) and -10 to +2% from anthropogenic land use change (Heald et al., 2008; Wu et al., 2011).
[e] Given that 63% of present-day ammonia emissions are associated with agricultural sources; we attribute all of these to land use change from pre-industrial to present-day
[f] RCP projected emissions
[g] Yienger and Levy (1995) estimate that one third of global soil NOx is currently due to fertilizer use. Assuming this is entirely associated with historical land use change implies a 50% increase from PI to PD.
[h] We estimate a 60% increase in soil NOx emissions due to future LUC, the sum of a 50% increase due to fertilizer use (based on the 1995-2025 estimate of Yienger and Levy (2005)) and an additional 10% increase associated with deforestation and plantations.
[i] We estimate a range of -11% to -30% for the reduction in aerosol emissions from pre-industrial to present day based on Kloster et al. (2010) who estimate that LUC and wood harvest have reduced fire carbon emissions by 11% and Yang et al. (2014) who predict that human activity has reduced global area burned by 30% from 1900 to present day. We assume that aerosol emissions scale with carbon emissions or area burned.
[j] We estimate the net effect of human activity on future fire (2075-2099 compared to 1985-2009) as -11% to -41% from Kloster et al. (2012) through combining a -5% to -35% change in fire due to LUC and wood harvest (range driven by different RCP scenarios) with a -6% due to increased fire suppression.
[k] Estimate that dust has historically increased due to LUC by 18% (Ward et al., 2014) to 25% (Ginoux et al.,2012)
[l] Assume 5-10% increase in dust from present-day to future as in Ward et al. (2014)

Table 3: Simulated changes in BVOC emissions and air quality due to land use change

Type of Land Use Change	Change in Isoprene Emissions	Change in Monoterpene Emissions	Change in O_3[a]	Change in SOA Burden	Time Horizon (geographical extent)	Study
CO_2 Fertilization						
	+27%	-	-10 to +50 ppb	-	1990 to 2090 (Global)	84 ii
	+27%	+51%	-	-	1990s to 2100 (Global)	85
	+26 to +41%	-	-	-	1981-2000 vs 2081-2100 (Europe)	88 b
	+25%	+10%	> -10 ppb	+20%	2000 to 2100 (Global)	86 ii
	+17%	-	+3%	-	2000 to 2095 (Global)	87 ii
	+7 to +14 %	-	-	-	1981 to 2000 (Global)	83
CO_2 Inhibition						
	-32 to -52%	-		-	2081-2100 (Global)	92
	-31%	-	-	-	2100 (Global)	93
	-55%	-	-10 ppb to +10 ppb	-	2000 to 2090 (Global)	95
	-47%		1%		2100-2109 (Global)	96
	-26%		0%	-25%	2050	94
	-31%		-4%		2095 (Global)	87
	+11%				1800 (Global)	109
Anthropogenic LUC						
Anthropogenic LUC	-15%	-10%	-	-14%	2000 to 2100 (Global)	76
Anthropogenic LUC	-40%	-	-5%[i]	-	2000 to 2095 (Global)	87
Anthropogenic LUC	-42%	-	-	-	1981-2000 to 2081-2100 (Europe)	88
Anthropogenic LUC	-52%	-32%	-5 ppb to +5 ppb	-28% to -45%	1990-1999 to 2045-2054 (United States)	111 ii
Anthropogenic LUC	-15%	-	-	-	1901 to 2002 (Global)	79
Anthropogenic LUC	-21%	negligible			1750 to 2000 (Global)	109
Anthropogenic LUC	-25%	-28%	-	-	1850 to 2000 (Global)	110

[a] Changes in surface concentrations given in ppb, changes in tropospheric burdens given in percent
[b] Also includes effects of climate change

Anthropogenic LUC	-12%	+2%	< + 5ppb	-20%	2000 to 2100 (Global)	[86]
Anthropogenic LUC	-22%	-	-	-	1880 to 2000 (Global)	[97]
Anthropogenic LUC	-2%				1981 to 2000 (Global)	[83]
Deforestation	-30%	-	< 2 ppb	-	Deforestation (Amazonia)	[112]
Tropical Deforestation	-29%	-29%	-	-	Local scenarios (Global)	[113]
Urbanization/Pasture	-9%		-4 to +4 ppb		Local Conversion Scenarios for US and Amazon (Global)	[114]
Afforestation						
Succession + Afforestation + Plantation	+7%	-	-2 to +2 ppb	-	1980s to 1990s (US)	[108, 290]
European Afforestation	+4%	-0.1%	-	-	Local scenarios (Global)	[113]
Biofuel Scenarios						
Tree Plantations	+37%		< - 7ppb		Local Conversion Scenarios for US and Amazon (Global)	[114]
Biofuel (oil palm, tropics) Biofuel (SRC, midlatitudes)	+1% (global) +1% (global)	-	< 1% < 1%	< 1% < 1%	Local scenarios to replace 1% of fossil fuel demand in 2020	[120]
Biofuel (A. donax)	-	-	+0.52 ppb (OR) 2.46 ppb (TX) 3.97 ppb (IL)	Negligible (OR, TX) +44% (IL)	Coal Power Plant Replacement Scenarios (local: OR, TX, IL)	[121]
Biofuel (poplar)	+45%	-	+6% (peak O3) +25% SUM035 +40% AOD40f	-	Convert 5% of crop and grassland to plantations (Europe)	[122]
Biofuel (oil palm, Borneo)	-	-	-5 to +18 ppb (Borneo)	-	Complete Conversion (Global)	[119]
Biofuel (oil palm, Borneo)		-	< 15 ppb (Borneo)	-	Complete conversion (Global)	[118]
Mixed Scenarios						
CO_2 Fertilization + Anthropogenic LUC	-12%	-	< +9ppb	-	2000 to 2050 (Global)	[50]
CO_2 Fertilization + Anthropogenic LUC	+19% -8%	-	-	-	1860-1869 to 2000-2009 2000-2009 to 2100-2109 (Global)	[96]

CO_2 Fertilization +CO_2 Inhibition	-36 to -12%	-	-	-	1981-200 to 2081-2100 (Europe)	[88]
Anthropogenic LUC + Biofuel (SRC)	-1.7% to +1.4%	-	+5 to +12 ppb (NH mid-lats) +0.2 to +0.4 ppb (tropics)	-	1990 to 2030 (Global)	[117]
CO_2 Fertilization + Anthropogenic LUC	-2%	+3%	-	-	1850s to 1990s (Global)	[85]
CO_2 Fertilization + CO_2 Inhibition	-21%	-	-	-	1901 to 2002 (Global)	[79]
CO_2 Fertilization + CO_2 Inhibition	+7%	-	-	-	1880 to 2000 (Global)	[97]

Table 4: Changes in global mean direct radiative effect due to historical and future (F, 2100) LUC. Pre-industrial (PI) is variously defined in the studies used here ranging from 1750 to 1850. Best estimate shown first, with range in brackets. Where not explicitly described in the footnotes, changes in direct radiative effect due to LUC are estimated by multiplying emission changes from Table 2 to the aerosol DRE. Similarly, unless otherwise stated, best estimates are mean of given ranges.

Atmospheric Species	**Direct Radiative Effect (Wm^{-2})**	**Change in Direct Radiative Effect due to LUC (Wm^{-2})**	
		PI to PD	**PD to F**
Tropospheric O_3[a]	-	-0.11	-
Biogenic SOA	-0.18[b]	+0.034[g] (+0.012 to +0.056)	+0.0145[c] (+0.007 to +0.022)
Nitrate[d]	-0.15[e]	-0.094[f] (-0.070 to -0.118)	-0.075[g] (-0.042 to – 0.158)
Dust	-0.26[h]	-0.056 (-0.047 to -0.065)	-0.020 (-0.013 to -0.026)
Smoke	-0.01 to -0.19[i]	+0.028[j] (0.0 to +0.057)	+0.039[k] 0.0 to +0.078
Total Aerosol	-0.6 to -0.78	-0.102 (-0.171 to -0.031)	-0.0415 (-0.177 to +0.045)
Net		-0.21 (-0.14 to -0.28)	-

[a] We do not quantify here the effect of LUC on tropospheric O_3 via changes in precursor emissions (as discussed in Section 2) given the non-linearities in ozone formation. Instead, we cite here Unger (2014) for historical impacts. The impact of future LUC on tropospheric ozone has not been estimated.
[b] Sum of DRE for monoterpene (-0.10 Wm^{-2}) and isoprene (-0.08 Wm^{-2}) from Scott et al. (2014).
[c] We apply the emissions changes for monoterpenes and isoprene from Table 2 separately to the DRE for each.
[d] This includes only the effects of ammonia emissions changes. To date, no studies have quantified the impact of land use change driven changes in NOx source (soil, fire) on nitrate abundance.
[e] We applied the DRE/DRF fraction from Heald et al., (2014) to the IPCC (2013) value for the DRF of nitrate.
[f] The impact of ammonia emissions on nitrate formation will vary with the local environment. Here for simplicity, we attribute 63% of the nitrate DRE to LUC given that 63% of present-day ammonia emissions are associated with agricultural sources. We give a ±25% range on this central value.
[g] Our best value represents a 50% increase in ammonia emissions projected by Erisman et al. (2008), with the range as in Table 2.We assume that there is sufficient NO_x available to form ammonium nitrate; thus this is an upper-limit.
[h] Heald et al. (2014).
[i] Rap et al. (2013) estimated -0.01 Wm^{-2}; Heald et al. (2014) estimated -0.19 Wm^{-2}.
[j] We estimate a range of -11% to -30% for the reduction in aerosol emissions from pre-industrial to present day. Based on Kloster et al. (2010) who estimate that LUC and wood harvest have reduced fire carbon emissions by 11%. Yang et al. (2014) predict that human activity has reduced global area burned by 30% from 1900 to present day. We assume that aerosol emissions scale with carbon emissions or area burned.
[k] We estimate the net effect of human activity on future fire (2075-2099 compared to 1985-2009) as -11% to -41% from Kloster et al. (2012) through combining a -5% to -35% change in fire due to LUC and wood harvest (range driven by different RCP scenarios) with a -6% due to increased fire suppression.

FIGURES

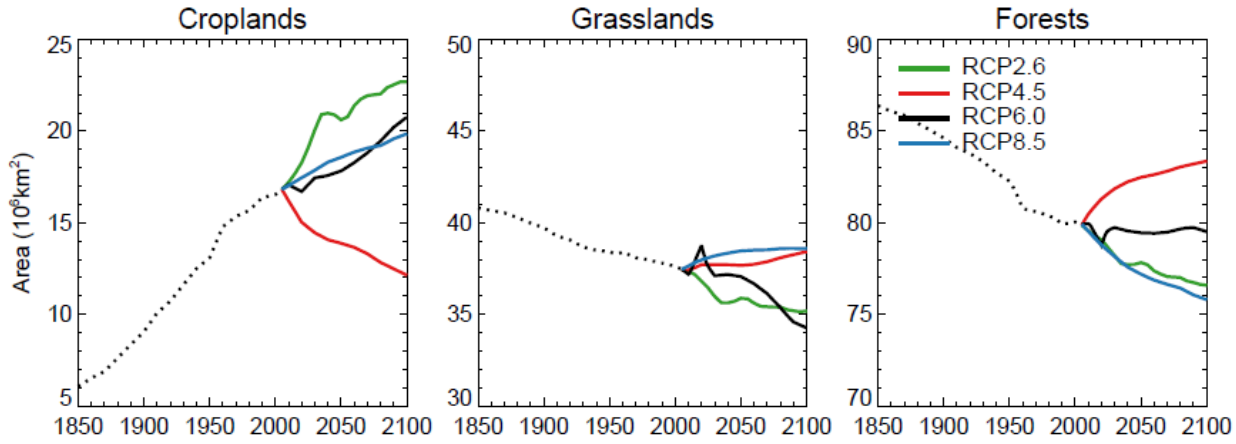

Figure 1: Historical (dashed) and projected (colored by RCP scenario) global land use area for 3 major categories. Data from Lawrence et al. [48]

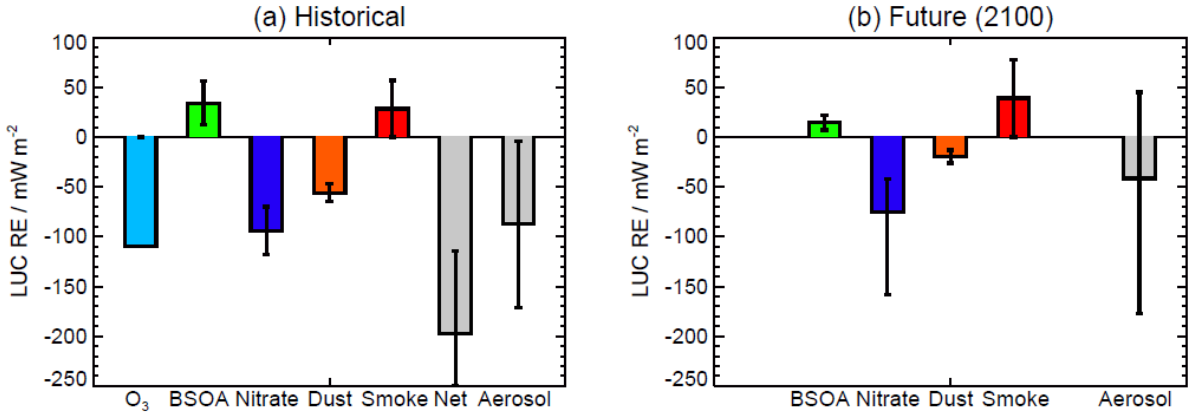

Figure 2: Changes in global mean d radiative effect (RE) due to (a) Historical (pre-industrial to present day) and (b) Future (2100) LUC. For aerosol RE is restricted to the direct radiative effect. See Table 4 for details.

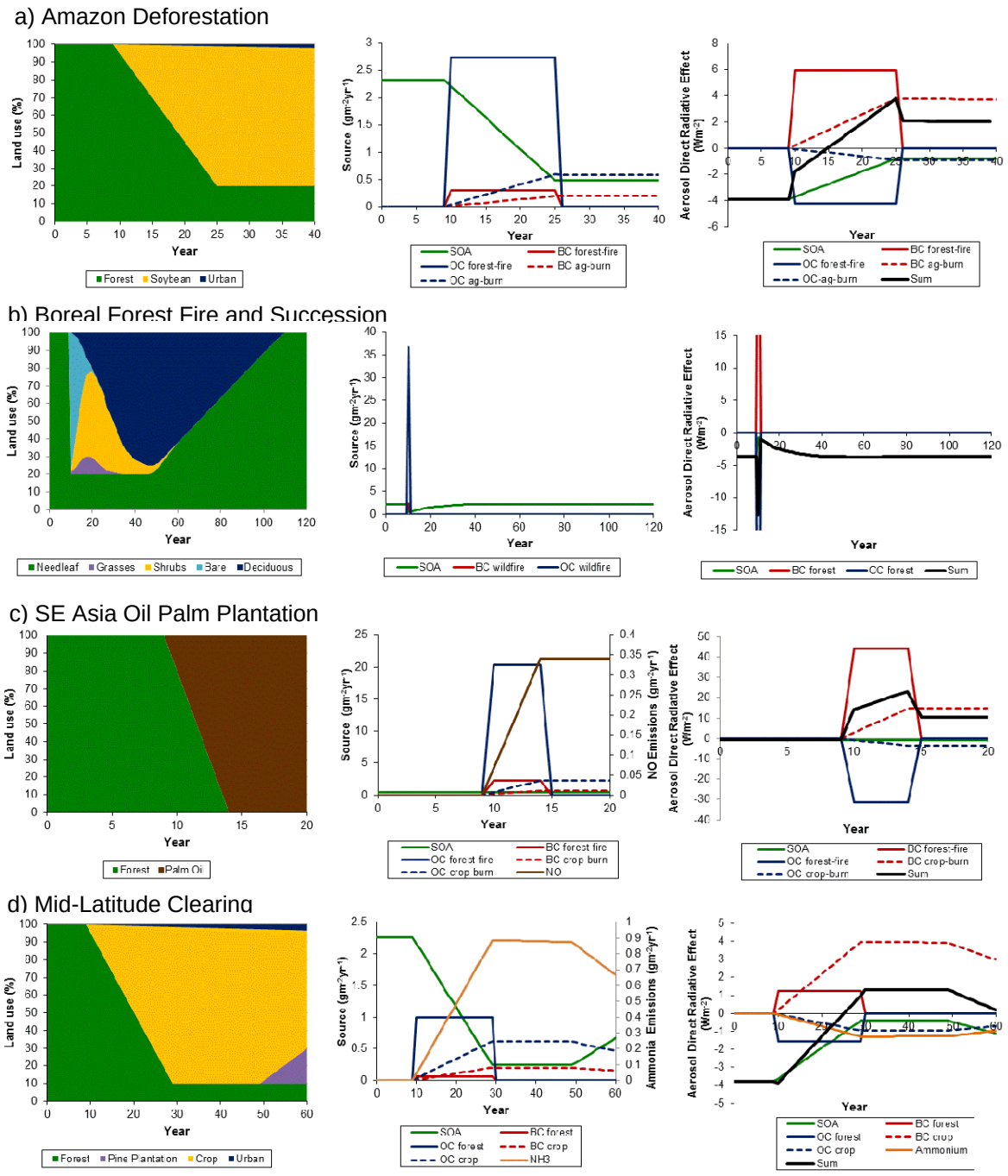

Figure 3: Local Land Use Change Scenarios (left) with Estimated Impacts on Aerosol Sources (center) and Radiative Effects (right). For each scenario land use change starts in year 10. Details on calculations provided in supplementary materials.

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