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Reviews and syntheses: influences of landscape structure and land uses on local to regional climate and air quality

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We present here the physical, chemical and biological theoretical backgrounds for the different interactions presented in this paper.

1. Physical processes

The different types of surfaces covering the earth tightly control (micro-) climate through their influences on the radiative budget, the energy balance, the water balance, and air flows. The radiative budget determines the energy received by the surface. For any surface, the net radiation (Q*) is defined as follows:

$$Q^* = (S \downarrow - S \uparrow) + (L \downarrow - L \uparrow) \qquad \text{(S1)}$$

where S and L are short- and longwave radiations, respectively, and ↑ and ↓ refer to upwelling and incoming components, respectively. By considering surface albedo (α) and surface and air emissivities (ε_s and ε_a, respectively), surface temperature (T_s) and high altitude air temperature (T_a), Eq. (1) becomes:

$$Q^* = (S \downarrow - \alpha S \downarrow) + (\varepsilon_a \cdot \sigma \cdot T_a^4 - \varepsilon_s \cdot \sigma \cdot T_s^4) \qquad \text{(S2)}$$

with σ the Stefan-Boltzmann constant (= 5.67×10^{-8} W m^{-2} K^{-4}). The energy balance for any surface is linked with its radiation budget through Q* and can be expressed as (assuming there is no energy stored with land, which excludes therefore permafrost regions or regions with snowy winters, for example):

$$\Delta Q_S = Q^* - H - -G \qquad \text{(S3)}$$

where ΔQ_S is the change of energy within the considered surface layer, H is the sensible heat flux (dry heat convectively exchanged between the surface and the atmosphere, that changes both the emitter and receptor temperatures), LE is the latent heat flux (i.e., energy dissipated during evapotranspiration, water vapour convectively exchanged between the surface and the atmosphere, that changes both the emitter and receptor moisture conditions). LE includes both water evaporation (E) (i.e., from soil, dew, water interception by leaves, lakes and oceans) and plant transpiration (T). G is the conductive ground heat flux from/to deeper layers. G is often small and negligible for minor scales compared to H and LE fluxes. The energy and water balances are connected through the evapotranspiration (i.e., the sum of E and T). The water balance for a surface including vegetation without considering lateral exchange between adjacent soil volumes can be expressed as:

$$\Delta S = P - E - T - R - D \qquad \text{(S4)}$$

where ΔS is the change of water content within the given layer, P is the precipitation (in case of surface layer) or percolation from the above layer, R is the surface runoff, and D is the drainage. Note that the term ΔS includes soil moisture, surface water, snow, ice cover, and — depending on the depth of the considered soil layer — groundwater. It also includes the interception storage. Finally, any convective fluxes between the atmosphere and the surface in the surface boundary layer can be expressed following the flux-profile relationships as:

$$F_\chi = -k \cdot u_* \cdot \frac{\chi_z - \chi_s}{ln\left(\frac{z-d}{z_0}\right) - \psi_\chi(z/L)} \tag{S5}$$

where F_χ is the bi-directional land-atmosphere turbulent flux of the scalar χ (e.g., temperature, water vapor, carbon dioxide, ozone), k is the von Karman constant (= 0.4), u_* is the friction velocity, z is the height above ground, d is the displacement height, z_0 is the roughness length, and $\psi_\chi(z/L)$ is the stability correction function accounting for atmospheric stability.

From the previous equations, it can be seen that any LULCC induces modifications in the surface radiative, energy, and water budgets, which may in turn modify the climate. First, the energy received by the surface is closely related to surface properties (i.e., α and ε_s) (Eq.2). Any darkening (brightening) of the surface by LULCC will decrease (increase) albedo and make more (less) energy available at the surface. This alteration may result in increased (decreased) surface and air temperature. Similarly, any increase (decrease) in surface emissivity due to LULCC modifies the radiative budget of the surface resulting in the decrease (increase) in surface and air temperature. Typical values of albedo (α), Bowen ration (β) and roughness length (z_0) are summarized in **Erreur ! Source du renvoi introuvable.**. Then, LULCCs modify the energy dissipation which occurs mainly through turbulent fluxes (*H* and *LE)* (Eq. 3), and the partitioning between H and LE that is often characterized by the so-called Bowen ratio (i.e., β the ratio *H*/*LE*, see Table 1 for typical values). This latter varies with surface properties: the largest the amount of evapotranspirative surface is, the lowest the Bowen ratio is. The Bowen ratio is controlled by the presence/absence of free water (e.g., lakes, oceans, rivers, soils) and as well by the presence/absence of vegetation (e.g., surface, density, phenology) and its physiological activity particularly linked with stomatal conductance (for details about the factors affecting stomatal opening and closure, see part 2 below on biological processes). The partitioning of turbulent heat fluxes influences local climatic conditions, especially air temperature: while a large Bowen ratio (i.e., *H* >> *LE*) induces local warming of the ambient air with consequences and feedbacks on ecosystem functioning (e.g., thermal stress) and air pollution (e.g., chemical production/depletion in the atmosphere), a small Bowen ratio due to larger *LE* allows surface cooling as energy is converted into latent heat, followed by air cooling as H is reduced. Yet, it also influences the water balance due to its link with *LE* (Eq. 4). Finally, although convective fluxes are closely related to local climatic conditions (e.g., wind speed and temperature influencing u_* and atmospheric stability respectively), surface largely influences the efficiency of convective fluxes through its impacts on *d* and z_0 (Eq. 5). Increasing surface roughness (e.g., through afforestation) enhances turbulent exchanges owing to the increase in *d* and z_0, and conversely. In a general manner, the higher the canopy is, the larger *d* and z_0 are (Table 1), even if they are influenced by other parameters (e.g., LAI for pseudo-natural ecosystems, building density for urbanized areas). However, it must be kept in mind that land-atmosphere exchanges are also dependent on scalar concentration difference between the surface and the atmosphere, meaning that air mass composition (e.g., temperature, H_2O, CO_2 or pollutants) and surface emissions (e.g., from manure application or anthropogenic activities) are crucial variables. Yet, plants can absorb or emit various compounds according to their development and functioning in link with meteorological conditions. In turn, the magnitude and direction (i.e., from or to the atmosphere) of the fluxes will affect the atmospheric composition.

2. Biogeochemical processes at the land/atmosphere interface

Biological activity occurs in soils and within the vegetation. It affects number of physical, chemical and biogeochemical processes and therefore also the exchanges between land and atmosphere.

Soil microbial activity primarily involves the production of energy by autotrophs through aerobic respiration. Organisms can obtain energy through anaerobic respiration that originates from the reduction of organic compounds, such as fermentation, or inorganic compounds, such as nitrate reduction, denitrification or methanogenesis. The oxidation of certain minerals, also called chemolithotrophy, can also be a source of energy for living organisms such as nitrification, anammox (ammonium anaerobic oxidation) or sulphur oxidation. As all metabolic pathways, environmental factors such as temperature, water presence or absence, and substrate availability control those processes and are therefore affected by LULCC. The different metabolic pathways release into the environment different reactive gases (NH_3, NO_x, BVOCs) and non-reactive (or less reactive) organic or mineral compounds (CO_2, N_2O, CH_4, H_2O) affecting the atmospheric composition. These compounds can have chemical (see Sect. 3.3) or physical effects (see Sect 3.1 change in the water and energy budget) and/or warming effect. In turn those atmospheric changes feed back on ecosystem functioning through direct and indirect effects.

Plants are considered as heterotroph and can therefore convert sunlight and CO_2 into organic carbon through photosynthesis. One of the major actors in photosynthesis is the stomatal movement, which allows the leaf to change both the partial pressure of CO_2 at the sites of carboxylation and the rate of transpiration interlinking the water and carbon budgets. Another important actor of photosynthesis is RuBisCO, the major enzyme involved in the fixation of CO_2. RuBisCO is a rate-limiting factor for potential photosynthesis under the present atmospheric air conditions (Spreitzer & Salvucci 2002). It contains relatively large amounts of N, accounting for 10 to 30% of total leaf N-content for C3 type plants and 5-10% of total leaf N for C4 type plants it is thus an important link between the C and N cycles in vegetated surfaces (Makino, 2003; Carmo-Silva et al. 2015).

The plant's photosynthetic enzymes and the functioning of the stomata are affected by: (i) changes in the physical environment of the leaves (water potential, temperature, and CO_2 concentration; Farquhar and Sharkey, 1982); (ii) contact with atmospheric chemical pollutants (oxidative gaseous compounds, nitrogen deposition); (iii) availability of other resources (nitrogen, phosphorous); and (iv) interaction with adjacent living organisms (competition for resources, invasion by pests). Climate change or land use and land cover changes can directly or indirectly modify all these factors. Moreover, stomatal conductance plays a major role in the surface energy budget when plants are involved, as explained in Sect. 3.1, and can be one of the pathways of feedbacks between the atmosphere and the surface since they regulate CO_2 input to the leaves and water output from the leaves. Vegetated surfaces are also involved in the exchange of other reactive species such as NH_3 in fertilized agricultural land as well as volatile organic compounds (VOCs) as a communication or defence tool that facilitate interactions with their environment, from attracting pollinators and seed dispersers to protecting themselves from pathogens, parasites and herbivores (Dudareva et al., 2013).

Some examples of how LULCC can affect climate through biological activity of soils and plants are given below:

- **Water Use Efficiency (WUE)** is defined as the ratio between the rates of carbon assimilation (photosynthesis) and transpiration. Plants that can have a lower transpiration rate without simultaneously decreasing their photosynthesis and thus biomass production are a desired trait in crop production. C4 type photosynthetic plants as opposed to C3 type photosynthetic plants have the capacity to concentrate CO_2 in their mesophyll cells and can therefore have a higher WUE. Plants in general respond to changing CO_2 concentrations, for example, it has been shown that an increased CO_2 concentration tends to reduce stomatal conductance while still increasing photosynthesis (Ainsworth and Rogers, 2007). This has several implications when considering different land uses in the context of climate change and increased CO_2 concentrations. Recent research demonstrate that most of C4 plants almost certainly display increasing water-use efficiency with increasing CO_2 concentrations, which allows them to better deal with conditions of water stress (Maroco et al., 1999; Conley et al., 2001). Consequently, this phenomenon should allow plants, in the future, to grow in areas where they currently cannot survive due to limited soil moisture availability. Those same plants will also be able to better resist drought periods and heat waves (Prior et al., 2011; Aparicio et al. 2015). WUE issues can be artificially overcome by irrigation, with consequences on plant phenology and local climate. Intensification of the water cycle or increased drought conditions because of climate change and LULCC modify the biological functioning of the soil-vegetation system and lastly influence the local climate.
- Increased **temperature** and frost-free days as well as atmospheric CO_2 concentrations affect the activity of RuBisCO. As a result, the growing season elongates and, if no other limiting factors are present, the net primary production (NPP) increases accordingly (Reyes-Fox et al. 2014; Fridley et al. 2016), which could be beneficial in temperate regions. However, longer growing seasons increase pressure on the water cycle therefore affecting local climate and resulting in potentially negative feedbacks on the carbon cycle (Wolf et al. 2016; Ciais et al. 2005). Due to temperature effects, species migrate to higher latitudes or altitudes (Hillyer and Silman, 2010; Brown et al. 2014; Spasojevic et al. 2013) resulting in LULCCs, changes in emissions of reactive trace gases and in habitat for biodiversity. Finally, higher temperatures enhance soil microorganism activity leading to higher mineralisation rates and consequently CO_2 release to the atmosphere.
- Rate of photosynthesis is directly correlated to leaf nitrogen content on a mass basis. **Nutrient Use Efficiency (NUE)** is defined as the ratio between the amount of fertilizer N removed from the field by the crop and the amount of fertilizer N applied. Increased anthropogenic synthesis of mineral fertilizers to intensify crop production impairs the global N cycle as illustrated by the N-cascade (Galloway et al., 2003;

Fowler et al., 2013) with impacts on biodiversity (Sutton et al., 2011), water and air quality (Billen et al., 2013; Erisman et al., 2013), and productivity and nutrient cycling (Phoenix et al., 2003; de Vries et al., 2009). Nitrogen and carbon cycles are interlinked through biosphere-atmosphere interactions via biological processes, as detailed here, and chemical processes in the atmosphere, as detailed in Sect. 3.3. Nitrogen is a limiting nutrient for plant growth. In the tropics, warmer and wetter climate induces high soil mineralization and biological fixation (Cleveland, 2006; Yang et al., 2010) therefore increasing N availability; however, this is not the case in mid- and high-latitude regions. Increasing N availability to vegetated surfaces raise NPP, at least temporarily, with increased C storage in soils and higher N values in the vegetation (Yue et al. 2016) with direct effects on climate but also indirect effects via impacts on the water and energy budgets of certain areas.

- Another example is the effect of elevated biotic or abiotic stress on plants. Increased **ozone** concentrations is a typical example, which affects stomatal conductance and photosynthesis (Fowler et al., 2009; Reich and Lassoie, 1984). Ozone is a strong oxidant that can alter the functioning of plant cell in different ways. At relatively high concentrations, we observe: (i) direct damage of leaf epidermis cells (Sandermann et al., 1997; Günthardt-Goerg et al., 2000), (ii) modification of stomatal resistance via damage of guard cells causing leaky stomates (Paoletti and Grulke, 2010; Wittig et al., 2007), and (iii) alteration of cell walls and cell membranes (Gunthardtgoerg and Vollenweider, 2007). At low concentrations, we observe also negative effects: (iv) ozone penetration to the mesophyll cells enhances production of reactive oxygen species (ROS) (Schraudner et al., 1998; Wohlgemuth et al., 2002), and it can also alter certain proteins and enzymes therefore affecting plant photosynthesis and biomass production (Heath, 1994). It is important to note that there is an accumulative effect of exposure to ozone concentrations by the plant (Fuhrer et al., 1997; Super et al., 2015). Different stresses affect different plant functioning but in most cases they induce the production of ROS and the emissions of biogenic VOCs with consequences on air quality.

In summary, the major biologically driven interactions from a LULCC or LMC perspective between the atmosphere and the terrestrial biosphere result from the following changes. (i) The total productivity of the ecosystem as affected by changes in photosynthesis and soil microorganism activity and conditioned by the availability of water and nutrients (N) thus resulting in the release or absorption of CO_2 to/from the atmosphere. (ii) Enhanced exchange of reactive trace gases (NH_3, BVOCs, NO_x) and their subsequent impact on nutrient availability in ecosystems and air quality. (iii) The indirect impacts of plant productivity on the energy and water budgets locally and regionally and their subsequent impacts on local and meso climates. In the sections below, we discuss some examples of these biological interactions as influenced by three LULCC and LMC.

3. Chemical processes in the atmosphere

Terrestrial ecosystems are both sources (nitrogen and organic species, particles) and sinks (ozone for instance through deposition on vegetative surfaces) of chemical compounds. Along their life, even trace amounts of these reactive gaseous and particulate matter (called aerosols) interact and influence the Earth system at large scales, regarding climate evolution, and at regional-local scales, regarding air quality. Air pollutants, both gases and aerosols, threaten human and ecosystem health and can be directly emitted (primary pollutants), or produced by reactions between primary pollutants (so called secondary pollutants). Any modification in the landscape structure, land-use or land management therefore has the potential to modify the air chemical composition. Some agricultural practices are shown in the literature to affect air quality. This is the case of fertilization as a source of ammonia, fires as a source of ozone precursors and aerosols, or fallow periods as a source of coarse aerosols. In this section we will focus especially on secondary pollutants such as ground-surface ozone (O_3) and secondary aerosols, that strongly affect air quality and whose production, lifetime and deposition involve the terrestrial biosphere, as demonstrated in several publications.

- Compound emissions

Natural sources contribute 90%of global annual VOC emissions (BVOCs, mainly from vegetation, with a minor contribution from oceans), while anthropogenic source (AVOCs, e.g., motor vehicle exhaust, solvents, biomass burning) only contribute 10% (Simpson et al., 1999). VOCs include thousands of different species. Among BVOCs, isoprene and monoterpenes are the most abundant, with isoprene that contributes around 50% of the total BVOC emissions and is mainly released by tropical and temperate vegetation, whereas monoterpenes contribute around 15% and are mostly emitted by boreal vegetation (Arneth et al., 2008). These secondary metabolites have been shown to play an important role for plants (thermotolerance, plant protection against abiotic stressors, plant-plant or plant-insect communication, etc.) (e.g., Peñuelas and Llusià, 2001). Broadleaf and needle-leaf forests are usually much stronger BVOC emitters compared to crops and grasslands. Temperature, radiation, water stress and atmospheric CO_2 concentration are strong external drivers of BVOC emissions (Peñuelas and Staudt, 2010). With a lifetime of a few minutes to hours, BVOCs are very reactive gases that play an important role in photochemistry (i.e., O_3 production), and contribute to the formation of biogenic secondary organic aerosols (BSOAs) (Atkinson and Arey, 2003).

Agricultural fertilization and natural soil processes of nitrification and denitrification are a significant source of nitrogen compounds, such as nitrogen oxide (NO) and nitrogen dioxide (NO_2). These two compounds are treated as a unique family (i.e., nitrogen oxides, NOx) due to the rapid cycling between NO and NO_2 during daytime (about one minute), while the NOx family is mainly composed by NO_2 at night-time. Overall, the lifetime of NOx is approximately one day. At the global scale, NOx are mainly emitted by anthropogenic sources (e.g., fossil fuel combustion, biomass burning) and more moderately by lightning.

- **Surface ozone**

Ozone is a highly reactive compound that is present in the stratosphere, where it protects life on Earth from ultra-violet (UV) radiations, in the troposphere and close to the surface, where it threatens human and plant health due to its oxidizing effect on living tissues. Ground-surface O_3 has a lifetime of one month and is mainly formed on sunny and warm days because of a complex and non-linear interplay between NOx and VOCs (Sillman, 1999). Surface O_3 production relies on the imbalance between O_3 production via NO_2 photolysis (i.e., NO reactions with peroxy radicals, HO_2) and O_3 removal via reaction of O_3 with NO. Organic peroxy radicals (i.e., RO_2) from the oxidation of VOCs in forested (BVOC-dominated) or highly polluted (AVOC-dominated) regions also contribute to O_3 production. While O_3 removal depends on O_3 photolysis, reactions with radicals (e.g., OH and HO_2) in remote regions, and dry deposition. The O_3 chemistry is characterized by two different photochemical regimes, driven by NOx and VOC concentrations: the NOx-sensitive regime, with relatively low NOx and high VOC concentrations, where O_3 increases with increasing NOx levels, with low sensitivity to VOCs; the VOC-sensitive regime, where O_3 increases with increasing VOC levels and decreases with increasing NOx (Sillman, 1999). Natural and anthropogenic ecosystems can therefore both influence the level of ozone concentration in the atmosphere, as sources of compounds involved in the ozone cycle, and be impacted by the ozone oxidizing effect, depending on the pollution level.

- **Secondary aerosols**

Atmospheric aerosol particles originate from a large variety of natural and anthropogenic sources. While primary aerosols are directly emitted as liquid droplets or solid particles (e.g., mineral dust, sea salt, pollen, black carbon from diesel engines or biomass burning), secondary aerosols result from gas-to-particle conversion. Secondary aerosols include inorganic (e.g., sulfate, nitrate) and organic species (named organic aerosols, OA), each species typically contributing about 10–30% of the overall mass load. However, both location and meteorological conditions strongly influence the air composition and the relative abundance of different aerosol types (Tunved et al., 2005; Deng et al., 2012).

In the last two decades, BVOCs have been identified as precursors of BSOAs, with monoterpenes and sesquiterpenes having a large potential to produce BSOAs (Kanakidou et al., 2005). Isoprene has a minor aerosol production yield but still significantly contributes to BSOA mass due to its abundance over total BVOC emissions and its large global source, especially during summer (Carlton et al., 2009). BSOA production shows a high variability that depends on external factors such as temperature and relative humidity (both playing a minor role), organic aerosol loading (which controls gas-particle partitioning of semi-volatiles), oxidants (which controls the extent and rate of reactions) and NOx levels. Carlton et al. (2009) observed the lowest SOA yields under “high NOx” conditions, whereas “NOx-free” conditions led to the highest measured SOA yields. Being involved into the absorption and scattering of radiation (direct effect) and into the alteration of cloud properties (indirect effect), BSOA, and SOA in general, can influence the radiative balance of the Earth, and therefore influence climate

(Forster et al., 2007). However, the exact contribution of BSOA to the radiative forcing is still very uncertain (Scott et al., 2014).

To form secondary aerosols, gas-to-particle conversion begins in the atmosphere with the oxidation, usually sustained by sunlight, of high volatility precursor gases (e.g., SO_2, NOx and VOCs, emitted especially from terrestrial ecosystems) into low volatility gases (e.g., sulfuric and nitric acid, ammonia, organics) that nucleate into stable molecular clusters (the ultra-fine mode, 10^{-3}–10^{-2} mm size range). Depending on ambient conditions, aerosols can still grow in size via condensation of gases onto the nucleated aerosol or coagulation (i.e., collision of two aerosols). The final aerosol size strongly determines multiple aerosol properties such as the interaction with radiation, impacts on human health, and aerosol lifetime and sinks. Typically, secondary aerosols belonging to the fine-mode have an atmospheric lifetime of about one-two weeks and can be removed from the atmosphere mainly via wet deposition (also termed scavenging), while coarse-mode aerosols, such as primary aerosols, are efficiently removed by dry deposition.

Among secondary aerosols, sulfates, nitrates and ammonium are produced primarily from atmospheric chemical reactions involving, respectively, sulfur dioxide (SO_2, mainly emitted from fossil fuel and biomass burning), NOx and ammonia (NH_3, largely emitted by domestic animals, synthetic fertilizers, biomass burning, and crops). Over half of atmospheric SO_2 is converted into sulfates, and half of emitted NH_3 is converted into ammonium aerosols. Together with nitrates, ammonium represents the main form of atmospheric nitrogen aerosols and may provide nutrients to vegetation growth in nitrogen limited systems (Mahowald et al, 2011). It is also worth mentioning phosphorus, a nutrient that plays a key role for many living organisms and is mainly present in the atmosphere in the aerosol mode. However, among atmospheric aerosols, the phosphorus composition, together with its size, geographical distribution and emission sources remain poorly characterized and investigated (Furutani et al., 2010).

Organic aerosols altogether contribute ~20–50% of the total fine aerosol mass at mid-latitudes and 90% in tropical forested regions (Kanakidou et al., 2005). Depending on the season and the location, secondary organic aerosols (SOAs) contribute 20–80% of measured mass of OAs.

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