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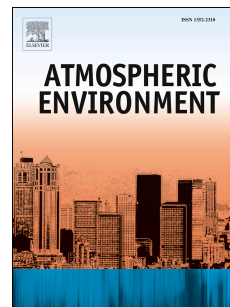
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Impact of LULCC on the emission of BVOCs during the 21st century

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Abstract

Land-use and land-cover change (LULCC) is one of the key drivers of anthropogenic climate change. In addition to greenhouse gases such as CO₂ or CH₄, LULCC affects also the emission of other carbon trace gases such as biogenic volatile organic compounds (BVOCs). We investigate the impact of changing LULCC on the emission of isoprene and monoterpenes during the 21st century using seven different land-use projections, applying the dynamic vegetation modelling framework LPJ-GUESS. Climate change, and atmospheric CO₂-concentration are based on the RCP2.6 scenario. The different LULCC-scenarios explore the impact of different land-based climate change mitigation strategies (such as afforestation and avoided deforestation, or bioenergy). We show that the increase of land area under crops or grassland would lead to a significant decrease of BVOC emissions, with a strong negative correlation between the fraction of managed global land area and the emission of isoprene and monoterpenes. But the choice of crops is important, especially for the bioenergy scenarios in which increasing fractional cover leads to decreasing BVOC emissions in our simulations; use of woody bioenergy crops can reverse this decrease. The strong impact of LULCC on the global and regional emission of BVOCs implies the need to include the impact of these changes in projections of atmospheric composition and air quality.

Keywords:

- 1 - BVOC
- 2 - LULCC
- 3 - Isoprene
- 4 - Monoterpene

5

6 **1. Introduction**

7 While in the year 1700 an estimated 95% of the ice-free land area was in a (semi) natural
8 state, by 2000 only about 45% remained relatively unaffected by human activities (Ellis et al.,
9 2010). Between 1990 and 2005 alone, nearly 9.000.000 ha of forest area were cleared for
10 cropland and pasture expansion, and human settlements (FAO and JRC, 2012). The rate of
11 deforestation has weakened over recent years, but a further growing population will still
12 need more cropland to meet its energy and food demand (Lambin and Meyfroidt, 2011,
13 Levis, 2010), even though the total area needed depends on many highly uncertain socio-
14 economic factors (Eitelberg et al., 2015). In addition, the need for land-based climate change
15 mitigation by providing biomass for bioenergy, conserving carbon-rich ecosystems from
16 agricultural expansion or enhancing terrestrial carbon sequestration by afforestation will be
17 an additional important driver of land-use change in future (Smith et al. 2014, Popp et al.
18 2014a).

19 Land-use and land-cover change (LULCC) is one of the key drivers of anthropogenic
20 greenhouse gas (GHG) emissions. LULCC-related emissions include direct and indirect
21 greenhouse gas emissions from agricultural activities such as livestock farming, manure
22 management, fertiliser use and paddy rice that contribute around 12% of today's total GHG
23 emissions and an additional ca. 10% arise as CO₂ emissions from deforestation (Smith et al.,
24 2008, Smith et al. 2014, Tubiello et al., 2015). Over the period from 1870 to 2014, for
25 example, emissions due to land-cover change were equivalent to nearly 30% of total
26 anthropogenic CO₂ emissions (Le Quéré et al., 2015, 2016). To evaluate future emission from
27 land clearance or the potential of mitigating climate change through land-ecosystem carbon
28 uptake, or through avoided future emissions from deforestation, future land-use scenarios
29 explore different pathways that are based on different assumptions such as technological
30 progress, population growth, or need for energy (e.g., O'Neill et al., 2015, Hurtt et al, 2011).
31 But not only the long-lived GHGs such as carbon dioxide (or methane and nitrous oxide) are
32 affected by LULCC, also atmospherically short-lived species are impacted. Among these are
33 biogenic volatile organic compounds (BVOC) (Rosenkranz et al., 2015). Atmospherically
34 short-lived, their high reactivity and large terrestrial source makes them a crucial component

1 of atmospheric chemistry. From a mass perspective, isoprene (2-methyl-1,3-butadiene, C_5H_8)
2 is the most important compound, with an estimated emission strength of around 400-600 Tg
3 $C a^{-1}$ (Guenther et al., 2012 Lathière et al., 2006, Arneth et al., 2008). A second important
4 compound group, monoterpenes, consist of two isoprene units ($C_{10}H_{16}$) and have
5 considerably lower (but equally uncertain to isoprene) global total emissions (Arneth et al.,
6 2008).

7 Once emitted, isoprene is primarily lost through oxidation by the hydroxyl radical, and in the
8 presence of sufficient NO_x leads to the increased formation of tropospheric ozone (O_3), a
9 greenhouse-gas which is also toxic to organisms (Atkinson, 2000, Atkinson and Arey, 2003,
10 Jerret 2009, Ainsworth 2012). Through its atmospheric oxidation by hydroxyl, isoprene is
11 believed to reduce the capacity of the atmosphere to oxidize methane, thus enhancing
12 methane lifetime (Poisson et al., 2000). However, uncertainty as to the extent to which
13 isoprene is a net sink for hydroxyl radicals means that the extent of any reductions is not well
14 understood (Lelieveld et al., 2008; Fuchs et al., 2013). Monoterpenes, but also isoprene,
15 contribute to the growth of secondary organic aerosol particles (SOA) (Carslaw et al., 2010,
16 Kulmala et al., 2005). As aerosols scatter and reflect radiation but also function as cloud
17 condensation nuclei (CCN), SOA (and hence BVOC) affect climate in various, complex ways
18 (direct and indirect effects) related to albedo, cloud lifetime, and surface-atmosphere
19 feedbacks (Spracklen et al., 2008, Carslaw et al., 2010, 2013, Paasonen et al., 2013, Kulmala
20 et al., 2014).

21 The BVOC total emission strength and underlying regional patterns are still largely unknown,
22 since large-scale observations are difficult to obtain or only indirectly possible. Isoprene
23 emissions, for instance, have been inferred from satellite remote sensing estimates of
24 formaldehyde (HCHO) in the air column. Formaldehyde is an isoprene oxidation product, but
25 interpretation of the signal is complicated by the need to combine such observations with
26 chemistry transport models and because of other processes, such as biomass burning, also
27 affecting HCHO levels (Barkley et al., 2012, Palmer et al., 2006). Ecosystem-atmosphere flux
28 measurements that have become standard over recent decades for monitoring and upscaling
29 of CO_2 exchange (Jung et al., 2011, Reichstein et al., 2007) still pose methodological
30 challenges for BVOC, due to the lack of appropriate fast and easy-maintenance sensors that
31 would allow long-term measurements at different locations. From the available data today it
32 appears that the main source of isoprene emissions is in the tropics, temperate forests emit
33 in a medium range, while boreal forests emit less isoprene, but comparatively large amounts
34 of monoterpene (Guenther et al., 1995, Monson et al., 2007, Arneth et al., 2008a, Harrison et
35 al., 2013). Besides this very general state of knowledge on relative source strengths, fine-

1 scaled emission patterns, interannual variability and/or seasonality of emissions, and
2 response to underlying biotic or abiotic drivers remain poorly understood.

3 Models of ecosystem BVOC emissions typically are based on algorithms that account for the
4 known strong leaf-temperature and, in case of isoprene and some monoterpene emitters,
5 radiation dependence, in combination with a number of other factors such as leaf age or
6 weather history (Niinemets et al., 2010). These algorithms are parameterized either for the
7 leaf- or the canopy scale, and are combined with vegetation maps or dynamic global
8 vegetation models to simulate the necessary ecosystem properties (Arnth et al., 2008,
9 Guenther et al., 2012 Niinemets et al. 2010). While using fixed vegetation maps allows to
10 project BVOC emissions into the future, accounting for effects of climate change, coupling
11 emissions algorithms with dynamic global vegetation models (DGVMs) is required to examine
12 combined effects of climate and atmospheric CO₂ concentration on emissions, for instance
13 through enhanced vegetation productivity and leaf area index, and to examine emission
14 response due to changes in vegetation cover (e.g. Arnth et al., 2007a, Schurgers et al., 2011,
15 Lathière et al., 2005).

16 Simulated BVOC emission potentials are greatly affected by the emission capacity (i.e.
17 emissions under standardized environmental conditions) for a given species or vegetation
18 type (Niinemets et al, 2010). Given that there are typically large differences between woody
19 and herbaceous vegetation both for isoprene, but in particular also for monoterpene
20 emissions, it seems surprising that so far only a relatively small number of studies has
21 focused on land-use change effects on historical and future BVOC emissions (e.g., Lathière et
22 al., 2006, 2010, Squire et al., 2014, Wu et al., 2012, Chen et al., 2009, Ganzeveld et al., 2010,
23 Heald and Spracklen, 2015, Unger, 2013, Unger, 2014, Hantson et al., 2017). From these
24 studies a consensus seems to emerge such that LULCC (in particular when expressed as
25 converting woody into crop or pasture vegetation) decreases global emissions of isoprene
26 and monoterpenes. For instance, Lathière et al. (2010) estimated that by the end of the 20th
27 century anthropogenic cropland expansion reduced isoprene emissions by 15% compared to
28 the beginning of the century. By the year 2095, Squire et al. (2014) estimated a 40% decrease
29 in isoprene emissions due to the effects of land-use change compared to the year 2000.
30 These projections were based on the Sheffield Dynamic Global Vegetation model with a
31 scenario of cropland expansion in the tropics and the SRES B2 emission and climate scenario.
32 Lathière et al. (2006) simulated effects of tropical deforestation over the period from 1983 to
33 1995 with the ORCHIDEE model, using the ISLSCP-II (International Satellite Land-Surface
34 Climatology Project) satellite-based climate archive and showed a decrease of 41% in
35 monoterpene emissions in the tropics, or 29% decrease globally.

1 Despite of the large effects of on emissions of isoprene and monoterpenes so far little focus
2 has been on comparing different future LULCC scenarios. In particular, future LULCC,
3 especially in the light of ambitious mitigation targets, will include not only deforestation but
4 also reforestation and/or expansion of bioenergy plantations. To study this further, we apply
5 the dynamic global vegetation model LPJ-GUESS (Smith et al., 2014) to examine how
6 emissions of isoprene and monoterpene compounds respond to changed global vegetation
7 cover, focusing especially on stylized scenarios of maximizing afforestation and reforestation
8 (jointly with avoided deforestation), as well as maximizing land used for growing dedicated
9 2nd generation bioenergy plants.

10 2. Methods

11 2.1 LPJ-GUESS

12 We used the LPJ-GUESS modelling framework (Smith et al., 2014), which simulates global
13 natural vegetation patterns as well as carbon and water cycles, on basis of climate data
14 input, atmospheric carbon dioxide concentration, nitrogen deposition and soil physical
15 properties. LPJ-GUESS is also able to simulate the emission of BVOCs in response to changing
16 climate and CO₂-concentrations in the atmosphere. Isoprene production is calculated
17 adopting the algorithm from Niinemets et al. (1999), based on the temperature and light
18 dependence as a function of the electron transport required for photosynthesis, and
19 temperature response adopted for rates seen for isoprene. The algorithm was supplemented
20 to include the frequently observed inhibition of emissions, seen at leaf scale, under
21 increasing atmospheric CO₂ (Arneth et al. 2007a). Monoterpene emissions are calculated
22 using similar algorithms for their production rates, but accounting also for storage, as
23 described in Schurgers et al (2009).

24 DGVMs represent vegetation through plant functional types (PFT), which – in case of LPJ-
25 GUESS - compete at each grid location for water, light and nutrients. Every gridcell has area
26 fractions prescribed for natural and agricultural land. For natural vegetation we used 11 plant
27 functional types as in Smith et al. (2014), 9 PFTs representing boreal, temperate and tropical
28 woody vegetation, while two PFTs represent C3 and C4 herbaceous species. Six different
29 land-use categories were specified: C3 and C4 pastures, and irrigated and non-irrigated crops
30 that were also simplified here by C3 and C4 vegetation. C4 plants are limited by a minimum
31 temperature of 15.5°C for establishment and can thus concur in some regions with C3 plants.
32 Representing harvest and grazing, 50% of the aboveground biomass on crops and pasture
33 areas, respectively, are removed each year and not returned into the soil through litter input.
34 Bioenergy crops were also assumed to be C3 or C4 crops, no woody bioenergy is considered
35 here, which for BVOC emissions is a conservative approach (Rosenkranz et al., 2015). A PFT-

1 specific standardized leaf-level BVOC emission capacity that corresponds to emission factors
2 (at 30°C temperatures and photosynthetically-active radiation flux of $1000\mu\text{mol m}^{-2} \text{s}^{-1}$
3 (Guenther et al, 1995, Arneth et al., 2007, Schurgers et al., 2009) was set for every PFT, with
4 values as in Schurgers et al. (2011). For crops we applied the values of C3 and C4 grasses.
5 The model was run with a spatial resolution of $0.5^\circ \times 0.5^\circ$. Since LPJ-GUESS contains some
6 stochastic features in its representation of growth dynamics (Smith et al., 2001, 2014), a
7 number of replicated patches (here set to 10) are specified for each gridcell. In each patch
8 PFTs are competing for resources, and patches are subsequently averaged to represent the
9 output per grid-cell. Atmospheric CO_2 concentration and N deposition for model spin-up and
10 historical simulations over the 20th century were as described in Smith et al (2014). For the
11 pre-industrial spin-up to 1901, we recycled the detrended climate data of 1901-1930 and ran
12 the model for 500 years until the carbon pools were in equilibrium. During the spin-up period
13 atmospheric CO_2 was kept at a constant mixing ratio of 296 ppmv (value in year 1900) and
14 the land-cover was held constant at the historical distribution in year 1901 as given in Hyde
15 3.1 (Goldewijk et al., 2011). As climate information, we used the IPSL-CM5A-LR Earth System
16 Model (ESM) simulation output (Dufresne et al., 2013), at daily resolution and bias-corrected
17 in the ISI-MIP project (Warszawski et al., 2014). The IPSL-CM5A-LR climate provides data for
18 the years 1950 to 2099. To provide climate data over the whole time period of the LU
19 scenarios (1901 to 2099) we extended the climate to the period 1901 to 1949 by randomly
20 choosing climate data of the years 1950 to 1959. Simulations over the 21st century are based
21 on the RCP2.6-emission scenario concerning evolution of CO_2 -concentration in the
22 atmosphere (van Vuuren et al. 2011).

23 **2.2 Land-use scenarios**

24 The land-use data over the 21st century is derived from two models, the Model of
25 Agricultural Production and its Impact on the Environment (MAGPIE) (Lotze-Campen et al.,
26 2008, Popp et al. 2014b) and the integrated model to assess the global environment (IMAGE)
27 (van Vuuren et al., 2011, Stehfest et al., 2014). Those models calculate future land use and
28 agriculture dynamics for a set of economic regions, considering population growth, economic
29 development, and technological progress affecting potential crop yields, as well as climate
30 change. Land-use changes over the 20th century were based on Hyde 3.1 (Goldewijk et al.,
31 2011) and the projected future output from MAGPIE and IMAGE were mapped onto Hyde 3.1
32 directly in order to create a harmonized historical-to-future time series. In addition to these
33 scenarios, we also used projections from Hurtt et al. (2011) for RCP 2.6, which are also based
34 on IMAGE.

1 Three scenarios from MAgPIE and IMAGE were implemented to investigate the impact of
2 different land-use projections on BVOC emissions. First, a scenario based on SSP2 (REF) that
3 does not include land-based climate mitigation (Sitch et al. in prep, see also Humpenöder et
4 al. 2015) was used as a reference. Second, ambitious land-based mitigation targets are
5 achieved through bioenergy in combination with carbon capture and storage (BECCS). In a
6 third scenario a high land-based mitigation target is achieved by means of avoided
7 deforestation and afforestation/reforestation (ADAFF). In each scenario, a number of basic
8 constraints had to be met (e.g. food supply and a minimum area set aside for conservation).
9 For comparison, we also calculated BVOC emission changes using the “standard” LULCC
10 historical-to-future estimates provided for RCP2.6 as described in Hurtt et al. (2011; H11).
11 The managed area fraction used in LPJ-GUESS (pasture, crops and irrigated crops) are derived
12 from the scenarios. Natural land, especially woody vegetation cover was not considered to
13 be managed for forest products in this version of the model.

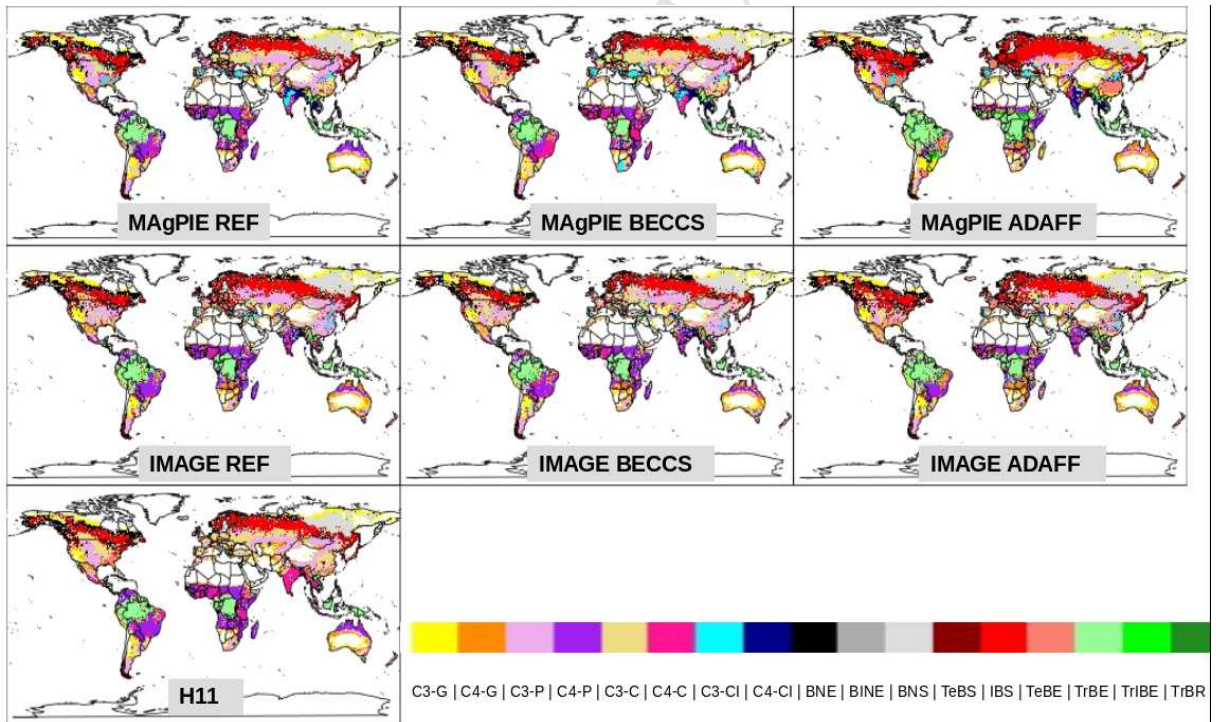
14 **2.3 Differences between the MAgPIE, IMAGE and H11 model frameworks**

15 The IMAGE and MAgPIE mitigation scenarios are based on the SSP framework and attempt to
16 maximize land-based mitigation but no explicit mitigation target was set, resulting in
17 different land requirements. The variation in land requirements also reflects different
18 modelling approaches. In IMAGE, land-use allocation follows a rule-based approach
19 according to sustainability criteria, which implies that bioenergy production or afforestation
20 can only take place on land that is not needed for food production. In contrast, competition
21 for land between food production and land-based mitigation is explicitly modeled in MAgPIE
22 based on a cost minimization procedure.

23 The H11 scenario is based on the IMAGE RCP 2.6 scenario (Hurtt et al., 2011, van Vuuren et
24 al., 2011). The largest part of the differences between H11 and the IMAGE REF, BECCS and
25 ADAFF scenarios arises from different socio-economic assumptions. In contrast to the BECCS
26 and ADAFF scenarios the H11 scenario also has a particular climate target (radiative forcing
27 of 2.6 Wm^{-2} in 2100) which considers a broad portfolio of mitigation measures across
28 different sectors. However, the H11 scenario did not include afforestation as a mitigation
29 measure. Lastly, the RCP 2.6 scenario which was the basis for the H11 scenario was produced
30 with IMAGE 2.4 (Bouwman et al., 2006) while the IMAGE mitigation scenarios were produced
31 with IMAGE 3.0 (Stehfest, 2014). Model developments such as the coupling to LPJmL
32 (Bondeau et al., 2007) also explain part of the differences.

34 **3. Results**

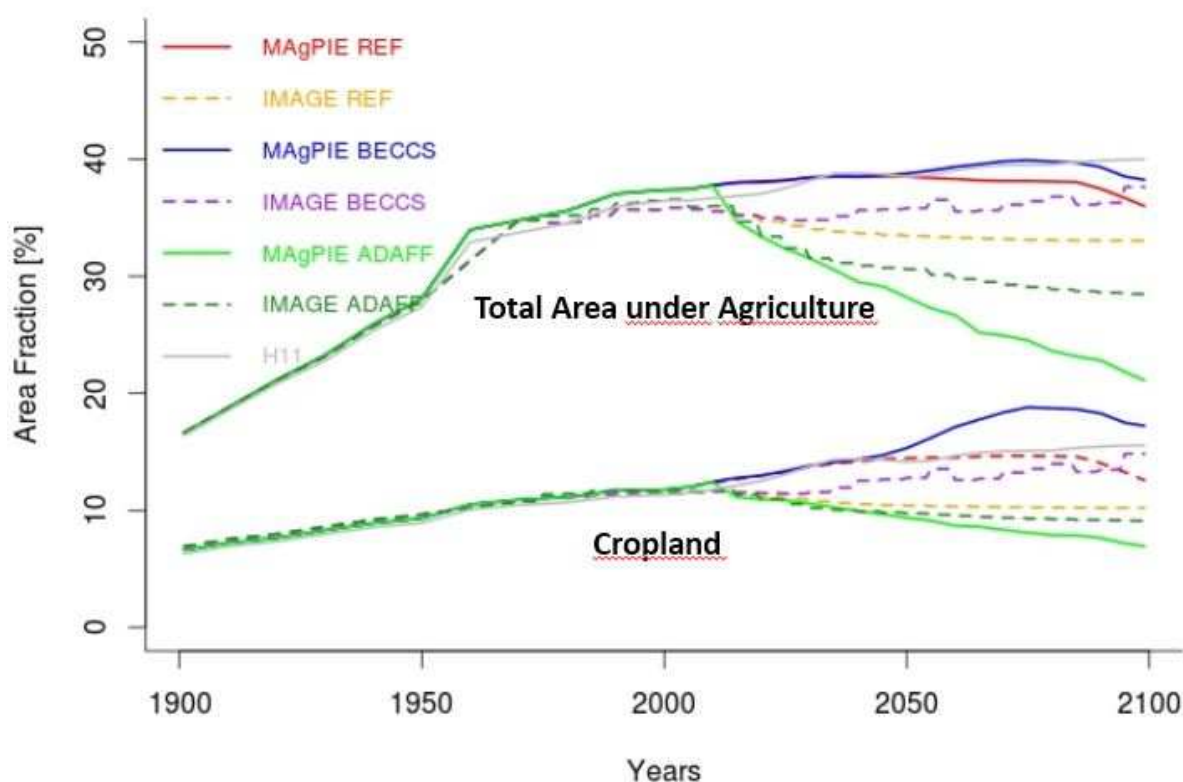
1 Maps of the different vegetation cover patterns in the LU scenarios, translated into the LPJ-
 2 GUESS dominant PFTs, for the end of 2100 are shown in Figure 1. As expected, the
 3 distribution differs strongly both between the scenarios, but also between the different LU
 4 models. The reference scenario in MAgPIE and H11 are fairly similar in terms of cropland and
 5 pasture distribution, but have some obvious differences e.g., in the assumption of where
 6 crops are irrigated (for instance in India). By contrast, the reference IMAGE case shows a
 7 larger area dominated by pastures, especially in the northern hemisphere. In both ADAFF
 8 stylized scenarios the tropical woody PFTs have a larger areal extent compared to REF, BECCS
 9 and also the H11 land cover, a similar observation can be made for parts of the temperate
 10 zone (e.g., in the central US and parts of Europe and Asia), where temperate woody
 11 vegetation expands. In the BECCS scenario the tropical woody PFTs are reduced by expansion
 12 of C4 cropland and C4 pasture, especially in southeastern Brazil and parts of central Africa. In
 13 temperate regions such as central USA, Europe or central Asia C3 croplands expand.



14

15 *Figure 1: Maps showing dominant PFTs per gridcell (PFTs with the highest LAI within each gridcell).*
 16 *Mean values for the period 2081-2100 for all seven different model projections. Abbreviations are:*
 17 *TrBR: Tropical broadleaved raingreen; TrIBE: Tropical shade intolerant broadleaved evergreen; TrBE:*
 18 *Tropical broadleaved evergreen; TeBE: Temperate broadleaved evergreen; IBS: Shade intolerant*
 19 *broadleaved summergreen; TeBS: Temperate broadleaved summergreen; BNS: Boreal needleleaved*
 20 *summergreen; BINE: Boreal shade intolerant needleleaved evergreen; BNE: Boreal needleleaved*
 21 *evergreen; C4-CI: C4 Crops Irrigated; C3-CI: C3 Crops Irrigated; C4-C: C4 Crops; C3-C: C3 Crops; C4-P:*
 22 *C4 Pasture; C3-P: C3 Pasture; C4-G: C4 Grass; C3-G: C3 Grass. (Crops and Pasture simulated as*
 23 *grasses, see methods).*

1 Figure 2 shows the corresponding development of the total fraction of area under some form of crop
 2 or pasture in all LU scenarios. At the end of the 21st century, the total land area under agriculture is
 3 smallest for the ADAFF scenarios and highest for the MAgPIE BECCS scenario, the latter being more
 4 or less similar to the H11 land cover estimates. For only cropland area the development of almost all
 5 the scenarios is equivalent to the development of the total fraction of area under some form of
 6 agriculture, except for MAgPIE BECCS, where the cropland area fraction shows a strong increase from
 7 2050 on and stays at an even higher value than H11.

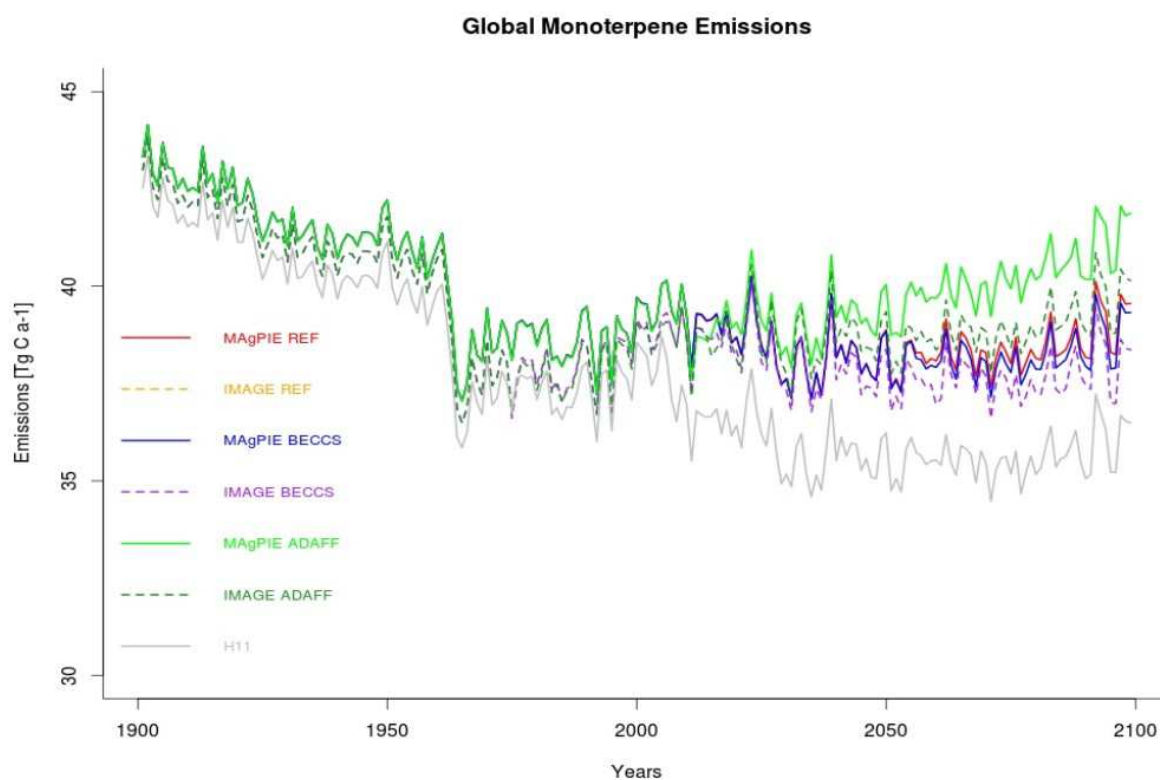
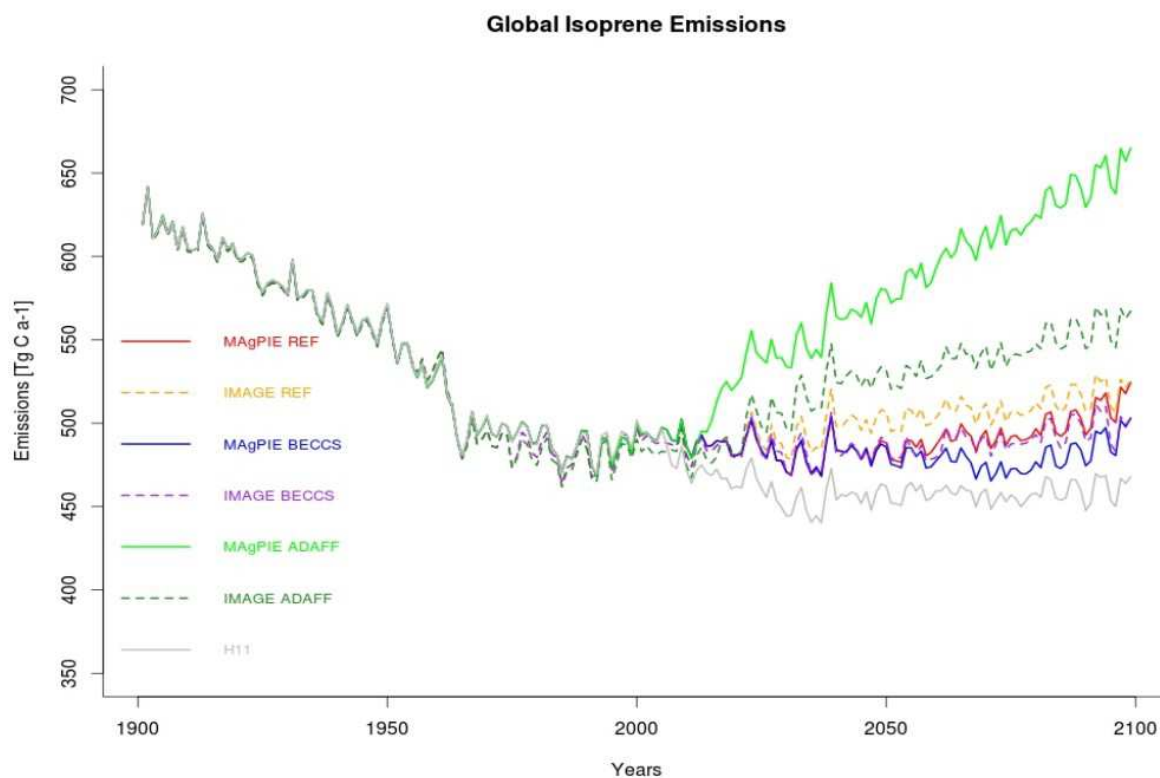


8

9 *Figure 2: Global area fraction under agriculture: total (Pasture and Crops), and cropland-only area*
 10 *fraction (Crops) for all seven LULLC-scenarios.*

11 The mean value of isoprene emission for the REF scenario of both IAMs and the H11 land cover for
 12 present day (2000-2010) is $\sim 480 \text{ Tg C a}^{-1}$ (Figure 3), which is well within the range of $412\text{-}601 \text{ Tg C a}^{-1}$
 13 given in Arneth et al. (2008). The mean value of monoterpene emission for the REF scenarios and the
 14 H11 land cover is $\sim 39 \text{ Tg C a}^{-1}$ (compared with $32\text{-}127 \text{ Tg C a}^{-1}$; Arneth et al., 2008). Figure 3 shows
 15 the change of the BVOC emissions in response to all simulated scenarios. Monoterpene as well as
 16 isoprene emissions follow a declining trajectory from 1901 until 2000. This decline is partially due to
 17 the CO_2 inhibition of isoprene production and reflecting the increase in the atmospheric CO_2
 18 concentration over the 20th century (Le Quéré et al., 2015). Historical land-cover change enhances

- 1 the decline in emissions, as woody vegetation with relatively high emission factors is replaced by
- 2 herbaceous vegetation cover. In the historical period the difference between H11 and the two IAMs
- 3 differ more for monoterpenes than isoprene due to small differences in vegetation cover in the
- 4 boreal biome (note also the different scales on the y-axes in Figure 3).



1 *Figure 3: Global sums of emissions for all seven LULCC-scenarios from 1900-2100 [Tg C a⁻¹]. Top*
 2 *panel: isoprene, bottom panel: monoterpene emissions.*

3 Over the 21st century, emissions in the two REF scenarios remain close to present-day levels, with
 4 around 470-500 Tg C a⁻¹ for isoprene and 38-40 Tg C a⁻¹ for monoterpenes. Lowest isoprene and
 5 monoterpene emissions overall are computed from the H11 land cover (Figure 3, 4, Table 1 & 2). In
 6 the ADAFF scenario emissions increase very strongly, especially for isoprene, until the end of the 21st
 7 century (Tables 1, 2) in both IAM stylized scenarios, with large differences arising from the
 8 differences in projected total vegetation cover changes in MAgPIE and IMAGE. Not surprisingly, the
 9 increase in BVOC emissions is dominated by woody PFTs. Emissions in the BECCS scenario show very
 10 small changes in different directions, with total isoprene emissions increasing by 1% for IMAGE, but
 11 decreasing by -1% in MAgPIE. Monoterpene emissions decrease by -2% (MAgPIE) and do not show
 12 any change for IMAGE. In the same period the area fraction under agriculture increases by ~1.5%.
 13 When considering cropped areas only (not irrigated) in the MAgPIE BECCS scenario emissions
 14 increase by 89% for isoprenes and by 100% for monoterpenes. This increase in emissions is closely
 15 linked to the area of cropland (Figure 2), which increases from 10% to 17% in MAgPIE BECCS for the
 16 same time period, but the effect on global emissions is small because of the low crop-PFT emission
 17 factors. For IMAGE, the increase in emissions from crops is 45% for isoprene and 10% for
 18 monoterpenes, while cropland increases by 5% compared to present day. The H11 land cover shows
 19 globally the highest increase in area fraction under management (+3% compared to present-day) and
 20 associated emissions decrease by -5% for isoprene and by -4% for monoterpene. Cropland increases
 21 in H11 are about +5% compared to present day.

22 *Table 1: Mean values of isoprene emission for the 2091-2100 period and the change in percentage*
 23 *compared to the mean value of a present day period (2001-2010) in brackets for each model*
 24 *projection. Regions are split in latitudinal bands (Boreal: 60°N-90°N, Temperate North: 30°N-60°N,*
 25 *Tropics: 30°S-30°N and Temperate South: 30°S-60°S).*

| | MAgPIE Ref | MAgPIE Beccs | MAgPIE Adaff | IMAGE Ref | IMAGE Beccs | IMAGE Adaff | H11 |
|-------|-------------|--------------|--------------|-------------|-------------|-------------|-------------|
| BNE | 5.8 (-26%) | 5.7 (-27%) | 6.2 (-21%) | 6.2 (-23%) | 5.6 (-28%) | 6.3 (-19%) | 5.4 (-31%) |
| BINE | 3.3 (-13%) | 3.2 (-16%) | 4.6 (+21%) | 3.7 (-5%) | 3.1 (-18%) | 4 (+5%) | 2.9 (-22%) |
| BNS | 1.4 (+27%) | 1.4 (+27%) | 1.4 (+27%) | 1.4 (+27%) | 1.4 (27%) | 1.4 (+27%) | 1.4 (+27%) |
| TeBS | 10.9 (+8%) | 10.7 (+6%) | 15.2 (+50%) | 12.4 (+19%) | 9.4 (-7%) | 14.3 (+42%) | 9.8 (-10%) |
| | | | 47.9 | | | | |
| IBS | 30.7 (+41%) | 29.9 (+38%) | (+121%) | 35.4 (+60%) | 28.8 (+33%) | 41.2 (+90%) | 29 (+31%) |
| TeBE | 35.6 (+5%) | 31.7 (-7%) | 56.2 (+65%) | 32.3 (+12%) | 28.6 (-16%) | 41.1 (+21%) | 34.4 (-5%) |
| | | | 243.9 | | | 214.4 | |
| TrBE | 199 (+4%) | 187.8 (-2%) | (+27%) | 204.4 (+4%) | 196.6 (+2%) | (+12%) | 179.6 (-9%) |
| | | | 117.6 | | | | |
| TriBE | 78.7 (-3%) | 74.1 (-9%) | (+44%) | 80 (0%) | 78.3 (-4%) | 91 (+12%) | 65.5 (-18%) |
| | | | 94.2 | | | | |
| TrBR | 51.5 (+20%) | 48.5 (+13%) | (+119%) | 54.8 (+27%) | 52.4 (+22%) | 66.1 (+54%) | 47.7 (+17%) |
| C3-G | 12.5 (-21%) | 11.8 (-26%) | 18.5 (+16%) | 13.6 (-15%) | 12.4 (-22%) | 15.1 (-5%) | 11.3 (-28%) |
| C4-G | 5.2 (+6%) | 4.9 (0%) | 7.2 (+47%) | 6 (+15%) | 5.7 (+16%) | 6.8 (+39%) | 4.5 (-10%) |
| C3-P | 21.3 (-19%) | 18 (-31%) | 11.4 (-57%) | 24.9 (-13%) | 24.9 (-5%) | 19.5 (-26%) | 21.6 (-15%) |
| C4-P | 15.6 (-5%) | 13.1 (-20%) | 6.5 (-60%) | 16.3 (+1%) | 16.3 (-1%) | 13.2 (-20%) | 15.7 (+1%) |

| | | | | | | | |
|--------------------|---------------------------|----------------------------|-----------------------|-------------------------|---------------------------|-----------------------|---------------------------|
| C3-C | 13.7 (+18%) | 20.3 (+75%) 12.6 | 4 (-66%) | 10.9 (-18%) | 16.9 (+46%) | 9.5 (-18%) | 19.3 (+18%) |
| C4-C | 6.5 (+12%) | (+117%) | 2.1 (-66%) | 6.7 (-4%) | 8.3 (+43%) | 5.8 (-5%) | 12.7 (+55%) |
| C3-CI | 11.8 (-6%) | 11.5 (-9%) | 9.7 (-24%) | 7.2 (-3%) | 7.2 (-43%) | 6.8 (-46%) | NA |
| C4-CI | 6.8 (+33%) | 5.2 (+2%) | 3.5 (-27%) 612.9 | 2.7 (+23%) | 2.7 (-47%) | 2.5 (-48%) | NA |
| Natural Pasture | 434.8 (+5%) 37 (--13%) | 409.8 (-1%) 31.1 (-27%) | (+47%) 17.9 (-58%) | 450 (+8%) 41.2 (-8%) | 422.3 (+2%) 41.2 (-3%) | (+21%) 32.7 (-23%) | 391.5 (-7%) 37.3 (-9%) |
| Total Crop | 510.5 (+3%) | 490.5 (-1%) | 650 (+32%) | 518.7 (+6%) | 498.5 (+1%) | (+13%) | 460.8 (-5%) |
| Rainfed Crop | 20.2 (+17%) | 32.9 (+89%) | 6 (-66%) | 17.7 (-13%) | 25.2 (+45%) | 15.3 (-14%) | 32 (+30%) |
| Irrigated | 18.6 (+4%) | 16.7 (-6%) | 13.2 (-24%) 587.2 | 9.9 (+4%) | 9.9 (-44%) | 9.3 (-47%) | NA |
| Woody | 417.1 (+6%) | 393.1 (0%) | (+49%) 504.8 | 430.5 (+9%) | 404.2 (+3%) | (+24%) 424.3 | 375.7 (-6%) |
| Tropics | 389.1 (+1%) | 372.2 (-3%) | (+32%) | 394.2 (+3%) | 382.9 (0%) | (+12%) | 349.1 (-8%) |
| Boreal | 7.4 (+45%) | 7.4 (+45%) | 7.5 (+47%) | 7.4 (+45%) | 7.4 (+45%) | 7.5 (+47%) | 7.4 (+45%) |
| Temperate N | 101.1 (+13%) | 97.8 (+9%) | (+36%) | 104.2 (+18%) | 95.6 (+9%) | (+29%) | 93 (+7%) |
| Temperate S | 12.9 (-16%) | 13.1 (-15%) | 15.8 (+3%) | 12.9 (-13%) | 12.7 (-14%) | 13.7 (-6%) | 11.3 (-22%) |

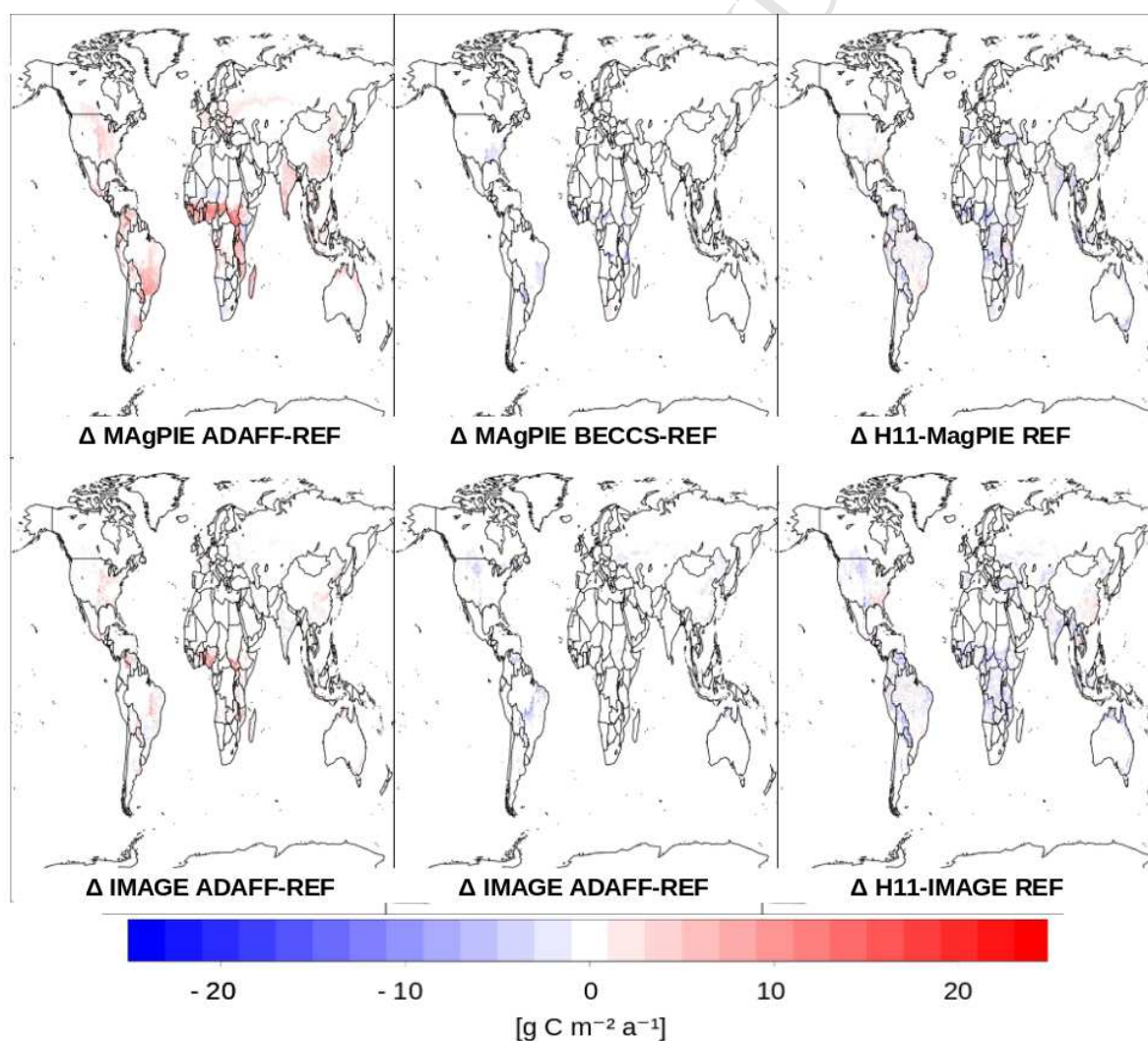
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2 *Table 2: Mean values of monoterpene emission for the 2091-2100 period and the change in*
3 *percentage compared to the mean value of a present day period (2001-2010) in brackets for each*
4 *model projection.*

| | MAGPIE Ref | MAGPIE Beccs | MAGPIE Adaff | IMAGE Ref | IMAGE Beccs | IMAGE Adaff | H11 |
|--------------------|-------------------------|--------------------------|---------------------------|-------------------------|-------------------------|---------------------------|------------------------|
| BNE | 3.4 (-26%) | 3.4 (-26%) | 3.7 (-20%) | 3.7 (-23%) | 3.3 (-18%) | 3.7 (-23%) | 3.2 (-30%) |
| BINE | 1.9 (-17%) | 1.9 (-17%) | 2.7 (+17%) | 2.2 (-4%) | 1.9 (-14%) | 2.4 (+4%) | 1.7 (-23%) |
| BNS | 0.9 (+29%) | 0.9 (+29%) | 0.9 (+29%) | 0.9 (+29%) | 0.9 (0%) | 0.9 (+29%) | 0.9 (+29%) |
| TeBS | 0.4 (0%) | 0.4 (0%) | 0.6 (+50%) | 0.5 (+25%) | 0.4 (0%) | 0.5 (+25%) | 0.4 (0%) |
| IBS | 1.2 (+50%) | 1.2 (+50%) | 1.9 (+137%) | 1.4 (+56%) | 1.1 (0%) | 1.6 (+100%) | 1.1 (+22%) |
| TeBE | 2.4 (+4%) | 2.1 (-9%) | 3.8 (+65%) | 2.2 (+16%) | 1.9 (0%) | 2.7 (+42%) | 2.3 (-4%) |
| TrBE | 6.6 (+3%) | 6.2 (-3%) | 8.1 (+27%) | 6.8 (+5%) | 6.5 (+3%) | 7.1 (+9%) | 6 (-9%) |
| TriBE | 2.6 (-4%) | 2.5 (-7%) | 3.9 (+44%) | 2.7 (0%) | 2.6 (0%) | 3 (+15%) | 2.2 (-19%) |
| TrBR | 2.8 (+22%) | 2.6 (+13%) | 5 (+117%) | 2.9 (+26%) | 2.8 (+8%) | 3.5 (+59%) | 2.6 (+18%) |
| C3-G | 1.4 (-22%) | 1.4 (-22%) | 2.1 (+17%) | 1.5 (-17%) | 1.4 (-7%) | 1.7 (-6%) | 1.3 (-28%) |
| C4-G | 1.6 (+7%) | 1.5 (0%) | 2.2 (+47%) | 1.8 (+12%) | 1.8 (+20%) | 2.1 (+40%) | 1.4 (-7%) |
| C3-P | 2.3 (-21%) | 2 (-31%) | 1.3 (-55%) | 2.7 (-13%) | 2.7 (-4%) | 2.1 (-32%) | 2.4 (-14%) |
| C4-P | 4.7 (-4%) | 4 (-18%) | 2 (-59%) | 4.9 (0%) | 4.9 (+4%) | 4 (-20%) | 4.7 (0%) |
| C3-C | 1.5 (+25%) | 2.2 (+69%) | 0.4 (-69%) | 1.2 (-4%) | 1.8 (+20%) | 1 (-33%) | 2.1 (+17%) |
| C4-C | 2 (+18%) | 3.8 (+124%) | 0.6 (-67%) | 2 (-5%) | 2.5 (+4%) | 1.7 (-23%) | 3.8 (+52%) |
| C3-CI | 1.3 (0%) | 1.2 (-8%) | 1 (-23%) | 0.8 (0%) | 0.8 (0%) | 0.7 (-13%) | NA |
| C4-CI | 2 (+33%) | 1.6 (+7%) | 1.1 (-21%) | 0.8 (+14%) | 0.8 (+14%) | 0.7 (0%) | NA |
| Natural Pasture | 25.3 (-2%) 7.1 (-9%) | 24.1 (-7%) 5.9 (-24%) | 34.9 (+35%) 3.2 (-59%) | 26.6 (+2%) 7.6 (-5%) | 24.6 (-2%) 7.6 (+1%) | 29.4 (+15%) 6.1 (-26%) | 23 (-12%) 7.1 (-5%) |
| Total Crop | 39.1 (-1%) | 38.8 (-2%) | 41.2 (+4%) | 39 (0%) | 38.1 (0%) | 39.7 (+2%) | 36.1 (-4%) |
| Rainfed Crop | 3.4 (+13%) | 6 (+100%) | 1.1 (-65%) | 3.2 (-11%) | 4.3 (+10%) | 2.8 (-22%) | 5.9 (+40%) |
| Irrigated | 3.3 (+14%) | 2.8 (-3%) | 2.1 (-25%) | 1.6 (+14%) | 1.6 (+7%) | 1.5 (+7%) | NA |
| Woody | 22.3 (-1%) | 21.2 (-6%) | 30.6 (+36%) | 23.2 (+3%) | 21.5 (-4%) | 25.6 (+15%) | 20.4 (-10%) |
| Tropics | 24.8 (+1%) | 24.7 (0%) | 25.7 (+4%) | 24.4 (+2%) | 24.3 (+1%) | 24.7 (+3%) | 22.9 (-2%) |
| Boreal | 2.1 (+17%) | 2.1 (+17%) | 2.1 (+17%) | 2.1 (+17%) | 2 (+11%) | 2.1 (+17%) | 2 (+5%) |
| Temperate N | 11 (-6%) | 10.8 (-8%) | 12.2 (+4%) | 11.4 (-3%) | 10.7 (-9%) | 11.8 (+1%) | 10 (-11%) |
| Temperate S | 1.2 (-14%) | 1.2 (-14%) | 1.3 (-7%) | 1.2 (-8%) | 1.1 (-15%) | 1.2 (-8%) | 1.1 (-15%) |

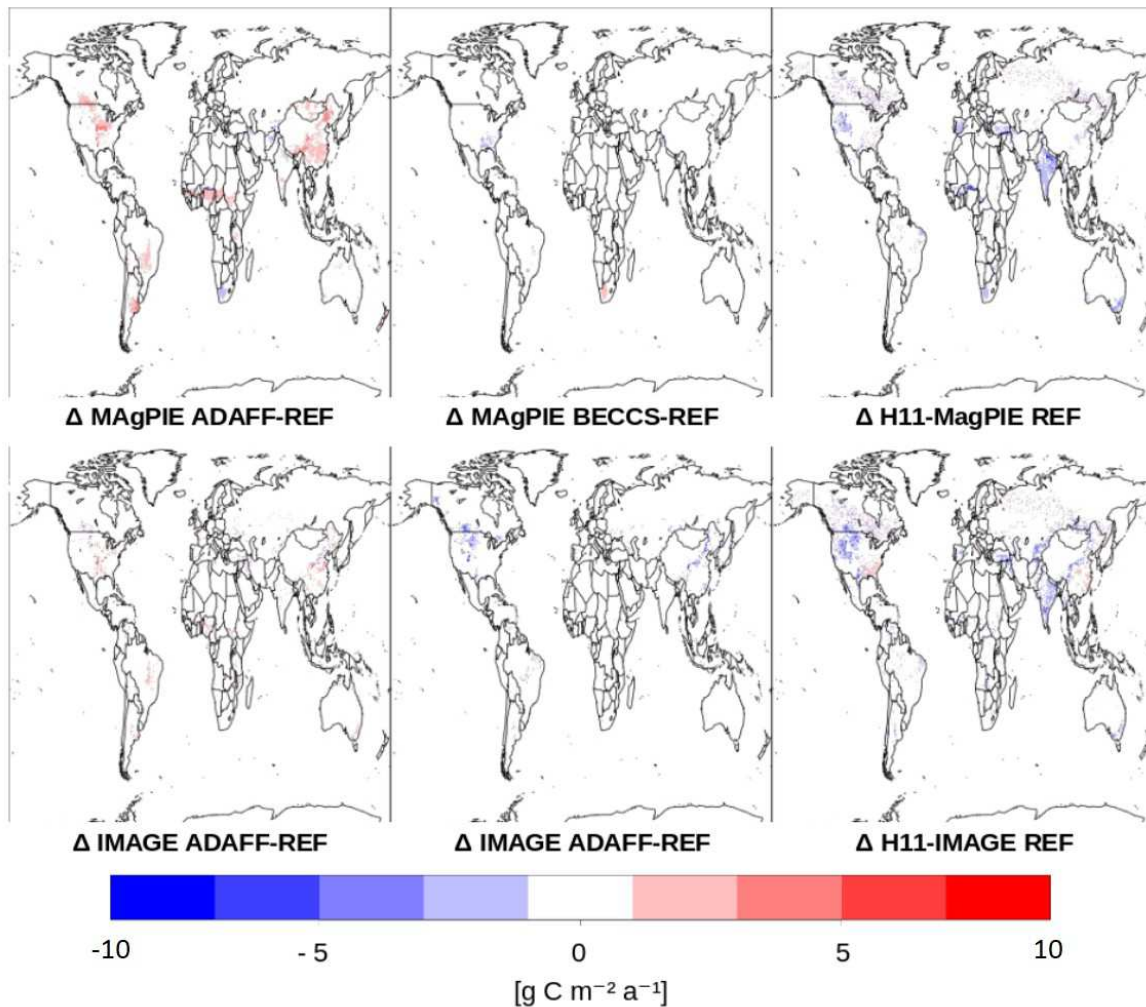
5

1 Figures 4 and 5 show isoprene and monoterpene emission difference maps for all scenarios. For
 2 isoprene, the clearest difference emerges between the ADAFF and the REF scenario, in particular for
 3 MAgPIE. Especially in the tropics the emission differences can be large, with up to $15 \text{ g C m}^{-2} \text{ a}^{-1}$
 4 higher emissions in the ADAFF scenario and reflecting the high emissions simulated for tropical
 5 regions (Guenther et al., 1995, Arneth et al., 2011). Differences between the ADAFF scenario and the
 6 reference cases for monoterpenes were lower and more broadly distributed between tropical and
 7 temperate regions for the MAgPIE case in which differences were up to $0.6 \text{ g C m}^{-2} \text{ a}^{-1}$. By contrast to
 8 ADAFF, differences between BECCS and REF were small for both isoprene and monoterpenes, and for
 9 both MAgPIE and IMAGE. This may be due to the fact that in this study bioenergy crops are only
 10 simulated as grasses, while also woody bioenergy crops could be used in reality. Those areas that
 11 show up in Figures 4 and 5 tend to show small decreases for the BECCS scenarios. When comparing
 12 H11 and REF landcovers, differences were visible over larger areas compared to the BECCS-Ref case,
 13 especially for monoterpenes.



14

15 *Figure 4: Isoprene emission differences (Δ) between the scenarios Adaff-Ref, Beccs-Ref and H11 - Ref*
 16 *of both MAgPIE and IMAGE. Mean values for the period 2081-2100 [$\text{g C m}^{-2} \text{ a}^{-1}$].*



1

2 *Figure 5: Monoterpene emission differences (Δ) between the scenarios Adaff-Ref, Beccs-Ref and H11 -*
 3 *Ref of both MAgPIE and IMAGE. Mean values for the period 2081-2100 [$\text{g C m}^{-2} \text{a}^{-1}$].*

4 Overall, the larger the amount of agricultural area fraction on global scale, the lower are the
 5 emissions of BVOCs. The difference between managed area fractions of the scenarios and the
 6 differences in global emissions between the scenarios have a strong negative correlation as indicated
 7 by the Pearson coefficient (Table 3), with values between -0.92 and -0.98 for isoprene and -0.8 and -
 8 0.94 for monoterpene emissions. This correlation is stronger for isoprene than for monoterpene
 9 emissions. Regarding regional differences, the correlations are strong for the tropical region, with
 10 values \sim -0.98 for both isoprene and monoterpenes, compared to values in the boreal region where
 11 correlations for both isoprene and monoterpenes are between -0.6 and -0.64 (not shown). The
 12 impact of LULCC on the emission of BVOCs seems bigger in the tropics than in other regions.

13 *Table 3 Pearson Correlation Coefficient between the global extent of area under agriculture and the global sum of Isoprene*
 14 *and Monoterpene emissions.*

| | MAgPIE Ref | MAgPIE Beccs | MAgPIE Adaff | IMAGE Ref | IMAGE Beccs | IMAGE Adaff | H11 |
|-------------|---------------|-----------------|-----------------|--------------|----------------|----------------|-------|
| Isoprene | -0.97 | -0.98 | -0.92 | -0.97 | -0.97 | -0.97 | -0.98 |
| Monoterpene | -0.91 | -0.91 | -0.80 | -0.89 | -0.91 | -0.85 | -0.94 |

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4. Discussion

So far, the number of studies that focused on the emission change of BVOCs due to LULCC, with or without also considering climate change is relatively limited. Lathière et al. (2010) worked with historical land-cover change data based on maps by Loveland et al. (2000), Ramankutty and Foley (1999) and Goldewijk (2001), on which basis emission changes were calculated using the Sheffield Dynamic Global Vegetation Model (SDGVM). Combining the effect of CO₂ inhibition, climate dynamics and LULCC, they showed a decline in isoprene emissions by ~24% from 1901-2002 (Lathière et al. 2010), and similar to our study, they found the highest impact of cropland expansion on isoprene emission in tropical regions, such as the Amazonian region, central Africa and south-east Asia, but also to some extent in central Europe and parts of the eastern USA. Unger (2013) used a biochemical model of isoprene emission within a global chemistry-climate framework to quantify the impact of LULCC, increasing CO₂-concentration and climate change on the emission of isoprene. She found a global decrease of isoprene emissions between 1880 and 2000 of 20% (from 534 to 449 TgCa⁻²), which is of similar magnitude to the earlier studies. Later Unger (2014a, 2014b) shows the same effects for BVOCs in general. In a literature review of the impact of LULCC on atmospheric composition and climate, Heald and Spracklen (2015) estimate the effect of LULCC from the preindustrial until today as a decrease of 15-36% in global isoprene emissions. In earlier work, Lathiere et al. (2006) used the DGVM ORCHIDEE combined with the Guenther et al (1995) BVOC emissions model, and satellite-based present-day climate from the International Satellite Land-Surface Climatology Project, ISLSCP-II. They found under otherwise similar conditions a decline in global isoprene and monoterpene emissions each by ~-30% in response to a tropical deforestation scenario, and very small changes only (+4% and -0.1% for isoprene and monoterpene, respectively) in response to an European afforestation scenario (although in case of the latter, changes when only the European simulation domain was considered were massive). Our results of a ~21% decline during the 20th century fall well within the previously published range.

For future land-use change effects, Wu et al. (2012) and Squire et al. (2014) used the IPCC A1B emission scenario. For land-use change, both studies were based on the IMAGE IAM, specified by Squire et al. (2014) to assume an increase in cropland area by 135% between 2001 and 2095. Despite of the land-use change, Wu et al. (2012) found an increase of isoprene emission by 8% for the period from 2000-2100, while our results for scenarios considering cropland expansion show either little change, or decreasing emissions (+/- 1% for BECCS and -5% for H11). For their simulations Wu et al. (2012) used climate data from the

1 GISS GCM3, combined with vegetation dynamics from LPJ DGVM (Sitch et al., 2003), and
2 using the MEGAN 2.0 framework for simulating the resulting BVOC emissions (Guenther et
3 al., 2006). The A1B CO₂ emissions pathway is higher than the RCP2.6 scenario used here,
4 however Wu et al. (2012) did not account for the CO₂ inhibition effect, which would act to
5 strongly reduce isoprene emissions relative to considering climate change alone (Arneeth et
6 al., 2007).

7 By contrast, a strong decrease in isoprene emissions by -41% over the 21st century was found
8 by Squire et al. (2014), who used the SDGVM to calculate the vegetation distribution, MEGAN
9 to calculate isoprene emissions, and climate projections from HadGEM1. Their simulation
10 setup combined SRES A1B LULCC and CO₂ emissions, and climate change as in the SRES B2
11 scenario. The large decline in isoprene emissions could be related to the fact that the authors
12 accounted for CO₂ inhibition in leaf-emissions. The large cropland expansion in their
13 simulations, together with the higher CO₂ concentrations in SRES B2 (comparable to a RCP4.5
14 pathway; van Vuuren et al. 2011) than used here are likely the chief reasons for the larger
15 emission reduction compared to our study. Likewise, Ganzeveld et al. (2010), applying the
16 EMAC chemistry/climate model with the Guenther et al. (1995) emission algorithms,
17 simulated a 12% decline in isoprene until 2050 when a SRES A2 land cover change scenario
18 was applied, without considering direct CO₂ inhibition or vegetation interacting dynamically
19 with climate change.

20 In a recent study, Hantson et al. (2017) also used the LPJ-GUESS modelling framework but
21 focusing on the effects of LULCC on the emission of BVOCs using land-use scenarios from
22 Hurtt et al. (2011). Across a broad range of LULCC (using the RCP4.5 climate and CO₂
23 concentrations) they identified large effects of LULCC on the emission of BVOCs, especially
24 for isoprene compared to a simulation with natural vegetation dynamics. Their results for
25 RCP2.6 LULCC show relatively similar dynamics to those we found here for H11 but with the
26 decline in isoprene emissions even bigger, likely due to the higher CO₂ concentration in
27 RCP4.5. Heald and Spracklen (2015) estimate a decline in isoprene emissions for the 21st
28 century due to the combined effects of increasing CO₂ concentration in the atmosphere and
29 anthropogenic LULCC of 24-27%.

30 Clearly, the studies available to date on future land-cover change effects on BVOC emissions
31 vary notably in their simulation set-up but unanimously show that BVOC emissions are
32 closely linked not only to climate and CO₂, but also to LULCC. BVOC emissions in regions
33 under cropland expansion decline, and all studies emphasize the crucial role tropical forests
34 play as main emitters of BVOCs on the one hand and as highly threatened regions by
35 agricultural expansion on the other.

1 Not surprisingly, effects of avoiding deforestation combined with afforestation had largest
2 effects on BVOC trajectories, because of the high emission potentials of woody vegetation
3 and because the difference in vegetation cover (compared to e.g., REF and BECCS) is quite
4 pronounced in tropical regions. As part of the current political debate, in particular after the
5 Paris COP21 agreement it is to be expected that land-based mitigation will become an even
6 larger contributor to land-use change than so-far assumed. While the scenarios we used here
7 are stylized and not calculated within a full Integrated Assessment framework they still serve
8 to highlight the potential effect of avoided deforestation and reforestation vs. BECCS on non-
9 greenhouse trace gas exchanges. In all scenarios we can demonstrate large effects on the
10 amount of BVOCs emitted, related to the total area under management, with less managed
11 area fraction on global scale relatively increasing BVOC-emissions globally and vice versa. But
12 especially for the bioenergy crops it is important to consider which type of vegetation is
13 used. In our scenario we assumed that bioenergy would be crop-based and used therefore a
14 plant functional type with low BVOC emission potential (Guenther et al, 1995, Arneth et al.,
15 2007, Schurgers et al., 2009, 2011). However, in tropical regions, for instance, oil palm is a
16 widely used bioenergy plant, which has an emission potential as high, or even higher than
17 tropical rainforests (Hewitt et al., 2009, Misztal et al., 2011, Silva et al., 2016). Likewise,
18 Ashworth et al. (2012, 2013) simulated for Europe the emissions from poplar plantations
19 used as bioenergy. Poplar is a strong isoprene emitter, and the authors found that the
20 planting of ca. 70 Mha of cropland with bioenergy crops such as poplar (or eucalyptus) would
21 result in an increase in isoprene emissions by roughly 4.5 Tg C a^{-1} . These studies
22 demonstrate that the total area undergoing change towards bioenergy (or other land uses) is
23 a large source of uncertainty for future BVOC emissions – and their effect on atmospheric
24 chemistry - but also that the type of vegetation grown in a certain region will affect emissions
25 substantially. For instance, if we compare our emission rates for C3 grasses, which we used
26 to calculate emissions from bioenergy crops to emission rates of bioenergy crops like short
27 rotation coppice (SRC) willow, the emissions in both BECCS scenarios would be significantly
28 higher. A recent study by Morrison et al. (2016) found isoprene emission factors of willow
29 growing at different locations in England between 0.1 and $16 \mu\text{g g}^{-1} \text{ h}^{-1}$, the upper value being
30 identical to emission factors we used for C3 crops ($16 \mu\text{g g}^{-1} \text{ h}^{-1}$). However, willow emission
31 factors well above $100 \mu\text{g g}^{-1} \text{ h}^{-1}$ were found in other studies (see Table 6, Morrison et al.,
32 2016). Since the calculated emissions scale linearly with the applied emission factor
33 (Niinemets et al., 2010) our calculated rates would, for instance, double if we had applied an
34 emission factor of 32 rather than $16 \mu\text{g g}^{-1} \text{ h}^{-1}$. A more detailed analysis of how bioenergy

1 plantations affect BVOC emissions would require estimates of the areas of woody and non-
2 woody plants used as bioenergy, and where these are located.

3 The emission hotspot for isoprene is located in the tropics (Guenther et al., 1995), and it is
4 thus not surprising that also the emission difference between the different LULCC scenarios
5 is largest there. It can be expected that different future trajectories would also impact on
6 tropical atmospheric chemistry (Young et al., 2009, Makkonen et al., 2012, Barkley et al.,
7 2012). However, the form of these impacts is not as clear as was once believed; work over
8 the last decade has revealed significant gaps our understanding of isoprene oxidation and
9 secondary organic aerosol formation (e.g. Lelieveld et al., 2008; Kiendler-Scharr et al., 2009;
10 Fuchs et al., 2013), as well as background chemical conditions over remote areas (Liu et al.,
11 2016). What can be expected with reasonable confidence is that increased BVOC emissions
12 in or near areas with relatively high background levels of NO_x will lead to increased ozone
13 formation. Such increases have been simulated previously for enhancements in isoprene
14 emissions due to land-use change (Chen et al., 2009; Ashworth et al., 2012, 2013; Wu et al.,
15 2012) and the chemistry is relatively well understood (Sillman, 1999). In environments with
16 low NO_x concentrations, such as the remote tropical rainforest, increased BVOC emissions
17 are expected to have the opposite effect, leading to decreased ozone concentrations (Hewitt
18 et al., 2009). Uncertainties in the isoprene oxidation pathway under low- NO_x do not appear
19 to have a large impact on projected changes in ozone concentration (Pugh et al., 2010), but
20 recent evidence suggests that there may be higher levels of NO_x in some of these remote
21 environments than previously thought (Liu et al., 2016), altering the effect of changed BVOC
22 emissions on ozone. Further, in our simulations we did not yet simulate NO_x emissions from
23 changing land-use, which we argue would be an important additional factor in assessing
24 land-use change/air quality interactions, as NO_x emissions will be enhanced in fertilized soils.
25 Combined BVOC/soil NO_x analyses thus will need to be done when assessing the potential
26 ozone formation effects of LULCC. For instance, Ganzeveld et al. (2010), in their study with
27 EMAC, investigated not only changes in isoprene but also altered soil NO_x emissions,
28 resulting in an increase in boundary layer ozone up to 9 ppbv over deforested areas in Africa
29 between present day and 2050 due to LULCC.

30 Methane lifetime has previously been strongly linked to BVOC emissions. As mentioned in
31 the introduction, however, the uncertainty in the level to which the hydroxyl radical is
32 regenerated in the isoprene oxidation chain (Lelieveld et al., 2008; Fuchs et al., 2013)
33 precludes clear statements as to the influence of changed BVOC emissions on methane
34 lifetime (e.g. Archibald et al., 2011). Further, the formation of secondary organic aerosol
35 (SOA) that takes place from oxidation of monoterpenes (Spracklen et al., 2008, 2011, Carslaw

1 et al., 2010, 2013) and isoprene (Claeys et al., 2004, Carlton et al., 2009) will also be affected
2 by LULCC, in particular if afforestation or avoided deforestation takes effect in conifer-
3 dominated vegetation of boreal and temperate regions. But very large uncertainties remain
4 regarding formation rates of SOA, for instance, isoprene has been found to inhibit new
5 particle formation in some studies (Kiender-Scharr et al., 2009, 2012). Moreover, climate
6 change has the potential to change the spectrum of different monoterpenes emitted
7 towards chemical species with higher (or lower) atmospheric reactivity (Jardine et al., 2017),
8 and the same can be speculated for land-cover change. Such a change in monoterpene
9 speciation would affect e.g., SOA formation and growth but is not considered in our
10 monoterpene emission algorithm. We therefore refrain from speculating on the implications
11 of our results for methane lifetime and SOA formation, recommending that dedicated
12 chemistry-transport studies are carried out to assess these important implications.

13
14 In summary, future pathways of LULCC highly affect the emission of BVOCs. As BVOCs play a
15 crucial role in atmospheric composition, a broader research focus is necessary, compared to
16 the few studies available to date. It seems evident that cropland expansion leads to
17 decreasing emissions, while afforestation leads to increasing emissions. However, not only
18 the impact of LULCC on the emissions itself is crucial, but also the choice of agricultural crop-
19 types, as well as dynamic vegetation response of natural vegetation to changing
20 environmental conditions. Future BVOC emissions and their impacts on chemistry-climate
21 interactions can only be assessed conjointly with dynamic vegetation responses.

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1 Appendix:

2 A: Supplementary Figures:

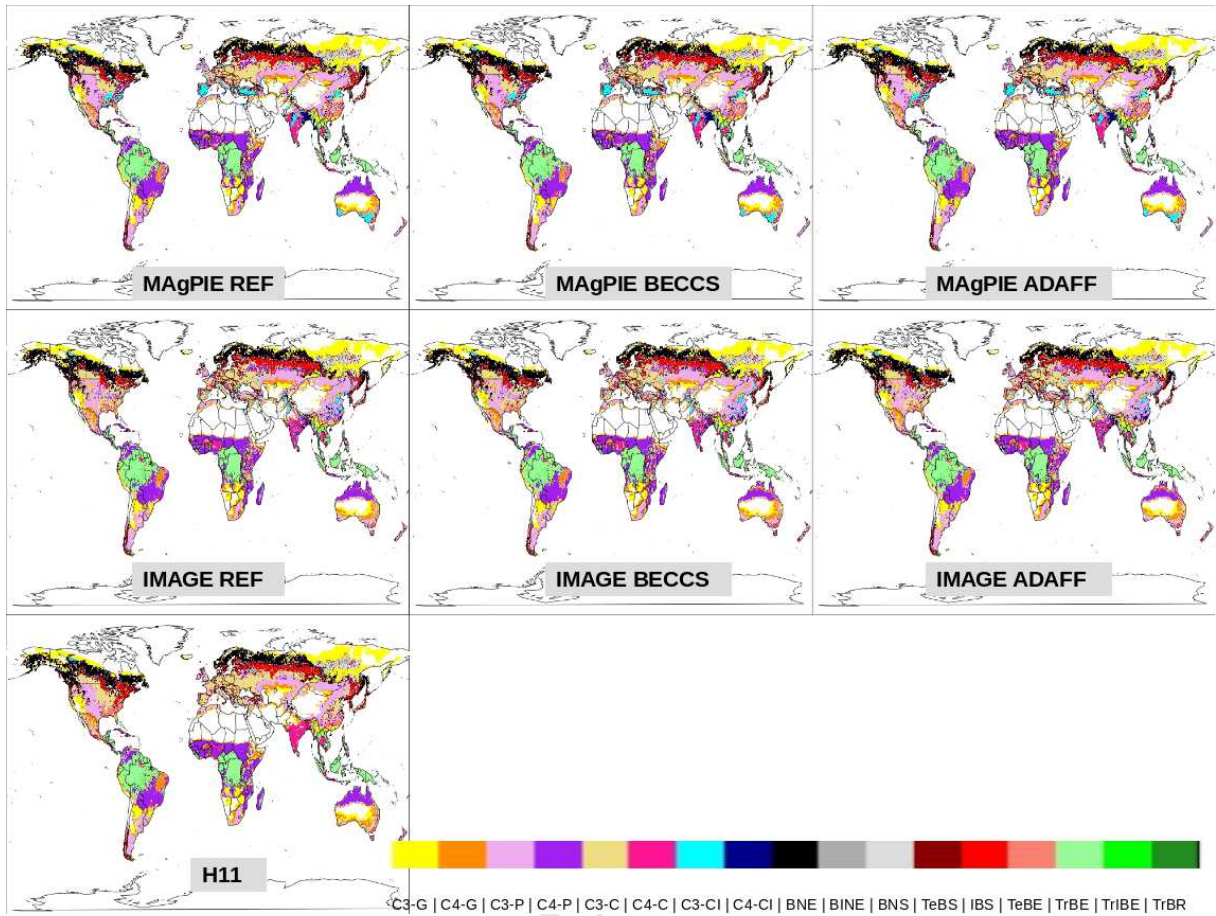
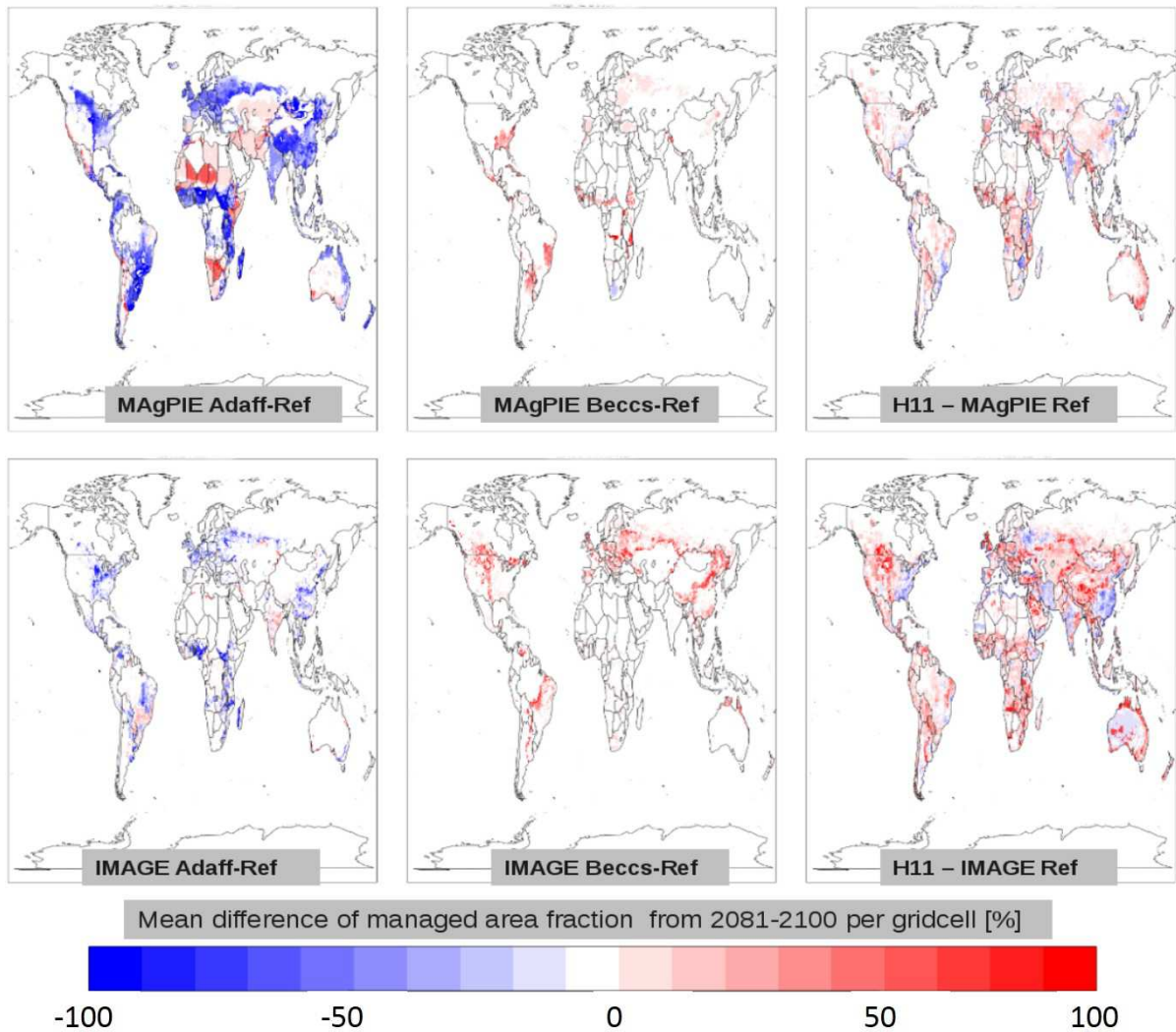


Figure 6: Maps showing present day distribution of the dominant PFTs per gridcell (gridcells show PFTs with maximum LAI). Mean values for the period 1981-2000 for all seven different model projections. Abbreviations are as in Figure 1.



1
 2 *Figure 7: Managed Area Fraction [%] - mean values for the period 2081-2100. Differences between*
 3 *the scenarios ADAFF-REF, BECCS-REF and H11-REF for both IAMs. Example equation for the*
 4 *calculation: Gridcell x (x:n) in MAgPIE ADAFF = 50% (0.5) area managed, Gridcell x (x:n) in MAgPIE*
 5 *REF = 30% (0.3) area managed. $0.5 - 0.3 = 0.2$ (20%) = higher extent of area managed in*
 6 *ADAFF than in REF for Gridcell x.*

7

8 **B: Method-Description – Land-use Datasets:**

9 **IMAGE3.0:**

10 IMAGE is an integrated assessment model framework that simulates global and regional
 11 environmental consequences of changes in human activities (Stehfest *et al* 2014). The model is a
 12 simulation model that uses a set of linked sub-models describing the energy system, the agricultural
 13 economy, land use, natural vegetation and the climate system. Most of the socio-economic
 14 parameters are modelled for 26 regions and most of the environmental parameters are modelled on
 15 a geographic grid.

16 The land-use component of the IMAGE framework is driven by demand for agricultural commodities,
 17 most importantly food crops, animal products and bio-energy. Food crops and animal product
 18 demand are modelled on a regional basis by the agro-economic model MAGNET, which takes into

1 account scenario-specific land availability. MAGNET determines the level of agricultural
2 intensification due to pressures on land availability, for example from policies for biodiversity
3 protection or REDD (Overmars *et al* 2014). The demand for bio-energy is modelled by the energy
4 model TIMER. The land availability for bio-energy production is determined according to a set of
5 sustainability criteria (Hoogwijk 2004): only land that has been used for agriculture in the past but is
6 not required for food production anymore, or areas with low carbon density are available. The
7 IMAGE-core model implements the demands for the various agricultural products (7 irrigated crop
8 groups, 7 rain-fed crop groups, grassland and sugarcane, grass and wood for bio-energy), on a
9 geographic grid of 5' by 5' arc-minutes according to a set of empirically based allocation rules
10 (Stehfest *et al* 2014). Production of animal products is calculated using a detailed description of the
11 livestock sector (Bouwman *et al* 2005). The expansion of built-up area is a function of scenario-
12 specific population growth and urbanization. Forestry activities are also modelled on the grid,
13 including forest degradation and deforestation that cannot be explained by land use expansion for
14 agriculture. In addition, the IMAGE-core model internally calculates climate change using an adapted
15 version of the MAGICC 6.0 model, and a pattern scaling to derive grid-specific temperature and
16 precipitation. The gridded land use is aggregated to 30' by 30' arc-minute fractions for use in the
17 coupled dynamic vegetation model LPJml (Bondeau *et al* 2007). Using land use and climate data,
18 LPJml models the carbon and water cycles, crop yields and natural vegetation dynamics.

19 In strict climate mitigation scenarios aiming for a maximum warming of two degrees by the year
20 2100, bio-energy production in combination with carbon capture and storage (BIO CCS) is an
21 important mitigation measure. IMAGE considers three types of bio-energy: ethanol from sugarcane
22 and methanol from lignocellulosic feedstock (grass or wood). The energy model TIMER uses land
23 availability, cost-based competition with other energy carriers and a carbon price to determine final
24 demand for bio-energy. While bioenergy can also be based on residues (Daioglou *et al* 2015), here
25 the bioenergy demand can only be fulfilled by the three explicit feedstocks to achieve optimal
26 comparability between MAgPIE and IMAGE.

27 Carbon dioxide removal (CDR) from avoided deforestation and afforestation (ADAFF) is not
28 dynamically modelled in IMAGE 3.0. For this study, a stylized implementation is used to determine
29 the mitigation potential of ADAFF under different agricultural intensification pathways. A stepwise
30 approach systematically increases the potential in a set of scenario runs to identify the combination
31 of intensification measures that achieves a specific CDR target. First, degraded forest areas are
32 afforested. Secondly, the efficiencies of the livestock sector are gradually increased until maximum
33 productivity levels are reached. Thirdly, the yield gap in crop production is closed by gradually
34 increasing the intensity towards the maximum potential. Finally, the intensity of livestock pasture
35 use is gradually increased towards the maximum potential. The intensification pathways result in
36 avoided deforestation and abandonment of agricultural land that can be used for afforestation,
37 which is assumed to take place as natural regrowth of vegetation. Areas with high afforestation
38 potential are abandoned first to maximize cumulative CDR. Only biomes with considerable forest
39 cover are accounted as part of the afforestation flux (boreal forests, temperate forests, tropical
40 forests, savannahs).

41 **MAgPIE:**

42 MAgPIE is a global multi regional partial equilibrium model of the agricultural sector (Lotze-Campen
43 *et al* 2008, Popp *et al* 2010, 2014). Objective function of the model is the minimization of global costs
44 for agricultural production throughout the 21st century (5-year time steps) in recursive dynamic
45 mode (**Error! Reference source not found.**). The model is driven by demand for agricultural
46 commodities, which is calculated based on population and income projections for the 21st century

1 consistent with SSP2 (O'Neill *et al* 2015). The production of agricultural commodities is associated
2 with costs for labor, capital, fertilizer, technological change, transport, and land conversion. Demand
3 and costs enter the model at the level of 10 world regions. For meeting the demand, the model
4 endogenously decides, based on cost-effectiveness, about the level of intensification (yield-
5 increasing technological change), extensification (land-use change), and production relocation
6 (international trade) (Dietrich *et al* 2014, Schmitz *et al* 2012). The optimization process is subject to
7 various spatially explicit biophysical conditions, which are derived from the global crop growth,
8 vegetation, and hydrology model LPJmL (Müller and Robertson 2014, Bondeau *et al* 2007). For
9 instance, climate-induced changes in crop yields can alter cropland requirements, and hence
10 deforestation. Due to computational constraints, spatially explicit input (0.5 degree resolution) is
11 aggregated to 600 simulation units for the optimization process based on a k-means clustering
12 algorithm (Dietrich *et al* 2013).

13 Land types in MAgPIE include cropland, pasture, forest, built-up and other natural land. Each grid-cell
14 can hold multiple land types at the same time, which add up to the total cell area. Cropland consists
15 of 15 food/feed crop types (e.g. cereals, oil crops, roots), and two bioenergy crop types (woody and
16 grassy). Pasture holds land needed for animal grazing. Forest includes primary forest, modified
17 natural forest and plantations. Built-up area is static in MAgPIE. Other natural land consists of non-
18 forest natural vegetation, abandoned agricultural land and deserts. The distinction between forest
19 and non-forest natural vegetation is based on a carbon density threshold of 20 tC/ha (Hurtt *et al*
20 2011). The carbon density on abandoned agricultural land increases over time due to regrowth of
21 natural vegetation (Humpenöder *et al* 2014). As soon as the carbon density of re-growing vegetation
22 passes the threshold of 20 tC/ha, the respective area is shifted to the forest land pool.

23 Demand for bioenergy is exogenous. In this study, bioenergy demand is based on Popp *et al* (2011)
24 and enters the model as a global number. Given the global demand, MAgPIE endogenously derives
25 spatially explicit bioenergy production patterns based on cost-effectiveness (Popp *et al* 2011, Klein *et al*
26 2014). In MAgPIE, bioenergy demand can only be fulfilled by the production of dedicated grassy
27 and woody energy crops, i.e. residues from agriculture or forestry are not considered. Woody
28 bioenergy, however, is not deployed by the model in this study because grassy bioenergy features
29 higher yields at the same factor costs.

30 Putting a price on CO₂ emissions from deforestation increases the relative costs of cropland
31 expansion into forests compared to other land conversions (e.g. pasture to cropland). As the
32 objective function of MAgPIE is cost minimization, the model tries to avoid the conversion of carbon-
33 rich forests under a carbon pricing scheme. The same mechanism can be used to incentivize
34 afforestation if carbon dioxide removal is rewarded besides punishing CO₂ emissions (Humpenöder *et al*
35 2014, 2015). Afforestation is implemented in MAgPIE as managed regrowth of natural vegetation.
36 In this study, we use the same CO₂ price trajectory as in Humpenöder *et al* (2014).

37 Literature Appendix:

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Highlights:

- LULCC shows a huge effect on the emission of BVOCs.
- Expanding croplands may lead to decreasing BVOC emissions
- Afforestation may lead to increasing BVOC emissions.