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Projected global ground-level ozone impacts on vegetation under different emission and climate scenarios

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ACCMIP models and RCP scenarios

All models simulate gaseous tropospheric chemistry, with various degrees of complexity: from 16 species in CESM-CAM to 110 species in MOCAGE depending on the degree of representation of non-methane hydrocarbon (NMHCs) chemistry in their chemical scheme. The aerosol indirect effects, including interactions between aerosols and gas-phase chemistry, are represented in GFDL-AM3, GISS-E2-R and MIROC-CHEM (Table 1). For future stratospheric O_3 projections, the models are grouped into 2 categories: the first group includes models with interactive or semi-offline chemistry and the second group includes models with prescribed O_3. Some models (e.g. GFDL-AM3, GISS-E2-R, MIROC-CHEM and MOCAGE) include full stratospheric chemistry schemes, while CESM-CAM is based on a linearized O_3 chemistry (i.e. LINOZ scheme, McLinden et al., 2000) and UM-CAM uses the CMIP5 dataset (Cionni et al., 2011) to prescribe offline O_3 in the stratosphere.

All models simulate temporal and spatial evolution of anthropogenic and natural emissions at global scale such as black carbon (BC), organic carbon (OC), secondary organic aerosols (SOA), non-methane volatile organic compounds (NMVOC), ammonia (NH_3), nitrogen oxides (NOx) from soils and lightning, and carbon monoxide (CO) from oceans and vegetation (see details in Stevenson et al., 2012). Surface methane (CH_4) concentrations, with spatial variation, are prescribed in all models using data from the database of Coupled Model Intercomparison Project Phase 5 (CMIP5), except for GISS-E2-R, in which interactive wetlands emissions for future are used (Shindell et al., 2012). Methane burdens vary with the time slices in all models (Tables 2). Natural biogenic emissions are those given by MEGAN v2.1 inventory, i.e. Model of Emissions of Gases and Aerosols from Nature (Guenther et al., 2012). Biogenic VOCs (e.g. isoprene) depend on meteorological conditions and may be responsible for near-surface O_3 changes (Guenther et al., 2006; Lin et al., 2008; Sicard et al., 2009). Only GISS-E2-R incorporates climate-driven isoprene emissions, the most abundant and reactive biogenic VOC. The other models prescribe fixed biogenic emissions, e.g. based on constant present-day isoprene emissions for all simulations. GISS-E2-R has interactive isoprene but fixed soil NOx (Lamarque et al., 2013; Young et al., 2013). Possible changes in lightning activity with climate change are recognized to impact the lightning NOx emissions (LNOx), therefore most models use the parameterization by Price and Rind (1992) based on the simulated convective activity (e.g. Fiore et al., 2006; Williams, 2009; Lamarque et al., 2013).

The three RCPs scenarios are named for their nominal radiative forcing level by 2100 (Thomson et al., 2011; San José et al., 2016). The RCP2.6 and RCP8.5 scenarios provide the boundary conditions in terms of 2100 climate change, i.e. by 2100, global mean surface air temperature at the land and ocean ranges from 1.0 to 4.5°C relative to the mean temperature over the pre-industrial period (Lamarque et al., 2013). The rise in global mean surface temperature by the end of the 21st century is in the ranges 2.6-4.8°C (RCP8.5), 1.1-2.6°C (RCP4.5) and 0.3-1.7°C (RCP2.6) at 68% of confidence interval. The greenhouse gas concentrations for the RCPs and their extensions beyond 2100 are deeply described by Meinshausen et al. (2011).

Results

Detailed description of the surface ozone concentration and AOT40 spatial pattern

Figure 1 shows the mean annual O_3 concentration at the lower model layer (Table 1) while Figure 2 shows the AOT40 distribution from the ACCMIP models for the historical and RCPs simulations.

In **CESM-CAM**, the historical highest surface O_3 mean concentrations (35-55 ppb) and AOT40 (>30 ppm.h) are observed in the Northern Hemisphere (NH), in particular over Greenland, Canada, Alaska, Mediterranean basin, East Asia and Tibetan plateau (>50 ppb and 60 ppm.h, respectively). The lowest O_3 mean concentrations (15-30 ppb) and AOT40 (<10 ppm.h) are recorded in particular below latitude 15°N and in the Southern Hemisphere (SH). Under RCP2.6 scenario, the surface O_3 mean concentrations decrease worldwide with mean concentrations of 25-30 ppb above latitude 15°N, except over the Tibetan plateau (around 40 ppb) and of 10-20 ppb below latitude 15°N. The AOT40 values are lower than 10 ppm.h worldwide (Fig. 2). Under RCP8.5, the O_3 mean concentrations increase up to50 ppb above latitude 15°N and reach 65 ppb over the Tibetan plateau. The AOT40 values increase from 20 to 60 ppm.h in the latitude band 15-45°N in the Western Hemisphere (WH) and the highest increases occur in Northwestern America, Greenland, Mediterranean basin, Near East and East Asia. The highest AOT40 values (>70 ppm.h) are found over the Tibetan plateau, Greenland and Near East.

In **GFDL-AM3**, a larger extent of high surface O_3 mean concentrations (>40ppb) is observed, i.e. covering North and Central America, Europe, Near East, Africa and Asia (Fig. 1). The highest O_3 levels (>50 ppb) are simulated in high-elevation areas (Rocky, Appalachian and Himalaya Mountains and Tibetan plateau) and over deserts (Near East, Colorado, Nevada and Gobi). The highest AOT40 values (>70 ppm.h) are observed in the latitude band 15-45°N (e.g. over Greenland and Tibetan plateau, East U.S, Near East). The lowest surface O_3 mean concentrations (<30 ppb)

and AOT40 (<10 ppm.h) are observed in South America, Oceania and Indonesia. Under RCP2.6, the global O_3 mean concentration is around 30 ppb, except in West and Equatorial Africa and South Asia (>40 ppb). A slighter decrease is observed in South America, Oceania and Indonesia where low historical O_3 mean levels (<30 ppb) are recorded. The AOT40 values are below 10 ppm.h worldwide, except in West and Equatorial Africa and South Asia (>70 ppm.h). Under RCP4.5, the O_3 mean concentrations range from 30 to 45 ppb, except in Northern South America and Indonesia (<20 ppb) and over the Tibetan plateau (>50 ppb). The AOT40 values exceed 30 ppm.h in Alaska, Greenland, East Asia and exceed 70 ppm.h over the Tibetan plateau and in Africa. The O_3 mean concentrations reach 50-60 ppb worldwide under RCP8.5, except in South America, Oceania and Indonesia (<35 ppb). The highest increases are observed in NH, in particular in Northwestern America, Greenland, Near East and South Asia (>65 ppb). The highest AOT40 values (>70 ppm.h) are found in Northern America, Greenland, in the latitude band 15-45°N in WH, in Equatorial and South Africa. Only South America and Oceania have AOT40 <10 ppm.h.

In **GISS-E2-R**, the highest historical surface O_3 mean concentrations (>40 ppb) and AOT40 (>70 ppm.h) are observed in NH above latitude 30°N (e.g. Greenland, Tibetan plateau, Eastern U.S, Mediterranean basin) and in the well-industrialized areas, close to Beijing and Los Angeles (>60 ppb). The lowest O_3 concentrations (<15 ppb) are recorded in Amazon, African and over Indonesian rainforests as well as in Australian Western plateau (shrubland). AOT40 range from 30 to 40 ppm.h in Central Europe and Asia, Siberia and North America while the lowest AOT40 (<10 ppm.h) are found in SH (Fig. 2). Under RCP2.6, the strongest decreases in surface O_3 mean concentrations and AOT40 are simulated in NH above latitude 15°N, where high historical O_3 concentrations (>40 ppb) and AOT40 (>40 ppm.h) are reported. Under RCP4.5, the lowest surface O_3 mean concentrations (10-20 ppb) and AOT40 (<10 ppm.h) are observed below latitude 15°N. Compared to the historical mean, the O_3 mean concentrations are lower in Western Africa and South Asia. Under RCP8.5, the strongest increases in surface O_3 mean concentrations are observed in NH above latitude 15°N, in particular over Greenland and South Asia (>70 ppb). Above latitude 30°N, high AOT40 values (30-60 ppm.h) occur, and exceed 70 ppm.h over Greenland, Near East, South Asia and around the Mediterranean basin. The lowest O_3 levels (<20 ppb) and AOT40 (<10 ppm.h) are reported in South America and Oceania. The O_3 burden is higher in NH while no significant change is simulated in SH as compared to historical data.

In **MIROC-CHEM**, the surface O_3 mean concentrations follow a clear latitudinal gradient, i.e. the highest mean concentrations (40-50 ppb) are observed in the latitude band 15-45°N, the range is 30-

40 ppb in the latitude band 45-60°N and <20 ppb above latitude 60°N. The concentrations are <30 ppb below latitude 15°N. AOT40 values, exceeding 30 ppm.h, are modelled in Eastern U.S, Central Europe and Africa, Near East and South Asia. Under RCP2.6, a homogeneous decrease is observed worldwide, i.e. above latitude 50°N, the O_3 mean concentrations are <20 ppb, as well as in South America, Oceania and Indonesia, and the AOT40 are <20 ppm.h worldwide. Slight changes are observed between the historical simulation and RCP8.5. The highest O_3 levels are simulated in Near East, Western U.S. and South Asia (45-55 ppb) while O_3 mean concentrations of 20-35 ppb occur worldwide. The mean AOT40 is <10 ppm.h worldwide, except in South Asia, Near East and a few locations in Africa (>65 ppm.h).

In **MOCAGE**, a heterogeneous spatial distribution of historical surface O_3 concentrations and AOT40 is found with a fine-scale resolution. The highest surface O_3 mean concentrations (>50 ppb) and AOT40 (>70 ppm.h) are observed in particular in the latitude band 15-45°N. Higher O_3 metrics values are simulated over Greenland, deserts (e.g. Gobi, Saudi Arabia), well-industrialized areas of North America and South Asia (e.g. Eastern U.S, Beijing) and at high-elevation sites (e.g. Summit in Greenland, Himalaya, Appalachian Mountains). The lowest concentrations (<20 ppb) are recorded in South America and Oceania while lower AOT40 (<10 ppm.h) are also observed in Northwestern America, Scandinavia, North Asia and along the African coastline. Under RCP2.6, the highest decreases in O_3 mean levels and AOT40 are reported where higher historical O_3 mean concentrations (40-60 ppb) and AOT40 (>70 ppm.h) are simulated, i.e. in the latitude band 15-45°N in WH, in North America and Greenland. Compared to the historical mean, no significant change is observed for the rest of the world. Under RCP4.5, the global O_3 mean levels (30-40 ppb) and AOT40 values (10-20 ppm.h) are lower than historical observations in NH, except in Canada, Greenland and East Asia where deterioration is observed compared to the historical time period. For mean concentrations and AOT40, no significant change is found in SH as compared to the historical simulation. Under RCP8.5, all the HN is strongly impacted by the increase in O_3 mean levels and AOT40. The highest O_3 mean concentrations (55-65 ppb) and AOT40 (>70 ppm.h) are observed in Greenland, Near East and across Asia.

In **UM-CAM**, the highest ground-level O_3 mean concentrations are simulated over deserts and remote areas (i.e. Greenland, Rocky Mountains, Sahara, Near East and Gobi), in particular in the latitude band 15-45°N with O_3 concentrations ranging from 40 to 60 ppb. The lowest O_3 concentrations (<15 ppb) are simulated over Amazon. Lower AOT40 values (<20 ppm.h) are found worldwide, except over Greenland and in latitude band 15-45°N in the Eastern Hemisphere (>30

ppm.h). The highest AOT40 values (>70 ppm.h) are observed in Near East and over the Tibetan plateau. Similarly to MOCAGE under RCP2.6, the highest decreases in O_3 mean levels and AOT40 are reported where higher historical O_3 mean concentrations (40-60 ppb) and AOT40 (>70 ppm.h) are simulated, i.e. in the latitude band 15-45°N in WH, in North America and Greenland. Under RCP4.5, the decreases in O_3 mean levels and AOT40 occur, in particular, in the latitude band 15-45°N in WH and in Northwestern America. The Tibetan plateau is an O_3 hot-spot (>50 ppb). Under RCP8.5, as compared to historical data, the highest increase in O_3 mean levels (40-55 ppb) and AOT40 (>70 ppm.h) is reported in Northwestern America, Greenland and in the latitude band 15-45°N in WH, reaching 70 ppb over the Tibetan plateau. Compared to the historical mean, no significant change is observed below latitude 15°N (<30 ppb).

In summary, the highest hemispheric O_3 mean concentrations (30.6 ppb in SH, 42.9 ppb in NH) are observed in GFDL-AM3 while the lowest (20.4 ppb in SH, 31.4 ppb in NH) are found in MIROC-CHEM for historical simulations (Table 3a) and across all RCPs (Table 3b). Averaged over the domain, the hemispheric AOT40 means range from 0.2 ppm.h (CESM-CAM) to 4.7 ppm.h (GISS-E2-R) in SH, while higher hemispheric AOT40 means, from 9.8 ppm.h (MIROC-CHEM) to 36.8 ppm.h (GISS-E2-R), are simulated in NH (Tables 3a).

Model-to-model changes in ozone burden related to emission changes

For RCP2.6, compared to historical simulations, annual total emissions of CO, NOx, VOCs and global CH_4 burden decrease by 40%, 44%, 5% and 27%, respectively and LNOx increase by 7% (Table 2b). The highest reduction in overall O_3 concentration (-29%) is recorded in CESM-CAM (Table 3b) and related to stronger changes in total NOx (-53%) and no change in VOCs emissions. Lower reduction in O_3 concentrations (-13%) in MIROC-CHEM is related to a slight decrease in VOCs emissions (-7%) and to the lowest decrease in total NOx emissions (-36%).

For RCP4.5, for which data from fewer models are available, there is generally a tendency for O_3 precursor emissions to decrease (Table 2b). The exception is the GISS-E2-R model, where we observe an opposite tendency for CH_4 burden (+5%) and VOCs emissions (+7%) for RCP4.5 and a slight increase (+0.5%) in VOCs for RCP2.6.

For RCP8.5, the global O_3 mean concentration increases by 23% in MOCAGE and 22% in CESM-CAM and by 4% in MIROC-CHEM (Table 3b). For RCP8.5, total emissions of CO and NOx are lower by respectively 32% and 20% compared to 2000, while CH_4 burden has strongly increased by

120% on average and LNOx emissions by 33%. For RCP8.5, VOCs emissions decreased by 1.3% on average, except for GISS-E2-R. The GISS-E2-R model shows a greater degree of variation than other models, with a stronger increase in CH_4 burden (+153%) and in VOCs emissions (+20%) for RCP8.5. In CESM-CAM, the strongest reduction in total NOx is found (-33%) while CH_4 (+112%) and LNOx (+30%) emissions strongly increased, and no change in VOCs emissions are observed over time (Table 2b). In MOCAGE, the strongest increase (+24%) in O_3 inputs is found (Table 3b). In MIROC-CHEM, VOCs (-3%) and NOx (-7%) emissions slightly decrease while an upward trend in CH_4 emissions (+116%) is simulated between 2100 and the early 2000s (Table 2b).

The decreases in AOT40, averaged over the whole domain, range from -97% in CESM-CAM to -54% in MOCAGE under RCP2.6 (Table 3c). Noting that MOCAGE and UM-CAM simulate an increase in AOT40 values in SH under RCP2.6 (+69 % and +92%) and RCP4.5 (+203% and +8%) scenarios. Under RCP8.5, an increase in averaged AOT40 is simulated in all models for both hemispheres (Table 3c), from +20% (MIROC-CHEM) to +138% (CESM-CAM).

Potential ozone impact on photosynthetic carbon assimilation under RCPs

Figure 3 shows the percentage of variation of IO3, i.e. the change in the potential impact of O_3 on vegetation for the ACCMIP models computed comparing the RCPs simulations with historical runs.

In **CESM-CAM**, compared to the historical mean, IO3 is homogenous worldwide under RCP2.6 with a strong decrease (more than -80%) above latitude 15°N and no significant change in Southeastern U.S., Central Asia and regions where AOT40 and O_3 mean concentrations are the lowest, i.e. South America, Africa and Australia in SH. Under RCP8.5, a strong increase in IO3 (more than +80 %) is simulated worldwide, except over Australia where no change is observed.

In **GFDL-AM3**, IO3 strongly decreases (more than-80%) in NH, except in South Asia (between -10 and -50%) under RCP2.6.Under RCP4.5, IO3 ranges from -15 to +15% above latitude 50°N, in South and Northeastern Asia, over Amazon and South Africa. The strongest percentages of change are observed in Equatorial Africa. IO3 is lower than -80% in the rest of the world (U.S., Europe, Central and Southeastern Asia). A larger extent of high IO3 percentage (>+60 %) is found under RCP8.5worldwide. However, lower increases (around +15%) are simulated in Southern Europe, Southeastern Asia and Central Africa while slight decreases (around -15 %) are observed over Amazon rainforests and in Eastern U.S. and China.

In **GISS-E2-R**, IO3 strongly decreases (more than-80%) in NH, except in South Asia (between -10 and -50%) under RCP2.6. Under RCP4.5, IO3 exceeds -80% in NH except in South Asia. No change occurs in South America and Australia while an increase in IO3 (>30%) is observed in Equatorial Africa. Under RCP8.5, the highest increase in IO3 (>+60%) is simulated across Africa, in Southeastern Asia and in Southwestern U.S. close to Los Angeles where O_3 mean concentrations exceeding 60 ppb are recorded. Lower increases (around +15 %) are simulated over Northwestern U.S. and in central and Northeastern Asia while a reduction in GPP is observed in Europe, East china and Eastern U.S (up to -70 %).

In **MIROC-CHEM**, no change in IO3 is observed worldwide except in South Asia and Equatorial Africa under RCP2.6. A strong decrease in IO3 is found under RCP4.5 in Eastern U.S, Central Europe and Southeastern China, close to Beijing surroundings. An increase of IO3 is simulated in Northern U.S., in South America, Asia and Africa.

In **MOCAGE**, the strongest decrease (more than -80%) under RCP2.6 occurs in Europe, East Asia and U.S., while a slight decrease or increase (between -15 and +15%) is found in Northern America, Southern and Northern Asia. Under RCP4.5, IO3 strongly increases (>+60%) above latitude 50°N. Across Africa, IO3 ranges from -15% to +60% over the rainforests. Slight decreases in IO3 (around -15%) are observed in Europe and South America (no change) while the strongest decrease (more than -60%) are found in Southern U.S., Southeastern Asia, South America. Under RCP8.5, the increase in IO3 is high worldwide, exceeding +80% above latitude 45°N, except in Central Europe where a slight decrease in IO3 (-15 %) is simulated. A reduction in IO3 (around -60 %) is found in Eastern U.S., Southeastern China and over the Amazon rainforests.

In **UM-CAM**, the strongest decrease in IO3 occurs in the latitude band 15-50°N in WH and Northwestern America under RCP2.6. No change in IO3 is found over latitude 50°N and in South America, Southeastern U.S. and Australia. Under RCP4.5, decreases in IO3 or no change (e.g. South America, Australia) occur worldwide, except in Africa where a few hot-spots (>+60%) are simulated. Slight increase in IO3 (around +15 %) is simulated in Europe and Central Asia under RCP8.5 and the increase in IO3 is high, exceeding +80%, above latitude 45°N, except in Central Europe where a slight decrease in IO3 (-15 %) is simulated.

In summary, averaged over the Globe, the highest decrease in the potential O_3 impact for vegetation, or risk factor IO3, is observed in CESM-CAM (-97%) while the lowest (-41%) is found

249 in MIROC-CHEM under RCP2.6 (Tables 3d). For RCP8.5, strong increases in IO3 are found for all
250 models and both hemispheres, from +21% (GISS-E2-R) to +130% (CESM-CAM). For all models,
251 generally a reduction in IO3 is simulated over Amazon rainforests (except in CESM-CAM), Central
252 Africa, Europe and East Asia, in particular under RCP8.5.

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