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Title:

Small global effect on terrestrial net primary production due to increased fossil fuel aerosol emissions from East Asia since the turn of the century

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Key points: Increase in FF aerosol emission over Eastern Asia appear not responsible for coincident increase in land carbon sink

Key words: Fossil fuel aerosol emission, Eastern Asia, aerosol optical depth, diffuse radiation, net primary production

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35 **Abstract**

36 The global terrestrial carbon sink has increased since the start of this century at a time of growing
37 carbon emissions from fossil fuel burning. Here we test the hypothesis that increases in atmospheric
38 aerosols from fossil fuel burning enhanced the diffuse light fraction and the efficiency of plant
39 carbon uptake. Using a combination of models, we estimate that at global scale changes in light
40 regimes from fossil fuel aerosol emissions had only a small negative effect on the increase in
41 terrestrial net primary production over the period 1998-2010. Hereby, the substantial increases in
42 fossil fuel aerosol emissions and plant carbon uptake over East Asia were effectively cancelled by
43 opposing trends across Europe and North America. This suggests that if the recent increase in the
44 land carbon sink would be causally linked to fossil fuel emissions it is unlikely via the effect of
45 aerosols but due to other factors such as nitrogen deposition or nitrogen-carbon interactions.

46
47 **1. Introduction**

48 Fossil fuel (FF) emissions of CO₂ have sharply increased since the turn of the century at a rate of
49 3% yr⁻¹, almost twice the rate of the prior three decades [*Hansen et al.*, 2013]. In contrast, global
50 atmospheric CO₂ growth rates were relatively constant during this period [*Ballantyne et al.*, 2012].
51 A coincident decline in land use carbon emissions [*Harris et al.*, 2012] as well as a moderate
52 strengthening of ocean carbon uptake [*Rödenbeck et al.*, 2014; *Le Quéré et al.*, 2015] may have
53 played a role but these contributions appear insufficient to explain the slow atmospheric growth rate
54 of CO₂, implying that terrestrial carbon sinks must have substantially increased in this period
55 [*Sarmiento et al.*, 2010].

56 The recent divergence of trends in carbon emissions and atmospheric CO₂ growth rates led
57 to speculations that key carbon sink processes may be strongly controlled by the increasing
58 emissions themselves, namely increased nitrogen deposition and a larger fraction of diffuse versus
59 direct solar radiation from predominantly increased sulfate aerosol emissions originating from East
60 Asia [*Hansen et al.*, 2013]. In regards to the latter, multiple studies have shown that the efficiency

61 of plant photosynthesis increases under more diffuse light conditions (e.g. resulting from increased
62 scattering of light by aerosols or clouds) since under such conditions radiation can penetrate deeper
63 into the canopy, illuminating previously shaded leaves [Roderick *et al.*, 2001; Gu *et al.*, 2003;
64 Mercado *et al.*, 2009]. However, these studies also show that a corresponding reduction in total
65 radiation may have a negative impact upon photosynthesis, whereby GPP tends to decline if the
66 diffuse fraction surpasses 0.4 [Mercado *et al.*, 2009]. The overall effect on photosynthesis and net
67 primary production (NPP) thus depends upon the balance between these two mechanisms. Recent
68 model results showed that increases in the fraction of diffuse radiation due to anthropogenic
69 aerosols in the period 1960-1999 (the global dimming period) enhanced the global carbon sink by
70 24% [Mercado *et al.*, 2009]. The extent at which the rapid increase in East Asian FF aerosol
71 emissions since the turn of the century may have impacted plant growth and the global carbon sink
72 is however not clear since anthropogenic aerosol emissions in Europe and United States have
73 decreased persistently since the late 1980s [Wild *et al.*, 2009].

74 Here we therefore test the hypothesis that an increase in the fraction of diffuse light
75 associated with increased FF aerosol emissions predominantly from East Asia have contributed to
76 increased global plant carbon uptake which would provide a mechanism for a potential link between
77 global carbon emissions and the land carbon sink. Using atmospheric models, including an aerosol
78 model with size-resolved aerosol microphysics, we first simulate aerosol distributions (originating
79 from fossil fuel and fires) and corresponding effects on light regimes over 1998 to 2010. We then
80 use these to drive a land surface model to estimate their relative contributions to changes in regional
81 and global NPP.

82

83 **2. Methodology**

84 The distribution of anthropogenic aerosols was simulated using a global aerosol model [Mann *et al.*,
85 2010]. The impact of aerosols and clouds on surface radiation was simulated using a radiative
86 transfer model [Edwards and Slingo, 1996]. Plant carbon uptake was simulated using a land surface

87 model [Best et al., 2011; Clark et al., 2011]. A similar combination of models has also been used in
88 a recent study by Rap et al. [2015].

89 **2.1 Aerosol model**

90 The aerosol distribution was simulated using the GLObal Model of Aerosol Processes (GLOMAP)
91 [Mann et al., 2010], which is an extension to the TOMCAT 3-D chemical transport model
92 [Chipperfield, 2006]. GLOMAP is a global aerosol microphysical model that simulates the
93 concentration, size, and mass of aerosol particles using a two-moment (mass per particle and
94 number concentration) modal scheme. This model includes various aerosol processes, including
95 nucleation, condensation, growth, coagulation, dry and wet deposition, and cloud processing. In the
96 GLOMAP version used here, the aerosol species included are black carbon (BC), particulate
97 organic matter (POM), sulfate, sea salt, and mineral dust. The horizontal resolution is $2.8^\circ \times 2.8^\circ$,
98 with 31 vertical levels ranging from the surface to 10hPa, with the layer thickness varying from 60
99 m (surface) to 1 km (tropopause). The model is driven with historical meteorology from the
100 European Centre for Medium-Range Weather Forecasts (ECMWF) at 6-hourly intervals and
101 interpolated onto the model time-step (30 minutes). Annually varying anthropogenic emissions
102 (BC, organic carbon (OC), SO₂) including fossil fuel and biofuel emissions are taken from the
103 MACCity inventory [Granier et al., 2011]. This dataset is based on historical ACCMIP (for years
104 1990 and 2000) and RCP 8.5 (2005 and 2010) emissions. The emissions were linearly interpolated
105 for the years between those given. Biomass burning emissions (BC, OC, SO₂) are taken from the
106 Global Fire Emissions Database version 3 (GFED3) [van der Werf et al., 2010] and are supplied as
107 annually varying monthly means.

108 GLOMAP has been evaluated extensively in previous work and generally found to match
109 ground-based station observations (e.g. AERONET) well [Mann et al., 2010; Reddington et al.,
110 2014, 2016; Rap et al., 2015]. In this study, we compared trends in simulated aerosol optical depth
111 (AOD) with satellite-based (MODIS, SeaWiFS) [Hsu et al., 2013; Platnick et al., 2015] estimates
112 for the period of overlapping data records 2001-2010. Results showed while there is generally good

113 agreement between the modelled and observed AOD trends in areas where fossil fuel emissions
114 dominate the AOD pattern which is the focus of this study (Figure 1 and Figures S1-S3 in the
115 supporting information), there are also notable differences in specific regions (e.g., Amazon Basin).
116 Some reasons for these discrepancies may involve the comparatively larger interannual variability
117 in the satellite AOD (Figure S2), requiring greater changes to be significant. In addition, the
118 GLOMAP 'baseline' AOD magnitudes tend to be somewhat lower than the satellite AOD (Figure
119 S2), therefore trends of equal size are more likely to be significant in the simulated AOD. In this
120 study we are interested in trends in AOD driven by changing anthropogenic aerosol emissions. To
121 exclude a contamination from dust, we calculate AOD only for the 4 aerosol size modes (aitken-
122 soluble, aitken-insoluble, accumulation-soluble, and coarse-soluble) that do not include dust. We
123 demonstrated that satellite aerosol trends are similar both during periods with and without a large
124 contribution from dust in East Asia
125 (Figure S1), demonstrating that observed trends are not due to trends in dust.

126

127 **2.2 Radiative transfer model**

128 The Edwards and Slingo [1996] radiative transfer model is used to quantify the aerosol effect on
129 direct and diffuse radiation [Rap *et al.*, 2013]. We used the aerosol optical properties (scattering,
130 absorption, and asymmetry coefficients) for each aerosol mode and spectral band based on Bellouin
131 *et al.* [2013]. The model is forced with monthly mean ECMWF climate (water vapour, temperature)
132 and ozone reanalysis data together with cloud fields and surface albedo from the International
133 Satellite Cloud Climatology Project (ISCCP-D2) [Rossow and Schiffer, 1999]. The simulated total
134 and direct radiation fluxes are used to calculate diffuse radiation (diffuse = total – direct). Due to
135 the uncertainty in aerosol-cloud interactions we do not allow changes in aerosol to alter cloud
136 properties (aerosol indirect effect). The Edwards-Slingo (ES) model has been validated in recent
137 studies to some extent [e.g. Rap *et al.*, 2015]. We performed additional validations at four FluxNet
138 (La Thuile ‘fair use’ database; <http://www.fluxdata.org>) sites in Europe and North America and also
139 found generally good agreement between observed and modelled light regimes, however at some of

140 the sites over-estimation of total radiation and under-estimation of diffuse radiation were apparent
141 (Figures S4 and S5). This may lead to an overestimation of the diffuse effect on NPP due to the
142 strong non-linear dependence of plant carbon uptake to changes in diffusivity [*Mercado et al.*,
143 2009].

144

145 **2.3 Land surface model**

146 The Joint UK Land Environment Simulator (JULES) land surface model used here simulates the
147 exchange of carbon, water, energy and momentum between the land surface and atmosphere [*Best*
148 *et al.*, 2011; *Clark et al.*, 2011]. The model includes a multilayer (10 levels) canopy
149 parameterization to scale photosynthesis from leaf to the canopy [*Mercado et al.*, 2007, 2009].
150 Photosynthesis is calculated at each level, and treats sunlit and shaded leaves separately. In our
151 simulations, we used the dynamic phenology (TRIFFID) version of JULES. To ensure the plant
152 pools and NPP are at steady state, the model was spun up for 60 years (10 in equilibrium mode and
153 50 in dynamical mode [see *Cox*, 2001]) using a repeated driver climatology for 1995. The control
154 simulation was then run with transient driving input for 1996-1998, providing a steady-state to start
155 our simulations from. The model is forced with ERA-Interim climate fields [*Weedon et al.*, 2014],
156 and runs at 0.5° spatial resolution with three hourly time steps. The climate drivers consist of 2m air
157 temperature, specific humidity, precipitation, 10m wind speed and surface pressure. Model drivers
158 also include downwards surface radiation (short-wave direct and diffuse, long-wave) from the ES
159 model. The JULES plant carbon uptake response to changes in solar radiation has also been
160 validated to some extent at temperate needleleaf and broadleaf forest sites [*Mercado et al.*, 2009]
161 and in tropical rainforests [*Rap et al.*, 2015]. We conducted further validations at the same four
162 FluxNet sites that were used in the ES validations (see above). Also in this case, the modelled GPP
163 responses to increases in PAR under both total and diffuse light regimes agree generally well with
164 observed responses (Figure S6).

165 We performed a set of factorial simulations with JULES over the period 1998-2010 to

166 isolate the impact of single drivers on NPP. The five drivers considered include (1) climate, (2)
167 atmospheric CO₂, and incoming solar radiation due to aerosols associated with (3) anthropogenic
168 emissions, (4) fire emissions as well as (5) cloud cover. We started with a 'control' simulation in
169 which only climate variables were varied and anthropogenic and fire aerosol emissions remained at
170 year 2000 values to avoid the anomalous 1998 ENSO year and atmospheric CO₂ was held fixed at
171 1998 levels whereas cloud cover was based on a climatology for whole study period 1998-2010.
172 Four additional simulations were carried out whereby in each simulation one additional driver was
173 varied, so that our final simulation had monthly varying fire emissions and cloud cover for the
174 whole period and anthropogenic emissions and the atmospheric CO₂ level varied annually. We first
175 calculated the trend (based on linear regression) in annual AOD, surface diffuse radiation (SDR),
176 and NPP for each simulation. The climate effect and combined effect can be inferred directly from
177 the first (only climate varied) and last (all drivers varied) model runs. To isolate the impact of the
178 remaining single drivers, the difference between the trends of two simulations that only differ by
179 that driver was used.

180

181 **3. Results**

182 The simulated impact of anthropogenic aerosol emissions on AOD and SDR from 1998 to 2010 is
183 shown in Figure 2. As anticipated, AOD changes were largest in regions of significant FF aerosol
184 emission change over this period. For example, East Asia show substantial increases in AOD and
185 SDR coinciding with increasing anthropogenic aerosol and aerosol precursor emissions [*Granier et*
186 *al.*, 2011]. In contrast, Europe and North America experienced declining AOD and SDR trends
187 driven by a reduction in FF aerosol emissions (Figure 2, Table 1 and Figure S3). The spatial
188 distribution of these trends in AOD and SDR are greatest close to the vicinity of the respective
189 source regions, although changes extend for thousands of km due to atmospheric transport of the
190 aerosols. Our results also show that changes in fossil fuel aerosol emissions play an important role
191 in the AOD trends compared to natural (e.g. sea spray) and fire induced changes in all three regions

192 of interest (Figure S3). A subsequent analysis that isolates the contribution of each factor (FF, fire,
193 and clouds) to trends in SDR further confirms this result, with fossil fuel burning also dominating
194 the trend in the three focus regions of East Asia, Europe and North America (Figure S7).

195 A factorial analysis based on multiple runs with the JULES land surface model (see
196 Methods) was used to quantify the contribution of single drivers (changes in light regimes due to FF
197 and fire emissions as well as changes in cloud cover, in addition to changes in near-surface climate
198 and increased atmospheric CO₂ concentrations) to the trend in NPP in the study period 1998-2010.
199 Results show that the spatial patterns in the overall NPP trends (Figure 3a) were generally
200 dominated by trends in near-surface climate (Figure 3b and Figure S8). In this regard, warming
201 across northern Eurasia and cooling across Canada appeared to be responsible for the pronounced
202 positive and negative NPP trends in these regions, respectfully (Figure 3b and Figure S9). Over
203 many land regions outside the northern high latitudes, trends in precipitation appeared to be the
204 dominant driver for trends in NPP (Figure 3b and Figure S9).

205 At more regional levels, changes in SDR associated with FF aerosols had a sizeable impact
206 on trends in NPP in East Asia, Europe, and Eastern USA (Figure 3c) broadly in line with the spatial
207 pattern of the corresponding AOD and SDR trends (Figure 2). Changes in NPP due to trends in
208 SDR resulting from changes in fire emissions and cloud cover were of similar magnitude but
209 displayed a more heterogeneous pattern across the continents (Figures 3d and 3e). Over central
210 African rainforests, a relatively strong cloud cover – SDR effect was observed, where a reduction in
211 SDR associated with a strong trend towards lower cloud cover (Figure S7) led to markedly lower
212 NPP. Conversely, and as expected, the CO₂ fertilization effect (Figure 3f) led to consistent increases
213 in NPP across most of the vegetated land surface, with the largest impact in the highly productive
214 tropics.

215 In Figure 4, regionally aggregated and global contributions from each single driver to the
216 overall NPP trends over the 1998-2010 study period are shown. Corresponding results show that
217 over East Asia, changes in climate (negative contribution) as well as atmospheric CO₂ (positive)

218 were the most dominant drivers of trends in NPP (Figure 4a). However, increases in SDR due to
219 increasing FF aerosol emissions caused a sizeable positive NPP trend (14 TgC yr^{-2} ; see also Table
220 1), which amounted to a substantial proportion (33%) of the total positive NPP trend over this
221 region. In Europe and North America, changes in climate and atmospheric CO_2 were generally also
222 the dominant drivers of NPP changes, whereas declining SDR (from decreasing FF aerosol
223 emissions) led to significant negative contributions to the overall NPP trends (Figure 4b and 4c;
224 Table 1). At global scale, we estimated an overall increasing NPP trend of 0.14 PgC yr^{-2} over the
225 study period 1998-2010 with changes in atmospheric CO_2 (0.25 PgC yr^{-2}) and near-surface climate
226 ($-0.09 \text{ PgC yr}^{-2}$) playing a dominant role (Figure 4d). At this global level, the aerosol radiative
227 effects from changes in FF emissions are relatively small (-6.8 TgC yr^{-2} , -4.9% of total NPP trend)
228 since the increasing contributions over East Asia are effectively cancelled out by the declining
229 contributions from Europe and North America.

230

231 **4. Discussion**

232 Our results suggest that the simulated increase in global NPP (0.14 PgC yr^{-2}) over the period 1998
233 to 2010 is largely driven by increasing atmospheric CO_2 , through a combination of direct CO_2
234 fertilization and the indirect effects of improved water use efficiency in line with previous model
235 studies [Schimel *et al.*, 2015; Sitch *et al.*, 2015]. The dominant contribution of the CO_2 fertilization
236 effect on trends in NPP should however be viewed with caution as more recent studies showed that
237 land surface models may overestimate corresponding impacts considerably [Brienen *et al.*, 2015;
238 Smith *et al.*, 2015]. At the global scale, climatic trends over this period contributed negatively to
239 changes in global NPP consistent with results based on a more data-constrained approach [Zhao and
240 Running, 2010].

241 Radiative effects associated with aerosol emissions from FF and fire activity and those
242 related to clouds on trends in NPP played only a minor role at global scale. Our results however do
243 show that at more regional levels, FF aerosol emissions and corresponding effects on diffuse

244 radiation are potent drivers of NPP changes, particularly over East Asia where they contribute 33%
245 to the total NPP trend. In this region, the recent trend in fossil fuel aerosol emissions are mainly
246 driven by increases in coal burning and associated sulfate aerosols [Lu *et al.*, 2010; Granier *et al.*,
247 2011]. Our results must be viewed with some caution since for example we did not consider
248 potential adverse effects of acidic sulfate deposition on NPP [Büntgen *et al.*, 2014] and the effect of
249 diffuse radiation on NPP at regional scales might be slightly overestimated due to a model bias (see
250 Section 2.2). But one important inference is that due to the importance of this ‘FF aerosol driver’
251 and the relatively short atmospheric lifetime of aerosols (days to weeks), a decline in regional-scale
252 FF aerosols (e.g. through implementing more strict air pollution standards) may reduce NPP and net
253 carbon uptake substantially at relatively short time scales.

254 Our findings presented here thus indicate that the marked post-2000 increase in the global
255 land carbon sink may not be explained by changes in light regimes resulting from coincident
256 changes in fossil fuel aerosol emissions and corresponding effects on NPP. This is to a large part a
257 result of the opposing contributions from Asia and from Europe and North America leading to a
258 relatively small global impact. This opens the door for investigations of alternative carbon sink
259 mechanisms that are causally linked to increasing FF emissions. In this regard, nitrogen deposition
260 may act as a potent driver through both its direct effect on photosynthesis, plant respiration and soil
261 respiration [Zaehle, 2013] as well as indirectly through easing nutrient constraints for NPP
262 enhancements via the CO₂ fertilization effect [Norby *et al.*, 2010]. In addition, decadal climatic
263 trends that are largely independent of FF emission trajectories may induce strong impacts on NPP
264 (as shown here) and also on plant and soil respiration. In this regard, the recent ‘hiatus’ in global
265 temperatures [IPCC 2013] may have reduced respiratory carbon fluxes thereby contributing to the
266 enhanced land carbon sink in this time frame.

267

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370

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Tables

Table 1: Trends in AOD, SDR, and NPP over the period 1998-2010 for global land and three focus regions. The linear trends shown are based on simulations in which all drivers are varied and where the effect of FF aerosol emissions is isolated (in parentheses). The three focus regions (only land areas) are outlined in Figure 2a and number of asterisks indicate statistical significance of trends at $P<0.05$ (*), $P<0.01$ (**) and $P<0.001$ (***) levels, respectively.

Region	AOD (yr^{-1})	SDR ($\text{W m}^{-2} \text{yr}^{-1}$)	NPP (TgC yr^{-2})
East Asia	0.0037*** (0.0035***)	0.31** (0.21***)	44.13* (14.44)
Europe	-0.0052*** (-0.0050***)	-0.56*** (-0.47***)	19.10 (-8.09)
North America	-0.0021*** (-0.0021***)	-0.16 (-0.18***)	17.32 (-9.78)
Global	-0.0002 (-0.0001)	-0.002 (-0.03)	140.13 (-6.82)

384 **Figure captions**

385 **Figure 1:** Comparison between modelled and satellite annual mean AOD trends (yr^{-1}) for the period
386 of overlapping data records 2001-2010. Panels depict linear trends for (a) GLOMAP, (b) MODIS,
387 and (c) SeaWiFS. In (d), linear trends in AOD (yr^{-1}) between 2001 and 2010 are shown for the three
388 focus regions (outlined in panel (a); land points only): Europe (EU), North America (NA), and East
389 Asia (EA) based on GLOMAP (green), MODIS (violet), and SeaWiFS (brown). The crosses
390 represent the mean trend, the middle bars the median, the boxes the 25th and 75th percentile values
391 and the error bars the minimum and maximum values with circles representing outliers (greater than
392 1.5 x interquartile range). White areas in (b) and (c) indicate regions where satellite retrievals were
393 not available and in all maps statistically significant ($P < 0.05$; Student's t-test) trends are highlighted
394 with stippling. Spatial resolutions in the original datasets differ between modelled (2.8°) and
395 satellite (MODIS (1.0°) and SeaWiFS (0.5°)), and for this comparison the satellite AOD fields were
396 aggregated to the coarser model resolution.

397
398 **Figure 2.** Spatial pattern of linear trends in simulated annual (a) AOD and (b) SDR due to changes
399 in fossil fuel aerosol emissions over the period 1998 – 2010. In (a) and (b), trends are calculated as
400 the difference in the trends based on two single simulations, with varying anthropogenic aerosol
401 emissions as the only difference between the two (see Methods). Statistically significant ($P < 0.05$)
402 trends are highlighted with stippling.

403
404 **Figure 3.** Spatial pattern of linear trends ($\text{gCm}^{-2}\text{yr}^{-2}$) in annual NPP for the period 1998-2010. The
405 maps depict trends in NPP based on factorial JULES simulations with (a) all drivers varied, and
406 corresponding to single drivers including (b) climate, as well as light regimes associated with (c)
407 fossil fuel aerosol emissions, (d) fire aerosol emissions and (e) cloud cover. Panel (f) shows trends
408 in NPP associated with atmospheric CO_2 . Statistically significant ($P < 0.05$) trends are highlighted
409 with stippling.

410 **Figure 4.** Global, regional and mechanistic attribution of trends in annual NPP for the period 1998-
411 2010. Trends are based on annual means of spatially aggregated NPP for the three focus regions (a)
412 East Asia, (b) Europe and (c) North America as well as for (d) all land regions. The three focus
413 regions are depicted in Figure 3a. Statistically significant ($P<0.05$) trends are highlighted (*).

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Figure 1. Figure

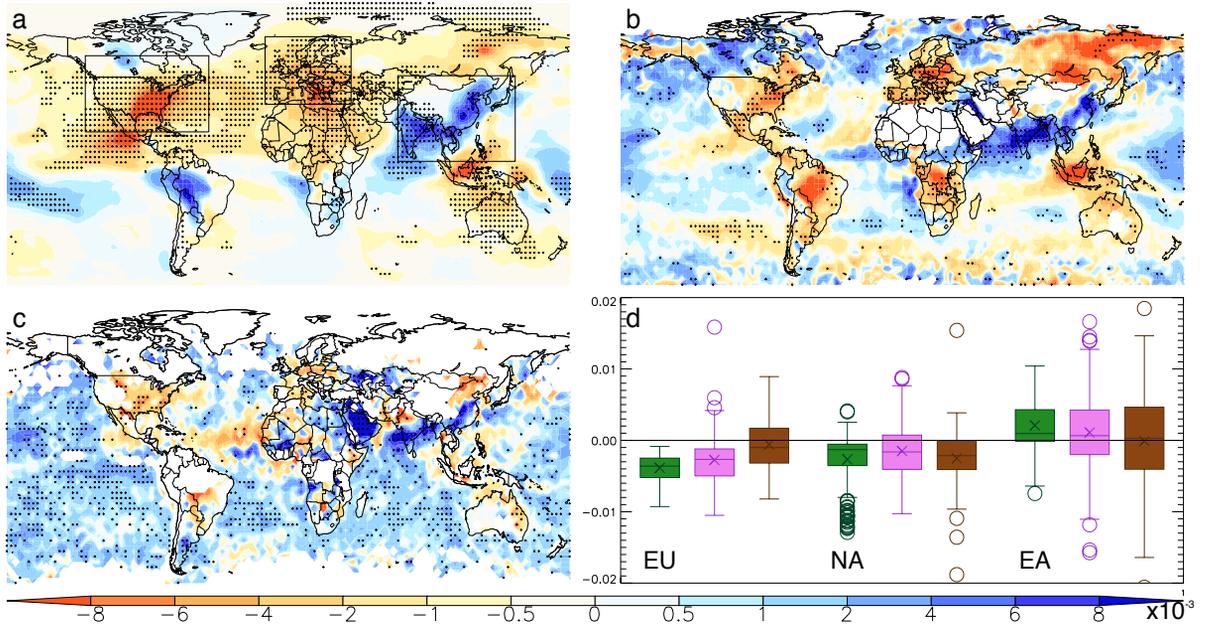


Figure 1

Figure 2. Figure

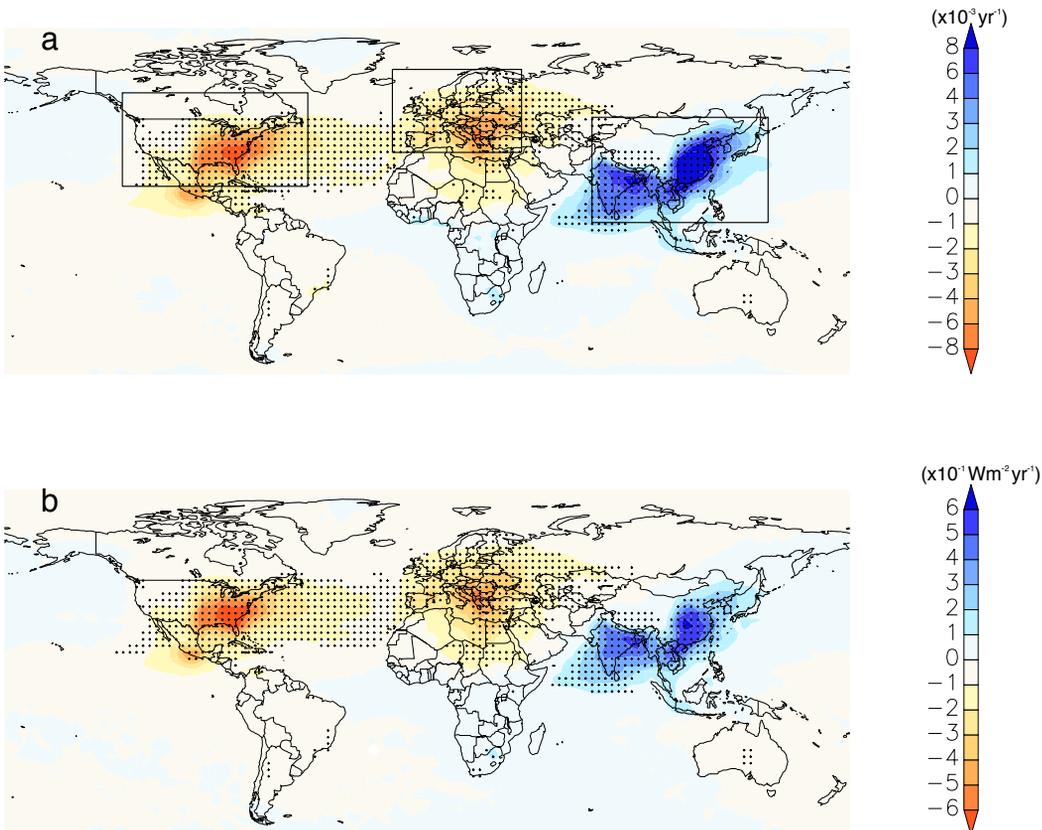


Figure 2

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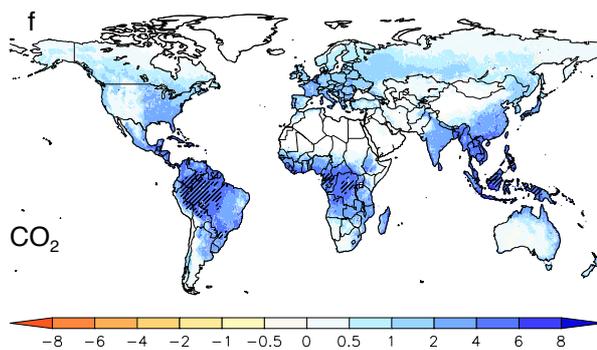
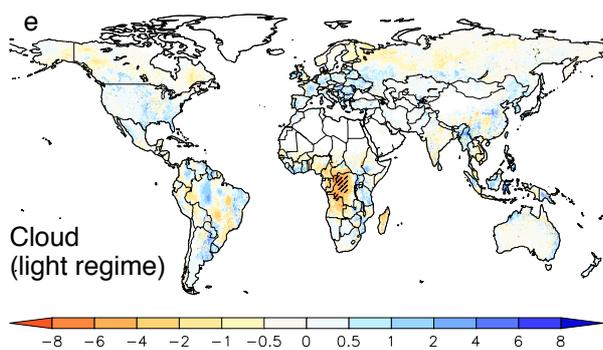
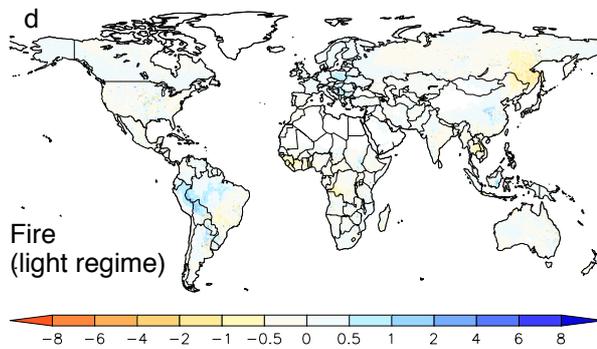
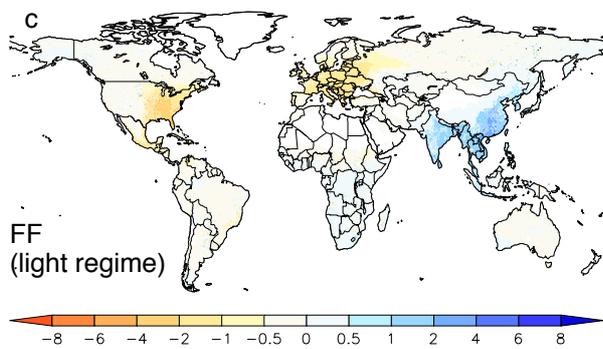
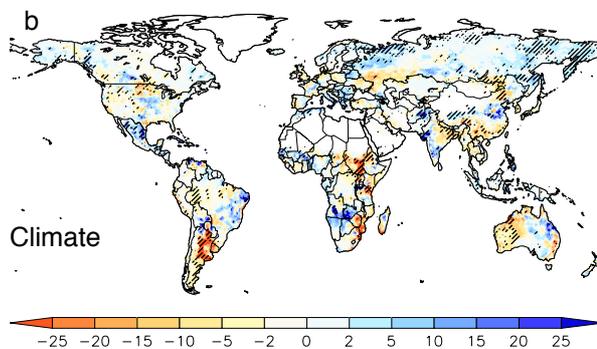
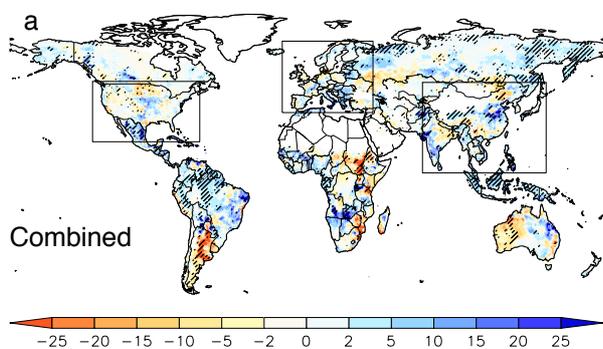


Figure 3

Figure 4. Figure

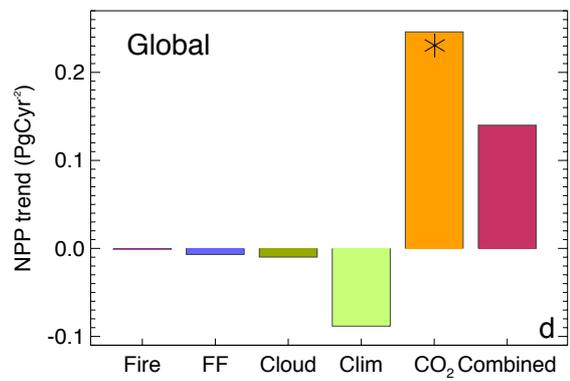
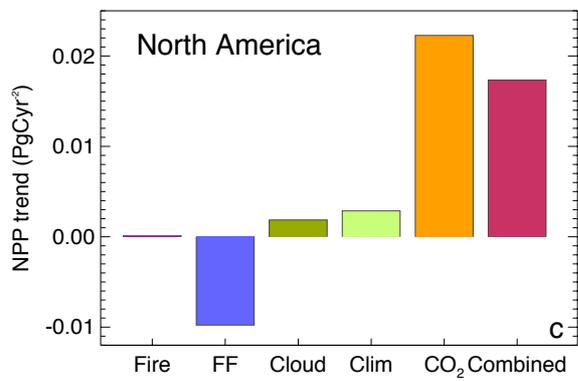
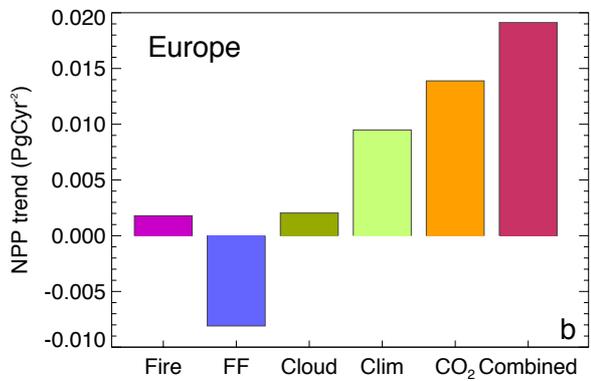
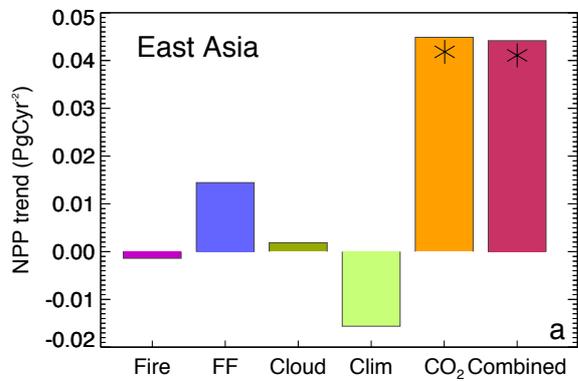


Figure 4