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Anthropogenic perturbation of the carbon fluxes from land to ocean

Supplementary Note: The terrestrial ecosystem C cycle

Contemporary global terrestrial NPP (FT_1) has been estimated to amount about 59 PgC y^{-1}, although satellite derived estimates are slightly lower[1,2]. Prior to significant human intervention (defined here as the 'natural' carbon cycle), the terrestrial net primary production was significantly lower. With an increasing human population, the demand for food, fiber and shelter was met through deforestation in favour of agricultural land-use and agricultural intensification through fertilisation, irrigation and species selection. Where deforestation is expected to have decreased the global NPP, intensified land-use in combination with increasing atmospheric CO_2 concentrations is expected to have increased the global NPP. Based on an estimate of potential NPP[1] (before human appropriation) and on DGVMs response to the historical atmospheric CO_2 increase, we estimate the net-effect of both processes to represent an increase in NPP of 4 PgC y^{-1} (FT_1). Hence, natural global terrestrial NPP is assessed to be around 55 PgC y^{-1}, but both this value and the contemporary one remain poorly constrained.

In addition, the increasing human population and affluence have increased the human appropriation of NPP (HANPP) to reach the current value of 4.4 PgC yr^{-1} and is due to crop harvest[3] (1.3 PgC yr^{-1}), wood harvest[4] (0.9 PgC yr^{-1}), biofuels production[5] (0.9 PgC yr^{-1};) and cattle grazing[1] (1.3 PgC yr^{-1}). Most of this HANPP, 4.1 PgC yr^{-1}, is emitted to the atmosphere, a small fraction (0.1 PgC yr^{-1}) is released to inland waters as sewage (F_4), and 0.2 PgC yr^{-1} are estimated to accumulate as harvest products[6]. We also accounted that a large fraction of these carbon fluxes is released to the atmosphere as CO_2 (3.85 PgC yr^{-1}; FT_2) and a small fraction (0.25 PgC yr^{-1}) as methane from cattle, rice paddies and landfills[7].

The effect of an increasing human population on fire intensity remains also difficult to assess. Average global fire carbon emissions have been estimated at 2.0 PgC yr^{-1} for present-day conditions[8], of which carbon emissions from anthropogenic disturbances such as tropical deforestation, degradation, and peatland fires contribute on average 0.5 PgC yr^{-1}. We also assume that 0.05 PgC yr^{-1} are emitted as

CH_4 from fires, thus leaving a CO_2 flux of 1.95 PgC yr^{-1} (FT_3). CH_4 emissions from fire, cattle and landfills are attributed to the terrestrial biomass reservoir (0.15 PgC yr^{-1}, FT_4), while emissions from rice paddies (0.15 PgC yr^{-1}) are attributed to soils, together with another 0.15 PgC yr^{-1} of CH_4 released by wetlands[7] (FT_6).

Typically croplands and grasslands consist of annual plants and as such the biomass accumulation is essentially restricted to forests. Consecutive forest inventories indicate a substantial 4.0 PgC yr^{-1} sink in forest with 2.9 PgC yr^{-1} in biomass, 0.9 PgC yr^{-1} in litter and soil, and 0.2 PgC yr^{-1} in harvest products[6]. However, this sink is partially offset by emissions from gross deforestation of tropical forests[6] (2.8 PgC yr^{-1}). As a result, the net C increase in forest is 1.2 PgC yr^{-1}, of which 0.2 PgC yr^{-1} is stored in harvest products. The partition of this net C increase (1.0 PgC yr^{-1} without the harvest products) between biomass, litter and soil is not reported in ref. [6]. Previous studies suggested that soil carbon loss accounts for 13 to 37% of total gross deforestation emissions[9]. Here we adopt an average value of 25%, leading to about 0.8 PgC yr^{-1} accumulating in biomass and 0.2 PgC yr^{-1} accumulating in litter and soil.

The annual carbon flux from the vegetation to the soil decreased from 53.5 PgC yr^{-1} prior to human disturbances to 51.95 PgC yr^{-1} at present (FT_5). Despite the decreasing C-inputs, soil C-sequestration is thought to have increased by 0.2 PgC yr^{-1} over forested areas[6]. However, this increase is offset by drainage of peatlands[10,11], which leads to an estimated carbon loss of 0.35 PgC yr^{-1}. The total present-day soil C reservoir is thus losing about 0.15 PgC yr^{-1}, compared to a natural sink of 0.05 PgC yr^{-1} due to peat accumulation[12] (ΔC = - 0.2 PgC yr^{-1}). Lateral C export to inland waters is thought to have increased by 0.8 PgC yr^{-1} up to its current level of 1.9 PgC yr^{-1} (F_1).

Simultaneously lower input fluxes into the litter and soil pool and higher output fluxes into inland waters are not fully compensated by the recorded decrease in soil carbon, hence the decomposition of litter and soil organic matter has also significantly decreased during the Anthropocene. Regionally such a decrease may be caused by increased harvest levels and increased N-deposition which may inhibit decomposition[13]. In the absence of data-driven global estimates of heterotrophic respiration, this flux was used to close the terrestrial budget.

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Anthropogenic perturbation of the land to ocean carbon fluxes

Supplementary Table 1: Synthesis of C fluxes for the Land-Ocean Aquatic Continuum (LOAC). Values are given for present-day conditions, as reported in the literature (black) and in our own budget analysis (red). The magnitude of the anthropogenic perturbation is also given. The anthropogenic transient is mainly attributed to land-use changes and soil erosion, liming, fertilizer and pesticide application, sewage water production, damming of water courses, water withdrawal and human-induced climatic change. The confidence in the selected values is specified for the present-day fluxes, using the SOCCR nomenclature[1]. ***: 95% certainty that the actual estimate is within 50% of the estimate reported; **: 95% certainty that the actual value is within 100% of the estimate reported; *: uncertainty greater than 100%. The method used in the published estimates is: r = review of existing numbers; d = data; m = model; b = budget. A brief justification of our proposed estimate for each of these present-day fluxes and anthropogenic perturbation is also provided.

Domain	Present day ($PgC\ yr^{-1}$)	Confi-dence	Perturbation ($PgC\ yr^{-1}$)	Method	Reference
F1: Total C input from soils to inland waters	1.9	**	0.8		This study
	1.9[a]			b	Battin et al., 2009[2]
	2.1[a]			b	Tranvik et al., 2009[3]
	0.9[a]			b	Cole et al., 2007[4]
	[0.4-1.4][b]			b	Richey, 2004[5]
			1	r	Richey, 2004[5]
			[0.3,0.55][b]	r(d)	Quinton et al., 2010[6]
			[0.47,0.61][b]	m	Van Oost et al., 2007[7]
			1.4[b]	d	Smith et al., 2001[8]
			[0.4-1.2][b]	d	Stallard, 1998[9]
F2: Inorganic C input from bedrock to inland waters	0.5	***	0.1		This study
	0.33			m	Hartmann et al., 2009[10]
	0.36			m	Amiotte Suchet et al., 2003[11]
	0.31			m	Munhoven et al., 2002[12]
	0.44			d	Gaillardet et al., 1999[13]
	0.37			d	Garrels & Mackenzie, 1971[14]
			0.1	m	Beaulieu et al., 2012[15]
F3: Atmospheric CO_2 uptake by bedrock	0.35	***	0.05		This study
	0.3		0.1	m	Beaulieu et al., 2012[15]
	[0.3,0.44]			m	Hartmann et al., 2009[10]
	0.26			m	Amiotte Suchet et al., 2003[11]
	0.22			m	Munhoven, 2002[12]
	0.29			d	Gaillardet et al., 1999[13]
			increase	d	Gislason et al., 2009[16]
			0.05	(d)	Oh and Raymond, 2006[17]
F4: Organic C input from sewage to inland waters	0.1	***	0.1		This study
	0.1		0.1	m	Mackenzie et al., 2001[18]
	0.1		0.1	m	Ver et al., 1999[19]
FR: Physical erosion of total recalcitrant C	0.3	*	0		This study
	0.1[c]			m	Copard et al., 2007[20]
	0.1[c]			m	Mackenzie & Lerman, 2006[21]
	0.17[d]			d	Meybeck, 1982 [22]
F5: Photosynthetically fixed C not respired in inland waters	0.3	*	0.1		This study
Emissions from inland waters to the atmosphere					
F6: CH_4	0.1	**	0		This study
	0.1			d	Bastviken et al., 2011[23]
F7: CO_2	1.1	**	0.5		This study
	0.63[e]		0.28[f]	d	Cole et al., 2007[4]
	1.2[g]			d	Aufdenkampe et al., 2011[24]
	1.25[e]			d	Tranvik et al., 2009[3]
	1.05[e]			d	Battin et al., 2009[2]
	1.0			d	Richey, 2004[5]
F8: Total C burial in inland waters	0.6	*	0.4		This study
	0.6		0.35[h]	d	Tranvik et al., 2009[3]
	[0.19-0.27][i]		[0.16-0.2][j]	d	Cole et al., 2007[4]
	[0.5,1.5]			r	Aufdenkampe et al., 2011[24]
	0.6			r	Battin et al., 2009[2]
			1	d	Smith et al., 2001[8]
			[0.6, 1.5]	m	Stallard, 1998[9]
			0.2[j]	r(m)	Richey, 2004[5]
			0.2[j]	d	Mulholland & Elwood, 1982[25]
F9: Total C inputs from rivers to estuaries	1.0	***	0.2		This study
	0.95		0.2	m	Andersson et al., 2005[26]
				m	Mackenzie et al., 2012[27]
	0.87			r(dm)	Cai, 2011[28]
	0.9			r	Battin et al., 2009[2]
	0.9			r	Tranvik et al., 2009[3]
	0.9			r(d)	Cole et al., 2007[4]
	[0.8 1.2]			r	Richey, 2004[5]
	0.7			m	Ludwig et al., 1998[29]; 1998[30]
	0.8			m	Sarmiento & Sundquist, 1992[31]
	0.85			d	Meybeck, 1982[22], 1991[32]
	0.2[k]			m	Beusen et al., 2005[33]
	0.53[l]			d	Stallard, 1998[9]
	0.4[l]			d	Schlesinger & Melack, 1981[34]
	0.45[m]			d	Kempe, 1979[35]
F10: Emissions from estuaries to the atmosphere					
CH_4	0		0		This study
	0.006			d	Borges & Abril, 2012[36]
CO_2	0.25	**	0		This study
	0.27			d	Borges & Abril, 2012[36]
	0.25			d	Cai, 2011[28]
	0.27			d	Laruelle et al., 2010[37]
	0.12			d	Cole et al., 2007[4]
	0.36			d	Chen & Borges, 2009[38]
	0.32			d	Borges et al., 2005[39]
	0.43			d	Borges, 2005[40]
	0.60			d	Abril & Borges, 2004[41]
F11: CO_2 uptake by coastal vegetation & organic C export to estuaries	0.3	**	-0.15		This study
	[0.17,0.4]			d	Cai, 2011[28]
	0.77 - 3.18			d	Duarte et al., 2005[42]

F12: Total C burial in estuarine sediments & coastal vegetation	**0.1**	**	**-0.05**		**This study**
	0.03[n]			d	Breithaupt et al., 2012[43]
	0.03-0.12[o]			r(d)	Mcleod et al., 2011[44]
F13: Total C inputs from estuaries to coastal ocean	**0.95**	**	**0.1**		**This study**
F14: Atmospheric CO_2 uptake by coastal ocean	**0.2**	**	**0.2**		**This study**
	0.2		0.2	m	Mackenzie et al., 2012[27]
	0.3		0.5	b	Liu et al., 2010[45]
	0.18			d	Wanninkhof et al., 2012[46]
	0.22			d	Cai, 2011[28]
	0.21			d	Laruelle et al., 2010[37]
	0.37			d	Borges, 2005[40]
	0.45			d	Borges et al., 2005[39]
	0.4			d	Thomas et al., 2004[47]
			[0.2,0.4]	m	Mackenzie et al., 2005[48]
				m	Andersson et al., 2005[26]
F15: Total carbon burial in coastal sediments	**0.35**	**	**0.15**		**This study**
	0.35		0.15	m	Lerman et al., 2004 [49]
	0.41[p]			r	Krumins et al., 2013[50]
	0.05-0.1[s]			r(d)	Mcleod et al., 2011[44]
	0.67[q]			m	Dunne et al., 2007[51]
	0.17[q]			b	Sarmiento & Gruber, 2006[52]
	0.28			m	Andersson et al., 2005[26]
	0.06[q]			m	Muller-Karger et al., 2005[53]
	0.36			b	Chen et al., 2003[54]
	0.49			m	Mackenzie et al., 1998[55]
	0.16[r]			d	Milliman & Droxler, 1996[56]
	0.2[q]			b	Wollast, 1991[57]
	0.23[q]			b	Wollast & Mackenzie, 1989[58]
F16: Total C inputs from coastal to the open ocean	**0.75**	*	**0.1**		**This study**
	0.9		0.45	b	Liu et al., 2010[45]
Inorganic C accumulation in coastal waters		*	**0.05**		**This study**
			0.05	m	Mackenzie et al., 2012[27]

[a] reported value minus 0.8 PgC y^{-1} attributed in this study to weathering and photosynthetic C fixation; [b] POC flux only. For the anthropogenic perturbation, values refer to agricultural soils and assume that erosion by water reach the aquatic continuum. It is also assumed that the C eroded is recently fixed atmospheric CO_2; [c] fossil organic carbon only; [d] particulate inorganic carbon only; [e] reported value minus 0.15 PgC y^{-1} reported by Cole et al. (2007)[4] for estuaries; [f] outgassing from reservoirs only; [g] reported value excluding wetlands; [h] storage in reservoirs and small agricultural ponds; [i] storage in lakes and reservoirs; [j] storage in reservoirs only; [k] particulate organic carbon only; [l] total organic carbon only; [m] dissolved inorganic carbon only; [n] mangroves only; [o] saltmarshes & mangroves only; [p] average of literature values; [q] POC only; [r] PIC only; [s] sea grass meadows only.

Present-day fluxes

F1: Estimated as the sum of literature values reported for aquatic-estuarine fluxes (F9), outgassing (F7) and burial (F8) minus the contribution from bedrock weathering (F2) and photosynthetic C fixation (F5). A higher weight was given to the more recent estimates by Battin et al. (2009)[2] and Tranvik et al. (2009)[3] which take into account a more complete estimate of outgassing and burial including the contributions from streams, smaller lakes and agricultural ponds

F2: Average of literature values from studies referring mainly to fairly pristine watersheds plus first order estimate of anthropogenic perturbations. Also consistent with values used in Earth System models for long-term climate studies (e.g. Climber 2)

F3: Average of literature values from studies referring mainly to fairly pristine watersheds plus first order estimate of anthropogenic perturbation

F4: Taken from Ver et al. (1999)[19] and Mackenzie et al. (2001)[18]

F5: Assuming that 20% of the C burial and export in inland waters is autochthonous

FR: Sum of literature estimates on particulate inorganic carbon and fossil particulate organic carbon stemming from rocks

F6: CH_4 flux taken from Bastviken et al. (2011)[23]

F7: Average of literature values excluding the study by Cole et al. (2007)[4], which did not include streams and smaller lakes and reservoirs, and excluding emissions from wetlands reported in Aufdenkampe et al. (2011)[24]

F8: The recent value by Tranvik et al. (2009)[3] was selected, which was updated from Cole et al. (2007)[4] based on suggestion by Downing et al. (2008)[59]. The very high values by Stallard (1998)[9] and Smith et al. (2001)[8] where discarded. This value could still be an upper-bound value considering the extreme environments investigated by Downing et al. (2008)[59] and used for upscaling. The fact that the natural C burial in all ecosystems combined (F8, F12 and F15) is somewhat larger than the geological fluxes could also corroborate this view.

F9: Calculated as mass balance on all preceding flux estimates. Our value is consistent with those reported in the literature

F10: Estimated from the three most recent publications

F11: Taken from the average reported in Cai et al. (2011)[28], discarding the high value from Duarte et al. (2005)[42] which also includes the contribution of seagrasses, macroalgae and coral reefs. Note that the burial in coastal vegetation and estuary have been lumped together

F12: Taken from McLeod et al. (2011)[44]

F13: Calculated from mass-balance on all preceding fluxes

F14: A lower estimate was chosen. Also consistent with the recent analysis by Wanninkhof et al. (2012)[46] and Mackenzie et al. (2012)[27]

F15: Average of literature values, excluding the high estimate from Dunne et al. (2007)[51]

F16: Calculated from mass-balance on all preceding fluxes, plus the change in inorganic C inventory in the global coastal ocean

Perturbation

F1: Estimated from a mass-balance on all other perturbed C fluxes through inland water systems. Broadly consistent with the anthropogenic POC erosion fluxes reported in the literature. Up to now, no estimates on perturbations of DOC and CO_2 fluxes to freshwater systems are available

F2: Taken from the literature. This estimate combines possible effects of increased CO_2 and land use

F3: Taken from the literature. This estimate combines possible effects of increased CO_2 and land use. Considering the high uncertainty of the effect of CO_2 increase on weathering rate, a lower-bound value was selected

F4: Taken from Ver et al. (1999)[19] and Mackenzie et al. (2001)[18]

F5: Assuming that 20% of the perturbation on C burial and export in inland waters is attributed to autochthonous C fixation

FR: Assumed 0. This flux has thus no effect on the anthropogenic CO_2 budget for the atmosphere

F7: Accounts for the outgassing of CO_2 from reservoirs (from Cole et al. 2007)[4] and increased CO_2 flux from other freshwater systems (due to e.g. sewage inputs and increased soil inputs), based on a linear scaling hypothesis on the increased riverine export flux (F9)

F8: Accounts for burial in reservoirs and agricultural ponds (Tranvik et al. 2009)[3] plus a highly uncertain enhanced burial (0.05 Pg C yr^{-1}) attributed to other freshwater bodies (linear scaling hypothesis)

F9: Taken from Andersson et al. (2005)[26] and Mackenzie et al. (2012)[27]

F10: Conservative estimate (no values reported in the literature). Changes in outgassing due to enhanced carbon inputs from terrestrial ecosystem could be partially offset by decreased carbon inputs from coastal vegetation

F11: Based on the surface area reduction of salt marshes and mangroves (Mcleod et al. 2011)[44]

F12: Based on the surface area reduction of salt marshes and mangroves (Mcleod et al. 2011)[44]

F13: Calculated as a mass-balance on all other estuarine carbon fluxes

F14: Based on the model-derived value by Mackenzie et al. (2012)[27] and arguments in Wanninkhof et al. (2012)[46] (conservative estimate). See also main text for further discussion

F15: Based on the model-derived value by Lerman et al. (2004)[49]

F16: Calculated as mass-balance on all other coastal ocean carbon fluxes, including increased inorganic C inventory in coastal waters (Mackenzie et al. 2012)[27]

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