



Supplement of

ORCHIDEE MICT-LEAK (r5459), a global model for the production, transport, and transformation of dissolved organic carbon from Arctic permafrost regions – Part 2: Model evaluation over the Lena River basin

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Text S1: Evaluation of Simulated NPP and Soil Respiration

5 Rates of yearly net primary production (NPP) for Russian and Siberian forests have been 6 inferred in situ from eddy flux and inventory techniques to range from 123-250 gC m⁻² 7 yr⁻¹ (Beer et al., 2006; Lloyd et al., 2002; Roser et al., 2002; Schulze et al., 1999; 8 Shvidenko and Nilsson, 2003). We likewise simulate a broad range of NPP carbon 9 uptake rates, of 61-469 gC m⁻² yr⁻¹ averaged per grid cell over the Lena basin, with a mean value of 210 gC m⁻² yr⁻¹. NPP is heterogeneously distributed over space and 10 between PFTs (SI, Fig. S4c), with forests averaging 90 gC m⁻² yr⁻¹ and grasslands 11 averaging 104 gC m⁻² yr⁻¹ over the basin as a whole. Low values tended to originate in 12 13 basin grid cells with elevated topography or high mean slope, while the maximum value was standalone, exceeding the next greatest by ~ 100 gC m⁻² yr⁻¹, and is most likely 14 caused by the edge effects of upscaling a coastal gridcell's small fraction of terrestrial 15 16 area where high productivity occurs in a small plot, to the grid cell as a whole. By 17 evaluating NPP we are also evaluating at a secondary level litter production, which is at a third level a major component of DOC production. 18

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Taken as a whole, gross primary production (GPP) was performed under simulations by four PFT groups, with the largest basin-wide bulk contributions coming from boreal needleleaf summer-green trees and C3 grasses (SI, Fig. S4a), the highest GPP uptake rates (3 TgC pixel⁻¹ yr⁻¹) generated by boreal needleleaf evergreen trees, and the remainder of GPP contributed by Boreal broad-leaved summer-green trees (SI, Fig. S4a).

- 26 Soil respiration rates, of combined soil heterotroph and plant root respiration in our Control simulation, averaged 208 gC m⁻² yr⁻¹ (0.57 gC m⁻² d⁻¹) over the Lena basin over 27 28 the period 1990-2000, which is somewhat higher than those found by Elberling (2007) 29 in tundra soils over Svalbard, of 103-176 gC m⁻² yr⁻¹ (0.28-0.48 gC m⁻² d⁻¹). Sawamoto, et 30 al. (2000) measured in situ summertime soil respiration over the central Lena basin and 31 found rates of 1.6-34 gC m⁻² d⁻¹, while Sommerkorn (2008) observed rates of 0.1-3.9 gC 32 m⁻² d⁻¹ at higher latitudes, these appearing to vary with vegetation and fire history, 33 water table depth and temperature. Mean heterotrophic respiration rates of 1.6 gC m⁻² 34 d⁻¹ are simulated here during July and August, in the range 0.0.5-2.2 gC m⁻² d⁻¹ for each of 35 the above PFT groups. The spatial distribution of, and difference in respiration rates 36 between PFT groups largely mirrors those for NPP (SI Fig. S4c), with maximum rates of 37 1.4 gC m² d⁻¹ over forested sites, versus a maximum of 2.2 gC m² d⁻¹ over 38 grassland/tundra sites (SI, Fig. S4b).
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40 Aggregated over the basin, results show that increases over the course of the 20th Century were simulated for NPP, GPP, River Discharge, DOC, CO_{2(aq.)}, autotrophic and 41 heterotrophic respiration and CO₂ evasion, with percentage changes in the last versus 42 first decade of +25%, +27%, 38%, +73%, +60%, +30%, +33% and +63%, respectively. 43 It thus appears that rising temperatures and CO₂ concentrations 44 (Fig. S12). disproportionately favoured the metabolisation of carbon within the soil and its 45 transport and mineralisation within the water column, fed by higher rates of primary 46 47 production and litter formation as well as an accelerated hydrological cycle.

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- 50 Text S2: Deficiencies in Modelled Hydrology
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52 Deficiencies in modelled hydrology correspond to those found in Fig. 12 of Guimberteau 53 et al. (2018), indicating that the modifications made in this model version, which focus on the DOC cycle, have not further degraded the hydrological performance of the model, 54 55 the causes of which are described below. Low simulated discharge for the Lena basin, 56 particularly during the late summer and autumn, is consistent with prior, Pan-Arctic 57 simulations conducted by Guimberteau et al. (2018), who ran ORCHIDEE-MICT using 58 both the GSWP3 and CRU-NCEP v7 datasets and evaluated them over the period 1981-59 2007. Despite the substantially better hydrological performance of ORCHIDEE under 60 GSWP3 climate, they described a near-systematic underestimation of summer/autumn discharge rates for both datasets over the Yukon, Mackenzie, Lena and Kolyma basins. 61 Furthermore, the discrepancy of model output between climatological datasets was 62 almost as large as the discrepancy between model output and observational data in that 63 study, which analysed this in great depth, suggesting that the source of error is both a 64 65 covariate of model process representation and parameterisation, as well as the climatological datasets themselves. Model hydrological representation and empirically 66 derived climate input data are then subject to interaction with modelled soil (e.g. 67 infiltration), vegetation (e.g. canopy interception) and thermodynamics (e.g. freezing 68 69 and consequent partitioning of water transport) from which river discharge is 70 computed, confounding full interpretation of sources of bias, briefly described below.

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72 Model process deficiency in this regard was identified by Guimberteau et al. (2018) as 73 residing in an overly restrictive representation of water impermeability through frozen topsoil, which decreases the residence time of running water by directing it to surface 74 75 runoff rather than subsurface flow, and in the process increases the susceptibility of the 76 total water volume to evapotranspiration from incoming shortwave radiation. This 77 would bias both the timing (over-partitioning of water to high runoff periods) and 78 volume of water (low bias) reaching the river stem and its eventual discharge into the 79 ocean, respectively, as demonstrated by model output. Guimberteau et al. (2018) 80 suggest that representation of sub-grid-scale infiltration mechanisms under frozen 81 conditions, such as soil freezing-drying that would enhance infiltration, be included in 82 future, yet-to-be implemented iterations of ORCHIDEE. Furthermore, we suggest that the lack of representation of lakes in ORCHIDEE, which serve to increase the time lag 83 84 between precipitation/melt and oceanic discharge, may likewise be a powerful source of 85 bias in the timing of discharge fluxes represented by the model.

86

87 Unsurprisingly, simulated surface runoff has been shown to be strongly affected by differences in precipitation between datasets (Biancamaria et al., 2009; Fekete et al., 88 89 2004), while biases in these and evapotranspiration datasets that are used to both drive 90 and evaluate the hydrological models, are a powerful source of water balance biases in high-latitude basins (Wang et al., 2015). Indeed, climatological dataset estimates for the 91 spatial distribution of high latitude winter snowfall are generally problematic, owing to 92 93 the low density of meteorological stations (Burke et al., 2013), wind-related issues with in-field collection and measurement that lead to systematic underestimates of snowfall 94 95 rates (Yang et al., 2005), creating biases in the climatological datasets that only show up 96 when the integrator of their model input -in this case river discharge -is modelled. In 97 addition, the wintertime partitioning of precipitation between rain and snow, a function 98 of 2m air temperatures in the forcing datasets, strongly affects the volume and timing of 99 runoff (Guimberteau et al., 2018; Haddeland et al., 2011). Indeed, 69% of the spatial 100 variance of the spring freshet has been attributed to snow water-equivalent bias during 101 the pre-melt season (Rawlins et al., 2007). In addition, errors in forcing of soil 102 evaporation due to inaccuracies in incoming shortwave radiation, as well as biases in the 103 parameterisation of canopy interception -a function of simulated LAI -can lead to 104 upward biases in evapotranspiration rates (Guimberteau et al., 2018).

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106 **Text S3: Deficiencies in Modelled DOC Discharge**

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108 Firstly, there is a quasi-linear positive relationship between DOC discharge and river 109 discharge (Fig. 3d). This relation is common to Arctic rivers, as DOC loading experiences 110 disproportionately large increases with increases in discharge (Fig. 4, Raymond et al., 111 2007), owing largely to the 'flushing' out of terrestrially fixed carbon from the previous 112 year's production by the massive runoff generated by ice and snow melt during the 113 spring thaw. Comparing simulated annual mean discharge rate (m³ s⁻¹) with long-term 114 observations (Ye et al. 2003) over years 1940-2000 (Fig. S3) shows that though absolute 115 discharge rates are underestimated by simulations, their interannual variation reasonably tracks the direction and magnitude of observations. Linear regressions 116 117 through each trend yield very similar yearly increases of 29 vs 38 m³ s⁻¹ yr⁻¹ for 118 simulations and observations, respectively. The observed vs. simulated mean annual 119 water discharge differential hovers at 36% (Figs. 3d, 4c), close to the 43% differential 120 between observed and simulated DOC discharge, giving some indication that, given the 121 linear relationship between water and DOC discharge, most of the DOC discrepancy can 122 be explained by the performance of the hydrology and not the DOC module, the latter of 123 which was the subject of developments added in ORCHIDEE M-L. Applying the 124 regression slope of the relationship in Fig. 3d (9E-06 mgC per m³s⁻¹) to the mean river 125 discharge discrepancy of 36%, we find that 84% of the differential between observed 126 and simulated discharge can be explained by the underperformance of the hydrology 127 module.

128

129 Further sources of error are process exclusion and representation/forcing limitations. 130 Indeed, separate test runs carried out using a different set of climatological input forcing 131 show that changing from the GSWP3 input dataset to input from bias-corrected 132 projections from the IPSL Earth System Model under the second Inter-Sectoral Impact 133 Model Intercomparison Project (ISIMIP2b (Frieler et al., 2017; Lange, 2016, 2018)) 134 protocol increases DOC discharge to the ocean to 4.14 TgC yr⁻¹ (+37%), largely due to 135 somewhat higher precipitation rates in that forcing dataset (see Table S3). Thus, the 136 choice of input dataset itself introduces a significant degree of uncertainty to model 137 output.

138

139 In addition, this model does not include explicit peatland formation and related dynamics, which is the subject of further model developments (Qiu et al., 2018) yet to 140 be included in this iteration. With peatlands thought to cover $\sim 17\%$ of the Arctic land 141 142 surface (Tarnocai et al., 2009), and with substantially higher leaching concentrations, this may be a significant omission from our model. The remaining biases likely arise 143 from errors in the interaction of simulated NPP, respiration and DOC production and 144 145 decomposition, which will impact on the net in and out -flow of dissolved carbon to the 146 fluvial system. However, the DOC relationship with these variables is less clear-cut than with river discharge. Indeed, regressions (Fig. S2) of annual DOC versus NPP (TgC yr⁻¹) 147

148 show that DOC is highly sensitive to increases in NPP, but is less coupled to it (more 149 scattered, R^2 =0.42) than other simulated fluvial carbon variables shown, i.e. aquatic CO₂ 150 evasion and soil CO_2 export to the river network. The differences in correlation and 151 slope of the variables in Fig. S2 are expected: aquatic CO_2 evasion is least sensitive yet 152 most tightly coupled to NPP ($R^2=0.52$), while CO_2 export to rivers is intermediate between the two ($R^2=0.43$). The greater correlation with NPP of DOC compared to 153 154 evasion is understandable, given that DOC leaching is a covariate of both NPP and runoff, 155 whereas evasion flux is largely dependent on organic inputs (production) and 156 temperature (see Part 1).

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158 **Text S4: Model Evaluation: DOC Discharge Seasonality**

159 160 Figure 4b shows that the bulk of the DOC outflow occurs during the spring freshet, accounting for \sim 50-70% of the total Arctic outflow (Lammers et al., 2001; Ye et al., 161 2009), in which DOC concentrations increase, as meltwater flushes out DOC 162 163 accumulated from the previous year's litter and SOC generation (Raymond et al., 2007; 164 Kutscher et al., 2017), reproduced in Fig. 4b. Simulation of the hydrological dynamic is 165 presented in maps of river discharge through the basin in Fig. 3b, which show low-flows in April with substantial hydrographic flow from upstream mountainous headwaters 166 167 and Lake Baikal inflow in the south, peak flow in June dominated by headwaters, and 168 little headwater input in September.

169

170 In Fig. 4b we observe the following: (i) DOC discharge fluxes closely track hydrological 171 fluxes. (ii) The simulated modern river discharge peak approximates the historical 172 observed discharge peak, but slightly overestimates spring fluxes and substantially 173 underestimates fluxes in the autumn, as explained above. (iii) The difference between the first and last decades of the simulation in Fig. 4b is mostly attributable to a large 174 175 increase in the DOC flux mobilised by spring freshet waters. This suggests both greater 176 peaks in simulated DOC flux and a shift to earlier peak timing, owing to an increase in 177 river discharge, indicative of an earlier spring and a progressively warmer environment 178 over the 20th Century. (iv) The maximum modelled modern monthly DOC flux rate of 179 \sim 1.3 TgC month⁻¹ is comparable to the mean maximum DOC flux rate measured in a 180 recent study (1.75 TgC month⁻¹, Kutscher et al., 2017, Fig. 2).

181

182 We compare the Raymond et al. (2007) modern DOC outflow (Fig. 4d, solid black line) 183 against simulated DOC outflow from both Zhigansk and Kusur (Fig. 4d). Simulated DOC 184 flux is underestimated for both sites. Peakflow at Zhigansk seems to be attenuated over 185 May and June in simulations, as opposed to May peakflow in observations. Peakflow at Kusur is definitively in June. This suggests that simulated outflow timing at Zhigansk 186 187 may slightly delayed, causing a split in peak discharge when averaged in the model 188 output. Thus the aggregation of model output to monthly averages from calculated daily and 30 minute timesteps can result in the artificial imposition of a normative temporal 189 190 boundary (i.e. month) on a continuous series. This may cause the less distinctive 'sharp' 191 peak seen in Fig. 4c, which is instead simulated at the downstream Kusur site, whose 192 distance some 500km away from Zhigansk more clearly explains the delay difference in 193 We further evaluate our DOC discharge at the sub-basin scale, to test seasonality. 194 whether the fractional contribution of different DOC flows from each sub-basin 195 correspond to those in their observed correlates from Kutscher et al., (2017). This 196 comparison is depicted in Fig. S5, where the observed and simulated percentage DOC 197 contributions of the Aldan, Vilui, and Upper and Lower Lena sub-basins to total flux 198 rates are 19 (24)%, 20(10%), 33 (38%) and 30 (28)% in simulations (observations) for 199 the four sub-basins, respectively. While deviations between simulated and observed 200 DOC fluxes can be expected, the nearly twofold value mismatch of the Vilui basin is due 201 to its real-word damming, not represented here. On the other hand, we cannot explain 202 the ~5% discrepancies in other sub-basin fluxes, particularly for the Aldan.

203

Text S5: Evaluation of Modelled DOC Concentrations in Stream, River, Ground Water

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207 The spatial distribution of DOC concentrations are shown in maps of mean monthly DOC 208 concentration for stream water, river water and groundwater (Fig. S6a,b,c, respectively) 209 in April, June and September. For both the stream and river water reservoirs, DOC 210 concentrations appear to have spatio-temporal gradients correlated with the flux of water over the basin during the thaw period, with high concentrations of 10-15 mgC L⁻¹ 211 212 driven by April meltwaters upstream of the basin, these high concentrations moving 213 northward to the coldest downstream regions of the basin in June. Lower DOC concentrations of ~ 5 mgC L⁻¹ dominate the basin in September when the bulk of 214 215 simulated lateral flux of DOC has dissipated into the Laptev Sea. In contrast, 216 groundwater DOC concentrations are generally stable with time, although some pixels 217 appear to experience some 'recharge' in their concentrations during the first two of the 218 three displayed thaw months. Significantly, highest groundwater DOC concentrations of 219 up to 20 mgC L⁻¹ are focussed on the highest elevation areas of the Lena basin on its 220 Eastern boundary, which are characterized by a dominance of Podzols (SI, Fig. S9b). 221 This region, the Verkhoyansk range, is clearly visible as the high groundwater DOC 222 concentration (2-20mgC L⁻¹) arc (in red) in Fig. S6a, as well as other high elevation areas 223 in the south-western portion of the basin (Fig. 3a), while the low-lying central basin 224 shows much smaller groundwater DOC concentrations (0-2mgC L⁻¹). The range of 225 simulated groundwater DOC concentration comes close to those aggregated from the 226 empirical literature by Shvartsev (2008), which finds from >9,000 observations that 227 groundwater in permafrost regions exhibit a mean concentration of $\sim 10 \text{ mgC L}^{-1}$ after 228 peatlands and swamps (not simulated here) are removed (Table 2).

229

230 The high groundwater reservoir DOC concentrations simulated in high altitude regions 231 by ORCHIDEE MICT-L is related to the fact that, in the model, DOC is rapidly produced 232 and infiltrated deep into soil above the permafrost table, to the point that it reaches the 233 simulated groundwater pool relatively quickly, allowing it to enter this reservoir before 234 being metabolised through the soil column -hence allowing for the relatively high 235 groundwater concentrations found in mountain areas. Because of the prevailing low 236 temperatures, this DOC is not quickly decomposed by microbes and instead feed the 237 groundwater DOC pool.

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239 Text S6: Riverine CO₂ Evasion

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In our model, the fate of DOC once it enters the fluvial system is either to remain as DOC and be exported to the ocean, or to be degraded to dissolved CO_2 ($CO_{2(aq.)}$), which is itself either also transported to the marine system or outgassed from the fluvial surface to the atmosphere (see Part 1, Section 2.10). The latter two outcomes also apply to $CO_{2(aq.)}$ produced in the soil by organic matter degradation and subsequently

- transported by runoff and drainage flows to the water column. As shown in Fig. 2, a 246 247 large proportion of DOC (38%, 2.1 TgC yr⁻¹) that enters the water column is degraded to $CO_{2(aq.)}$ during transport, which adds to the 1.65 TgC yr⁻¹ of direct $CO_{2(aq.)}$ input from the 248 249 terrestrial land surface. Of this bulk CO₂ exported into and generated within the water 250 column, 3.6 TgC yr⁻¹ evades from the water surface to the atmosphere before reaching 251 the river delta. In what follows, we evaluate first inputs of $CO_{2(ac.)}$ to the water column in 252 terms of their seasonality, before evaluating CO₂ evasion rates and the relation of this to 253 smaller and larger water bodies (river versus stream).
- 254
- 255 To our knowledge, no direct measurements for CO₂ evasion from the surface of the Lena 256 river are available in the literature. We refer to Denfeld et al. (2013) for evaluating our 257 evasion flux results, since their basin of study, the Kolyma River, is the most 258 geographically proximate existing dataset to the Lena, despite biogeographical 259 differences between the two basins -namely that the Kolyma is almost entirely 260 underlain by continuous permafrost. The Kolyma River CO₂ evasion study measured 261 evasion at 29 different sites along the river basin (~158-163°E; 68-69.5°N), with these sites distinguished from one another as 'main stem', 'inflowing river' or 'stream' on the 262 263 basis of reach length. The study showed that during the summer low-flow period 264 (August), areal river mainstem CO_2 evasion fluxes were ~0.35 gC m⁻² d⁻¹, whereas for 265 streams of stream order 1-3 (widths 1-19m), evasion fluxes were up to $\sim 7 \text{ gC m}^{-2} \text{ d}^{-1}$, 266 and for non-mainstem rivers (widths 20-400m) mean net fluxes were roughly zero 267 (Table 3 of Denfeld et al., 2013). Thus, while small streams have been observed to 268 contribute to roughly 2% of the Kolyma basin surface area, their measured percentage 269 contribution to total basin-wide CO_2 evasion ~40%, whereas for the main stem the surface area and evasion fractions were $\sim 80\%$ and 60%, respectively. Likewise, mean 270 271 annual evasion rates of <0.8 up to around 7 gC m⁻² d⁻¹ have been found for the Ob and 272 Pur rivers in Western Siberia (Serikova et al., 2018).
- 273

274 Results such as these, in addition to permafrost soil incubation experiments (e.g. Drake 275 et al., 2015; Vonk et al., 2013, 2015b, 2015a) suggest that small streams, which 276 represent the initial (headwater) drainage sites of these basins, rapidly process 277 hydrologically leached carbon to the atmosphere, and that this high-reactivity carbon is 278 a mix of recently thawed ancient permafrost material, as well as decomposing matter from the previous growth year. This is given as evidence that the total carbon 279 280 processing of high-latitude rivers is significantly underestimated if only mainstem carbon concentrations are used in the accounting framework, since a large amount of 281 282 carbon is metabolised to the atmosphere before reaching the site of measurement.

283

284Text S7: Spatio-Temporal Heterogeneity in CO2 Evasion

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286 The heterogeneity of CO₂ evasion from different sources in the model is most evident in 287 terms of their geographic distribution and relative intensity, as shown in the evasion 288 flux rate maps over stream and river areas in April, June and September (Fig. S8a-b). 289 Stream evasion (Fig. S8a), tends to be broadly distributed over the whole basin, 290 representing the fact that small streams and their evasion are the main hydrologic 291 connectors outside of the main river and tributary grid cells, whereas river evasion (Fig. 292 S8b) is clearly linked to the hydrographic representation of the Lena main stem itself, 293 with higher total quantities in some individual grid cells than for the stream reservoir, 294 yet distributed amongst a substantially smaller number of grid cells. Whereas the

295 stream reservoir has greatest absolute evasion flux rates earlier in the year (April-May), 296 maximum evasion rates occur later in the year and further downstream for the river 297 reservoir, reflecting the fact that headwaters are first-order integrators of soil-water 298 carbon connectivity, whereas the river mainstem and tributaries are of a secondary 299 order.

300

301 The spatio-temporal pattern of increasing evasion over the simulation period is shown 302 in Fig. S7 as a Hovmöller difference plot, between the last and first decade, of log-scale 303 average monthly evasion rates per latitudinal band. This shows that the vast majority of 304 outgassing increase occurs between March and June, corresponding to the progressive 305 onset of the thaw period moving northwards over this timespan. Although relatively 306 small, outgassing increases are apparent for most of the year, particularly at lower 307 latitudes. This would suggest that the change is driven most acutely by relatively greater 308 temperature increases at higher latitudes ('Arctic amplification' of climate warming, e.g. 309 Bekryaev et al., 2010) while less acute but more temporally homogenous evasion is 310 driven by seasonal warming at lower latitudes.

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312 Text S8:

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314 This is because in ORCHIDEE MICT-L, the 'stream' water reservoir is water routed to the 315 river network for all hydrologic flows calculated to not cross a 0.5 degree grid cell 316 boundary (the resolution of the routing module, explained in Part 1, Section 2.6), which 317 may not be commensurate with long, <20m width streams in the real-world, that were 318 used in the Denfeld et al. (2013) study. In addition, this 'stream' water reservoir in the 319 model does not include any values for width or area in the model, so we cannot directly

- 320 compare our stream reservoir to the <20m width criterion employed by Denfeld et al. 321
- (2013) in their definition of an observed stream. Thus our 'stream' water reservoir
- 322 encompasses substantially greater surface area and hydrologic throughput than that in 323 the Denfeld et al. study.
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325 Text S9:

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327 The 'approximate' caveat refers to the fact that model output doesn't define a precise 328 surface area for the stream water reservoir, which is instead bundled into a single value 329 representing the riverine fraction of a grid cell's total surface area. To approximate the 330 areal outgassing for the stream versus river water reservoirs, we weight the total nonfloodplain inundated area of each grid cell by the relative total water mass of each of the 331 332 two hydrological pools, then divide the total daily CO₂ flux simulated by the model by this value. The per-pool areal estimate is an approximation since it assumes that rivers 333 334 and streams have the same surface area: volume relationship. This is clearly not the 335 case, since streams are generally shallow, tending to have greater surface area per increment increase in depth than rivers. Thus, our areal approximations are likely 336 337 underestimated (overestimated) for streams (rivers), respectively. Note that from \sim 700 338 non-zero simulation datapoints used to generate Fig. 6d were omitted as 'outliers' from 339 the stream reservoir efflux statistics described below, because very low stream:river 340 reservoir values skewed the estimation of total approximate stream surface area values very low, leading to extreme efflux rate values of 1-3000gC m⁻² d⁻¹ and are thus 341 342 considered numerical artefacts of the areal approximation approach used here. 343

345 **Text S10: Emergent Phenomena: DOC and Topographic Slope**

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347 This relationship was found in temperate rivers by Lauerwald et al. (2012), and in a recent Pan-Arctic synthesis paper Connolly et al. (2018). The reasoning for the negative 348 349 slope-DOC concentration relationship is that as elevation increases, temperature and 350 primary production decreases. This leads to a thinner organic soil layer, meaning that 351 mineral soil plays a stronger role in shallow hydrologic flowpaths, allowing for deeper 352 infiltration and shorter residence time in a given soil layer. Further, steeper terrain 353 leads to a lower soil water residence time and lower moisture than in flat areas. As a 354 result, a given patch of soil matter will be exposed to leaching for less (residence) time, 355 while the organic matter that is leached is thought to be adsorbed more readily to 356 mineral soil particles, leading to either their re-stabilisation in the soil column or 357 shallow retention and subsequent heterotrophic respiration in situ, cumulatively 358 resulting in lower DOC concentrations in the hydrologic export (Kaiser and Kalbitz, 359 2012; Klaminder et al., 2011). This line of reasoning was recently shown to apply also to 360 deep organic permafrost soils (Zhang et al., 2017), although the degree to which this is 361 the case in comparison to mineral soils is as yet unknown.

362

363 In addition, and as described in Part 1 (Section 2.5) of this study, MICT-L contains a 364 provision for increased soil column infiltration and lower decomposition rates in areas 365 underlain by Podzols and Arenosols. The map from the Harmonized World Soil Database 366 (Nachtergaele, Freddy, Harrij van Velthuizen, Luc Verelst, N. H. Batjes, Koos Dijkshoorn, 367 V. W. P. van Engelen, Guenther Fischer, Arwyn Jones, Luca Montanarella, Monica Petri, 368 Sylvia PrielerB, Xuezheng Shi, Edmar Teixera and David Wiberg, 2010), which is used as 369 the input to this criterion, shows areas underlain by these soils in the Lena basin to also 370 be co-incident with areas of high topographic slope (Fig. 3a, SI, Fig S9b). The 'Podzol 371 effect' is to increase the rate of decomposition and infiltration of DOC, relative to all 372 other soil types, thus also increasing the rate of DOC flux into groundwater (see Part 1 of 373 this study, Section 2.5). Thus, our modelling framework explicitly resolves the 374 processes involved in these documented dynamics -soil thermodynamics, solid vertical 375 flow (turbation), infiltration as a function of soil textures and types, adsorption as a 376 function of soil parameters (see Part 1 of this study, Section 2.11), DOC respiration as a 377 function of soil temperature and hence depth (Part 1, Section 2.12), and lagging of DOC 378 vertical flow behind hydrological drainage flow (summary Figure in Part 1, Fig. 1). We 379 thus have some confidence in reporting that the simulated negative relationship of DOC 380 concentration with topographic slope may indeed emerge from the model.

381

382 Text S11: Emergent Phenomena: DOC and Mean Annual Air Temperature

383

A key emergent property of DOC concentrations in soils and inland waters should be their positive partial determination by the temperature of the environment under which their rates of production occur, as has been shown in the literature on permafrost regions, most notably in Frey & Smith (2005) and Frey & McClelland (2009). Increasing

temperatures should lead to greater primary production, thaw, decomposition and microbial mobilisation rates, and hence DOC production rates, leading to (dilution effects notwithstanding) higher concentrations of DOC in thaw and so stream waters. Looking at this emergent property allows us to evaluate the soil-level production of both

392 DOC and thaw water at the appropriate biogeographic and temporal scale in our model.

This provides a further constraint on model effectiveness at simulating existingphenomena at greater process-resolution.

395

396 Figure 7 compares three datasets (simulated and two observational) of riverine DOC 397 concentration (in mgC L⁻¹) plotted against mean annual air temperature (MAAT). The 398 simulated grid-scale DOC versus MAAT averaged over July and August (for 399 comparability of DOC with observational sampling period) of 1998-2007 is shown in 400 red, and observed data compiled by Laudon et al. (2012) and Frey and Smith (2005) for 401 sites in temperate/cold regions globally and peatland-dominated Western Siberia, 402 respectively. The Laudon et al. (2012) data are taken from 49 observations including MAAT over the period 1997-2011 from catchments north of 43°N, and aggregated to 10 403 404 regional biogeographies, along with datapoints from their own sampling; those in the 405 Frey and Smith study are from 55-68°N and ~65-85°E (for site locations, see Laudon et 406 al. (2012), Table 1 and 2; Frey and Smith (2005), Fig. 1).

407

408 Fig. 7 can be interpreted in a number of ways. First, this MAAT continuum spans the 409 range of areas that are both highly and moderately permafrost affected and permafrost 410 free (Fig. 7, blue and green versus orange shading, respectively), potentially allowing us 411 a glimpse of the behaviour of DOC concentration as the environment transitions from 412 the former to the latter. Simulated Lena DOC concentrations, all in pixels with MAAT < -413 2°C and hence all bearing continuous or discontinuous permafrost ('permafrost-414 affected' in the figure), only exhibit a weakly positive response to MAAT on the scale used ($y=6.05e^{0.03MAAT}$), although the consistent increase in DOC minima with MAAT is 415 clearly visible. Second, the Laudon et al. (2012) data exhibit an increasing then 416 417 decreasing trend over the range of MAAT (-2°C to 10°C) in their dataset, which they propose reflects an 'optimal' MAAT range (0-3°C) for the production and transport of 418 419 DOC (Fig. 7, red shading). Below this optimum range, DOC concentrations may be 420 limited by transport due to freezing, and above this, smaller soil carbon pools and 421 temperature-driven decomposition would suppress the amount of DOC within rivers. 422 Third, the lower end of the Laudon et al. (2012) MAAT values correspond to a DOC 423 concentration in line with DOC concentrations simulated by our model. Fourth, DOC 424 concentrations in the Frey and Smith (2005) data exhibit a broad scattering in 425 permafrost-affected sites, with concentrations overlapping those of our simulations (Fig. 426 7, green shading), before rapidly increasing to very high concentrations relative to the 427 Laudon et al. (2012) data, as sites transition to permafrost-free (red shading, y=3.6_{MAAT}+29.4). 428

429

Their data highlight the difference in DOC concentration regime between areas of high 430 (Frey and Smith, 2005) and low (Laudon et al., 2012) peatland coverage and the 431 different response of these to temperature changes. Fifth, because our simulation 432 433 results largely correspond with the observed data where the MAAT ranges overlap 434 (green shading), and because our model lacks peatland processes, we should expect our 435 model to follow the polynomial regression plotted for the Laudon et al. (2012) data as 436 temperature inputs to the model increase. Figure 7 implies that this increase should be 437 on the order of a doubling of DOC concentration as a system evolves from a MAAT of -438 2°C to 2°C. With warming, we expect the response of DOC concentrations to reflect a 439 mix of both observationally-derived curves, as a function of peatland coverage.

- 440
- 441 **Text S12: LOAC drivers**

The constant climate (CLIM) and constant CO₂ (CO2) simulations described in Section 3
were undertaken to assess the extent –and the extent of the difference –to which these
two factors are drivers of model processes and fluxes. These differences are
summarised in Figs. S10 (a-b), in which we show the same 1998-2007 –averaged yearly
variable fluxes as in the CTRL simulation, expressed as percentages of the CTRL values
given in Fig. 2. A number of conclusions can be drawn from these diagrams.

449

450 First, all fluxes are lower in the factorial simulations, which can be expected due to 451 lower carbon input to vegetation from the atmosphere (constant CO_2) and colder 452 temperatures (constant climate) inhibiting more vigorous growth and carbon cycling. 453 Second, broadly speaking, both climate and CO₂ appear to have similar effects on all 454 fluxes, at least within the range of climatic and CO₂ values to which they have subjected 455 the model in these historical runs. With regard to lateral export fluxes in isolation, 456 variable climate (temperature increase) is a more powerful driver than CO₂ increase 457 (see below). Third, the greatest difference between the constant climate and CO_2 458 simulation carbon fluxes appear to be those associated with terrestrial inflow of 459 dissolved matter to the aquatic network, these being more sensitive to climatic than CO₂ variability. This is evidenced by a 49% and 32% decline in CO₂ and DOC export, 460 461 respectively, from the land to rivers in the constant climate simulation, versus a 27% 462 and 23% decline in these same variables in the constant CO₂ simulation. Given that the 463 decline in primary production and respiration in both factorial simulations was roughly 464 the same, this difference in terrestrial dissolved input is attributable to the effect of 465 climate (increased temperatures) on the hydrological cycle, driving changes in lateral 466 export fluxes.

467

468 This would imply that at these carbon dioxide and climatic ranges, the modelled DOC 469 inputs are slightly more sensitive to changes in the climate rather than to changes in 470 atmospheric carbon dioxide concentration and the first order biospheric response to 471 this. However, while the model biospheric response to carbon dioxide concentration 472 may be linear, thresholds in environmental variables such as MAAT may prove to be 473 tipping points in the system's emergent response to change, as implied by Fig. 7, 474 meaning that the Lena, as with the Arctic in general, may soon become much more temperature-dominated with regard to the drivers of its own change. 475

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478 **Supplementary Tables and Figures**

- 479
- **Table S1:** Data type, name and sources of data files used to drive the model in the studysimulations.
- 482 483

Data Type	Name	Source		
Vegetation Map	ESA CCI Land Cover Map	(Bontemps et al., 2013)		
Topographic Index	STN-30p	(Vorosmarty et al., 2000)		
Stream flow direction	STN-30p	Vörösmarty et al., 2000		
River surface area		(Lauerwald et al., 2015)		
Soil texture class		(Reynolds et al., 1999)		
Climatology GSWP3 v0, 1 degree		http://hydro.iis.u-tokyo.ac.jp/GSWP3/		

Potential floodplains	Multi-source global wetland maps	(Tootchi et al., 2019)	
		(Nachtergaele, Freddy, Harrij van Velthuizen,	
		Luc Verelst, N. H. Batjes, Koos Dijkshoorn, V.	
		W. P. van Engelen, Guenther Fischer, Arwyn	
		Jones, Luca Montanarella, Monica Petri, Sylvia	
		PrielerB, Xuezheng Shi, Edmar Teixera and	
Poor soils	Harmonized World Soil Database map	David Wiberg, 2010)	
Spinup Soil Carbon Stock	20ky ORCHIDEE-MICT soil carbon spinup	(Guimberteau et al., 2018)	

Table S2: Literature sources for empirical evaluation of model output.

	Empirical Evaluation Sources		
DOC Discharge	Cauwet and Sidorov (1996); Dolman et al. (2012); Holmes et al. (2012); Lara et al. (1998); Raymond et al. (2007); Semiletov et al. (2011); Kutscher et al. (2017).		
Water Discharge	Ye et al. (2009); Lammers et al. (2001)		
DOC concentration	Shvartsev (2008); Denfeld et al. (2013); Mann et al. (2015); Raymond et al. (2007); Semiletov et al. (2011); Arctic-GRO/PARTNERS (Holmes et al., 2012)		
NPP	Beer et al. (2006); Lloyd et al. (2002); Roser et al. (2002); Schulze et al. (1999); Shvidenko and Nilsson, (2003)		
Soil Respiration	Elberling (2007); Sawamoto et al. (2000); Sommerkorn (2008).		
CO2 Evasion	Denfeld et al. (2013); Serikova et al. (2018).		

Table S3: Observed versus simulated DOC discharge (1998-2007), where we compare

- the output of two separate climatological datasets used as input to the model (GSWP3
- and ISIMIP 2b). Also shown are the simulated versus observed DOC discharge for the six
- 491 largest Arctic rivers (the "Big Six") and for the Pan-Arctic as a whole.

	Simulated DOC to Ocean	Simulated DOC to Ocean	Observations (Holmes et al., 2012)	
	GSWP3	ISIMIP 2b	PARTNERS/Arctic-GRO	
Lena	3.16	4.14	5.68	
Big 6		19.36	18.11	
Pan-Arctic		32.06	34.04	

Table S4: Mean observed groundwater CO₂ and DOC concentrations for global
permafrost regions subdivided by biogeographic province and compiled by Shvartsev
(2008) from over 9000 observations.

	Permafrost Groundwater Provinces				
	Swamp	Tundra	Taiga	Average	Average (-Swamp)
CO_2 (mgC L ⁻¹)	12.3	14	10.8	12.4	12.4
DOC (mgC L ⁻¹)	17.6	10.1	9.3	12.3	9.7

(a)





514 in the model. Red boxes indicate meta-reservoirs of carbon, black boxes the actual pools 515 as they exist in the model. Black arrows indicate carbon fluxes between pools, dashed red arrows give carbon loss as CO₂, green arrows highlight the fractional distribution of 516 DOC to SOC (no carbon loss incurred in this transfer), a feature of this model. For a given 517 temperature (5°C) and soil clay fraction, the fractional fluxes between pools are given 518 519 for each flux, while residence times for each pool (τ) are in each box. The association of 520 carbon dynamics with the hydrological module are shown by the blue arrows. Blue 521 coloured boxes illustrate the statistical sequence which activates the boolean 522 floodplains module. Note that for readability, the generation and lateral flux of 523 dissolved CO₂ is omitted from this diagram, but is described at length in the Methods 524 section. (c) (Left) Soil carbon concentrations per depth level for each soil carbon 525 reactivity pool at the end of the spinup period. (Right) Evolution of each soil carbon pool 526 over the course of the 400-year spinup quasi-eqliuibration period. 527





Figure S2: Summed yearly lateral flux versus NPP values for DOC discharge, CO₂

discharge and CO₂ evasion (FCO₂) over the entire simulation period, with linear
 regression lines shown.





Figure S3: Observed versus simulated mean annual water discharge from the Lena river, where observations are taken from (Ye et al., 2003). 535 536



538 539







544 Figure S4: (a) Absolute yearly gross primary productivity (GPP, TgC yr⁻¹) for the four 545 relevant PFT groups over the Lena basin, averaged over 1998-2007. (b) Mean July and 546 August soil heterotrophic respiration rates (g $m^2 d^{-1}$) for the same PFT groups as in (a), 547 548 during the period 1998-2007. (c) Average yearly NPP (gC m² yr⁻¹) averaged over the period 1998-2007. All maps have the Lena basin area shaded in the background. (d) 549 550 Mean monthly carbon uptake (GPP) versus its heterotrophic respiration from the soil 551 (Het_Resp) in TgC per month, over the period 1998-2007. 552





555 Figure S5: Map adapted from Fig. 2 in Kutscher et al. (2017) showing proportional sub-556 basin contributions of TOC outflow to total TOC discharge in June and July (designated as their sampling period 'p⁻¹') of 2012-2013, as observed in Kutscher et al., 2017 (black 557 arrows), and DOC export contributions as simulated over the period 1998-2007 by 558 559 ORCHIDEE MICT-L (red boxes). Simulation pixels used in the calculation are correlates of the real-world sampling locations unless the site coordinates deviated from a 560 mainstem hydrographic flowpath pixel -in which case a nearest 'next-best' pixel was 561 used. Here the percentages are out of the summed mean bulk DOC flow of each 562 tributary, not the mean DOC discharge from the river mouth, because doing so would 563 negate the in-stream loss of DOC via degradation to CO₂ while in-stream. 564



567 0.0 5.0 10.0 15.0 20.0
Figure S6: Maps of (a) DOC concentrations (mgC L⁻¹) in groundwater ('slow' water pool), (b) stream water pool, (c) river water pool in April, June and September (first to third rows, respectively), averaged over the period 1998-2007. The coastal boundary and a water body overlay have been applied to the graphic in red and black, respectively, and the same scale applies to all diagrams. All maps have the Lena basin area shaded in the background.

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Figure S7: Log-scale Hovmöller diagram plotting the longitudinally-averaged difference
(increase) in total CO₂ evaded from the Lena River basin between the average of the

- periods 1998-2007 and 1901-1910, over each montly timestep, in (log) gC m⁻² d⁻¹. Thus
 as the river drains northward the month-on-month difference in water-body CO₂ flux,
 between the beginning and end of the 20th Century is shown.
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583 0.0 0.1 1.0 10.0 100.0584 **Figure S8:** Maps of CO₂ evasion from the surface of the two fluvial hydrological pools in 585 the model, (a) streams and (b) rivers in April, June and September. All maps use the 586 same (log) scale in units of (mgC m⁻² d⁻¹).

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- Figure S9: (a) Maximum floodable fraction of grid cells for the Lena basin per the input
 map from Tootchi et al. (2018). (b) Podzol and Arenosol map (Nachtergaele, Freddy,
 Harrij van Velthuizen, Luc Verelst, N. H. Batjes, Koos Dijkshoorn, V. W. P. van Engelen,
 Guenther Fischer, Arwyn Jones, Luca Montanarella, Monica Petri, Sylvia PrielerB,
 Xuezheng Shi, Edmar Teixera and David Wiberg, 2010) used as input to the 'poor soils'
- 600 module, basin mask in the background.
- 601
- 602



607 **Figure S10: (a-b)**: Schematic diagrams detailing the major yearly carbon flux outputs 608 from simulations averaged over the period 1998-2007 as they are transformed and 609 transported across the land-aquatic continuum. Figures **(b)** and **(c)** give the same fluxes 610 as a percentage difference from the Control (CTRL-Simulation), for the constant climate 611 and CO_2 simulations, respectively.

612



614 **Figure S11:** Simulated basin-mean annual DOC concentrations (mg L⁻¹) for the

- 615 floodplain water pool regressed against mean annual simulated discharge rates at Kusur 616 $(m^3 s^{-1})$ over 1901-2007. A linear regression with R² is plotted.
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Figure S12: Time series showing the decadal-mean fractional change in carbon fluxes

- normalised to a 1901-1910 average baseline (=1 on the y-axis) for NPP, GPP,
 autotrophic and heterotrophic respiration, DOC inputs to the water column, CO₂ inputs
- autotrophic and heterotrophic respiration, DOC inputs to the water column, CO₂ inputs
 to the water column, CO₂ evasion from the water surface (FCO2), and discharge.
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