

Fluvial carbon dioxide emission from the Lena River basin during spring flood

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Abstract

Greenhouse gas (GHG) emission from inland waters of permafrost-affected regions is one of the key factors of circumpolar aquatic ecosystem response to climate warming and permafrost thaw. Riverine systems of central and eastern Siberia contribute a significant part of the water and carbon (C) export to the Arctic Ocean, yet their C exchange with the atmosphere remain poorly known due to lack of *in-situ* GHG concentration and emission estimates. Here we present the results of continuous *in-situ* pCO₂ measurements over a 2600-km transect of the Lena River main stem and lower reaches of 20 major tributaries (together representing watershed area of 1,661,000 km², 66% of the Lena's basin), conducted at the peak of the spring flood. The pCO₂ in Lena (range 400-1400 μatm) and tributaries (range 400-1600 μatm) remained generally stable (within ca. 20 %) over the night/day period and across the river channels.

32 The pCO₂ in tributaries increased northward with mean annual temperature decrease and permafrost
33 increase; this change was positively correlated with C stock in soil, the proportion of deciduous needle-
34 leaf forest and the riparian vegetation. Based on gas transfer coefficients obtained from rivers of the
35 Siberian permafrost zone, we calculated CO₂ emission for the main stem and tributaries. Typical fluxes
36 ranged from 1 to 2 g C m⁻² d⁻¹ (>99% CO₂, < 1 % CH₄) which is comparable with CO₂ emission measured
37 in Kolyma, Yukon and Mackenzie and permafrost-affected rivers in western Siberia. The areal C
38 emissions from lotic waters of the Lena watershed were quantified via taking into account the total area
39 of permanent and seasonal water of the Lena basin (28,000 km²). Assuming 6 months of the year to be
40 open water period with no emission under ice, the annual C emission from the whole Lena basin is
41 estimated as 8.3 ± 2.5 Tg C y⁻¹ which is comparable to the DOC and DIC lateral export to the Arctic
42 Ocean.

43

44

45 **Introduction**

46 Climate warming in high latitudes is anticipated to result in mobilization, decomposition and
47 atmospheric release of significant amounts of carbon (C) stored in permafrost soils, providing a positive
48 feedback (Schuur et al. 2015). Permafrost thawing is expected to also increase the lateral C export to
49 rivers and lakes (Frey and Smith, 2005). The exported permafrost C is relatively labile and largely
50 degraded to greenhouse gases (GHG) in recipient freshwaters (e.g. Vonk et al., 2015). As a result,
51 assessment of GHG emission in rivers of permafrost affected regions is crucially important for
52 understanding the high latitude C cycle under various climate change scenarios (Chadburn et al., 2017;
53 Vonk et al., 2019). Among six great Arctic rivers, Lena is most emblematic one, situated chiefly within
54 the continuous permafrost zone and exhibiting the highest seasonal variation in discharge. Over the past
55 two decades, there has been an explosive interest to the Lena River hydrology (Yang et al., 2002;
56 Berezovskaya et al., 2005; Smith and Pavelsky, 2008; Ye et al., 2009; Gelfan et al., 2017; Suzuki et al.,
57 2018), organic C (OC) transport (Lara et al., 1998; Raymond et al., 2007; Semiletov et al., 2011;
58 Goncalves-Araujo et al., 2015; Kutscher et al., 2017; Griffin et al., 2018) and general hydrochemistry

59 (Gordeev and Sidorov, 1993; Cauwet and Sidorov, 1996; Huh et al., 1998a,b; Huh and Edmond, 1999;
60 Wu and Huh, 2007; Kuzmin et al., 2009; Pipko et al., 2010; Georgiadi et al., 2019; Juhls et al., 2020)
61 including novel isotopic approaches for nutrients (Si, Sun et al., 2018) and trace metals such as Li
62 (Murphy et al., 2019) and Fe (Hirst et al., 2020). This interest is naturally linked to the Lena River
63 location within the forested continuous permafrost/taiga zone covered by organic-rich yedoma soil.
64 Under on-going climate warming, the soils of the Lena River watershed are subjected to strong thawing
65 and active (seasonally unfrozen) layer deepening (Zhang et al., 2005) accompanied by overall increase
66 in river water discharge (McClelland et al., 2004; Ahmed et al., 2020), flood intensity and frequency
67 (Gautier et al., 2018). The Lena River exhibits the highest DOC concentration among all great Arctic
68 rivers (i.e., Holmes et al., 2013) which may reflect weak DOC degradation in the water column and
69 massive mobilization of both contemporary and ancient OC to the river from the watershed (Feng et al.,
70 2013; Wild et al., 2019). In contrast to rather limited works on CO₂ and CH₄ emissions from water
71 surfaces of Eastern Siberia (Semiletov, 1999; Denfeld et al., 2013), extensive studies were performed on
72 land, in the polygonal tundra of the Lena River Delta (Wille et al., 2008; Bussman, 2013; Sachs et al.,
73 2008; Kutzbach et al., 2007) and the Indigirka Lowland (van der Molen et al., 2007). Finally, there have
74 been several studies of sediment and particular matter transport by the Lena River to the Laptev Sea
75 (Rachold et al., 1996; Dudarev et al., 2006) together with detailed research of the Lena River Delta
76 (Zubrzycki et al., 2013; Siewert et al., 2016).

77 Surprisingly, despite such extensive research on C transport, storage, and emission in Eastern
78 Siberian landscapes, C emissions of the Lena River main stem and tributaries remain virtually unknown,
79 compared to a relatively good understanding of those in the Yukon (Striegl et al., 2012; Stackpoole et
80 al., 2017), Mackenzie (Horan et al., 2019), Ob (Karlsson et al., 2021; Pipko et al., 2019) and Kolyma
81 (Denfeld et al., 2013). The only available estimates of C emission from inland waters of the Lena basin
82 are based on few indirect (calculated gas concentration and modelled fluxes) snapshot data with very low
83 spatial and temporal resolution (Raymond et al., 2013). Similar to other regions, this introduces
84 uncertainties and cannot adequately capture total regional C emissions (Abril et al., 2015; Denfeld et al.,
85 2018; Park et al., 2018; Klaus et al., 2019; Klaus and Vachon, 2020; Karlsson et al., 2021). In particular,

86 no detailed studies at the peak of spring flood have been performed and the information on various
87 contrasting tributaries of the Lena River remains very limited. As a result, reliable estimations of
88 magnitude and controlling factors of C emission in the Lena River basin are poorly understood. The
89 present work represents a first assessment of CO₂ and CH₄ concentration and fluxes of the main stem
90 and tributaries during the peak of spring flow, via calculating C emission and relating these data to river
91 hydrochemistry and GIS-based landscape parameters. This should allow identifying environmental
92 factors controlling GHG concentration and emission in the Lena River watershed in order to use this
93 knowledge to foresee future changes in C balance of the largest permafrost-affected Arctic river.

94

95 **2. Study Site, Materials and Methods**

96 *2.1. Lena River and its tributaries*

97 The sampled Lena River main stem and 20 tributaries are located along a 2600 km latitudinal
98 transect SW to NE and include watersheds of distinct sizes, geomorphology, permafrost extent, lithology,
99 climate and vegetation (Fig. 1, S1 A; Table S1). The total watershed area of the rivers sampled in this
100 work is approximately 1.66 million km², representing 66% of the entire Lena River basin. Permafrost is
101 mostly continuous except some patches of discontinuous and sporadic in the southern part of the Lena
102 basin (Brown et al., 2002). The mean annual air temperatures (MAAT) along the transect ranges from -
103 5 °C in the southern part of the Lena basin to -9 °C in the central part of the basin. The range of MAAT
104 for 20 tributaries is from -4.7 to -15.9 °C. The mean annual precipitation ranges from 350-500 mm y⁻¹ in
105 the southern and south-western part of the basin to 200-250 mm y⁻¹ in the central and northern parts
106 (Chevychelov and Bosikov, 2010). The lithology of the Siberian platform which is drained by the Lena
107 River is highly diverse and includes Archean and Proterozoic crystalline and metamorphic rocks, Upper
108 Proterozoic, Cambrian and Ordovician dolostones and limestones, volcanic rocks of Permo-Triassic age
109 and essentially terrigenous silicate sedimentary rocks of the Phanerozoic. Further description of the Lena
110 River basin landscapes, vegetation and lithology can be found elsewhere (Rachold et al., 1996; Huh et
111 al., 1999a, b; Pipko et al., 2010; Semiletov et al., 2011; Kutscher et al., 2017; Juhls et al., 2020).

112 The peak of annual discharge depends on the latitude (**Fig. 1**) and occurs in May in the south
113 (Ust-Kut) and in June in the middle and low reaches of the Lena River (Yakutsk, Kysyr). From May 29
114 to June 17, 2016, we moved downstream the Lena River by boat with an average speed of 30 km h⁻¹
115 (Gureyev, 2016). As such, we followed the progression of **the** spring and moved from the **southwest** to
116 the **northeast**, thus **collecting river** water at approximately the same stage of maximal discharge. Note
117 that transect sampling is a common way to assess river water chemistry in extreme environments (Huh
118 and Edmond, 1999; Spence and Telmer, 2005), and generally, a single sampling during high flow season
119 provides the best agreement with time-series estimates (Qin et al., 2006). Regular stops each 80-100 km
120 along the Lena River allowed sampling for major hydrochemical parameters and CH₄ **along** the main
121 stem. We also moved 500-1500 m upstream of selected tributaries to record **CO₂ concentrations** for at
122 least 1 h and to sample for river hydrochemistry; see examples of spatial coverage in **Fig. S1 B**. From
123 late afternoon/evening to the next morning, we stopped for sleep but continued to record pCO₂ in the
124 Lena River main stem (15 sites, evenly distributed over the full 2600 km transect) and two tributaries
125 (Aldan and Tuolba).

126

127 2.2. CO₂ and CH₄ concentrations

128 **Surface water CO₂ concentration** was measured continuously, *in-situ* by **deploying a portable**
129 infrared gas analyzer (IRGA, GMT222 CARBOCAP® probe, Vaisala®; accuracy ± 1.5%) of two ranges
130 (2 000 and 10 000 ppm). This system was mounted on a small boat in a perforated steel pipe ~0.5 m
131 below water surface. The tube had two necessary opening of different diameter, which allowed free water
132 flow with a constant rate during the moving of the boat. The probe was enclosed within a waterproof and
133 gas-permeable membrane. **The key to aqueous deployment of the IRGA sensor is the use of a protective**
134 **expanded polytetrafluoroethylene (PTFE) tube or sleeve that is highly permeable to CO₂ but**
135 **impermeable to water (Johnson et al., 2009). The material is available for purchase as a flexible tube that**
136 **fits over the IRGA sensor (Product number 200-07; International Polymer Engineering, Tempe, Arizona,**
137 **USA). We also used a copper mesh screen to minimize biofouling effects (i.e., Yoon et al., 2016).**
138 **However these effects are expected to be low in cold waters of the virtually pristine Lena River and its**

139 tributaries. During sampling, the sensor was left to equilibrate in the water for 10 minutes before
140 measurements were recorded.

141 The probe was enclosed and placed into a tube which was submerged 0.5 m below the water
142 surface. Within this tube, we designed a special chamber that allowed low-turbulent water flow around
143 the probe without gas bubbles. Previous studies (Park et al., 2021; Crawford et al. 2015; Yoon et al.,
144 2016) reported some effects of boat speed on sensor CO₂ measurements due to turbulences. Although
145 the turbulences were minimized in the tube/chamber design used in the present study, on a selected river
146 transect (~10 km) we have also tested the impact of the boat speed (5, 10, 20, 30 and 40 km h⁻¹) on the
147 sensor performance and have not detected any sizable (> 10%, p < 0.05, n = 25) difference in the CO₂
148 concentrations recorded by our system.

149 A Campbell logger was connected to the system allowing continuous recording of the CO₂
150 concentration (ppm), water temperature (°C) and pressure (mbar) every minute during 5 minutes over 10
151 minute intervals yielding 4,285 individual pCO₂, water temperature and pressure measurements in total.
152 These data were averaged for 3 consecutive slots of 5 min measurements, which represented the
153 approximate 20-km interval of the main stem route. CO₂ concentrations in the Lena River tributaries
154 were measured over the first 500-2000 m distance upstream of the tributary mouth, and comprised
155 between 5 and 34 measurements for day-time visits and between 305 and 323 individual pCO₂ readings
156 for each tributary for day-time and night-time monitoring.

157 Sensor preparation was conducted in the lab following the method described by Johnson et al.
158 (2009). The measurement unit (MI70, Vaisala®; accuracy ± 0.2%) was connected to the sensor allowing
159 instantaneous readings of pCO₂. The sensors were calibrated in the lab against standard gas mixtures (0,
160 800, 3 000, 8 000 ppm; linear regression with R² > 0.99) before and after the field campaign. The sensors'
161 drift was 0.03-0.06% per day and overall error was 4-8% (relative standard deviation, RSD). Following
162 calibration, post-measurement correction of the sensor output induced by changes in water temperature
163 and barometric pressure was done by applying empirically derived coefficients following Johnson et al.
164 (2009). These corrections never exceeded 5% of the measured values. Furthermore, we tested two
165 different sensors in several sites of the river transect: a main probe used for continuous measurements

166 and another probe used as a control and never employed for continuous measurements. We did not find
167 any sizable (>10%) difference in measured CO₂ concentration between these two probes.

168 For CH₄ analyses, unfiltered water was sampled in 60-mL Serum bottles and closed without air
169 bubbles using vinyl stoppers and aluminum caps and immediately poisoned by adding 0.2 mL of
170 saturated HgCl₂ via a two-way needle system. In the laboratory, a headspace was created by displacing
171 approx. 40% of water with N₂ (99.999%). Two 0.5-mL replicates of the equilibrated headspace were
172 analyzed for their concentrations of CH₄, using a Bruker GC-456 gas chromatograph (GC) equipped with
173 flame ionization and thermal conductivity detectors. After every 10 samples, a calibration of the detectors
174 was performed using Air Liquid gas standards (i.e. 145 ppmv). Duplicate injection of the samples showed
175 that results were reproducible within ±5%. The specific gas solubility for CH₄ (Yamamoto et al., 1976)
176 was used in calculation of total CH₄ content in the vials and then recalculated to μmol L⁻¹ of the initial
177 waters.

178

179 *2.3. Chemical analyses of the river water*

180 The dissolved oxygen (CelloX 325; accuracy of ±5%), specific conductivity (TetraCon 325;
181 ±1.5%), and water temperature (±0.2 °C) were measured in-situ at 20 cm depth using a WTW 3320
182 Multimeter. The pH was measured using portable Hanna instrument via combined Schott glass electrode
183 calibrated with NIST buffer solutions (4.01, 6.86 and 9.18 at 25°C), with an uncertainty of 0.01 pH units.
184 The temperature of buffer solutions was within ± 5°C of that of the river water. The water was sampled
185 in pre-cleaned polypropylene bottle from 20-30 cm depth in the middle of the river and immediately
186 filtered through disposable single-use sterile Sartorius filter units (0.45 μm pore size). The first 50 mL of
187 filtrate was discarded. The DOC and Dissolved Inorganic Carbon (DIC) were determined by a Shimadzu
188 TOC-VSCN Analyzer (Kyoto, Japan) with an uncertainty of 3% and a detection limit of 0.1 mg/L. Blanks
189 of MilliQ water passed through the filters demonstrated negligible release of DOC from the filter
190 material.

191

192

193 2.4. Flux calculation

194 CO₂ flux (F_{CO_2}) was calculated following Cai and Wang (1998):

$$195 F_{CO_2} = K_h k_{CO_2} (C_{water} - C_{air}), \quad (1)$$

196 where K_h is the Henry's constant corrected for temperature and pressure ($\text{mol L}^{-1} \text{atm}^{-1}$), k_{CO_2} is the gas
197 exchange velocity at a given temperature, C_{water} is the water CO₂ concentration, and C_{air} is the CO₂
198 concentration in the ambient air. We used the average CO₂ concentrations of 402 ppm in May-June 2016
199 (from 129 stations all over the world, <https://community.wmo.int/wmo-greenhouse-gas-bulletins>), which
200 is consistent with the value recorded at the nearest Tiksi station in 2016 (404 ± 0.9 ppm, Ivakhov et al.,
201 2019). Temperature-specific solubility coefficients were used to calculate respective CO₂ concentrations
202 in the water following Wanninkhof et al. (1992). To standardize k_{CO_2} to a Schmidt number of 600, we
203 used the following equation (Alin et al., 2011; Vachon et al., 2010):

$$204 k_{600} = k_{CO_2} \left(\frac{600}{Sc_{CO_2}} \right)^{-n} \quad (2)$$

205 where Sc_{CO_2} is CO₂ Schmidt number for a given temperature (t , °C) in the freshwater (Wanninkhof, 1992):

$$206 Sc_{CO_2} = 1911.1 - 118.11t + 3.4527t^2 - 0.041320t^3 \quad (3)$$

207 The exponent n (Eqn. 2) is a coefficient that describes water surface (2/3 for a smooth water surface
208 regime while 1/2 for a rippled and a turbulent one), and the Schmidt number for 20°C in freshwater is
209 600. We used $n = 2/3$ because all water surfaces of sampled rivers were considered flat and had a laminar
210 flow (Alin et al., 2011; Jähne et al., 1987) with wind speed always below 3.7 m s^{-1} (Guérin et al., 2007).

211 In this study, we used a k_{CO_2} (a median gas transfer coefficient) value of 4.464 m d^{-1} measured in
212 the 4 largest rivers of Western Siberia Lowland (WSL) in June 2015 (Ob', Pur, Pyakupur and Taz rivers,
213 Karlsson et al., 2021). These rivers are similar to Lena and its tributaries in size, but exhibit lower velocity
214 than those of the Lena River. In fact, due to more mountainous relief, the Lena River main stem and
215 tributaries present much higher turbulence than that of the Ob River and tributaries and as such the value
216 k_{CO_2} used in this study can be considered rather conservative. This value is consistent with the k_{CO_2}
217 reported for the Kolyma River and its large tributaries ($k = 3.9 \pm 2.5 \text{ m d}^{-1}$, Denfeld et al., 2013),

218 tributaries and main stem of the Yukon river basin ($k_{600} = 4.9 - 7.6 \text{ m d}^{-1}$, Striegl et al. 2012), large rivers
219 in the Amazon and Mekong basins ($k_{600} = 3.5 \pm 2.1 \text{ m d}^{-1}$, Alin et al., 2011) and with modelling results
220 of k for large rivers across the world ($k = 3 - 4 \text{ m d}^{-1}$, Raymond et al., 2013). Note that decreasing the k
221 to most conservative value of 3 m d^{-1} of Raymond et al. (2013) will decrease specific emissions by ca.
222 30 %.

223 Instantaneous diffusive CH_4 fluxes were calculated using an equation similar to 1 with k from
224 western Siberia rivers (Serikova et al., 2018), concentrations of dissolved CH_4 in the water and air–water
225 equilibrium pCH_4 concentration of 1.8 ppm, and mean annual pCH_4 concentration in the air for 2016
226 (Mauna Loa Observatory ftp://aftp.cmdl.noaa.gov/products/trends/ch4/ch4_annmean_gl.txt) following
227 standard procedures (Serikova et al., 2018, 2019).

228

229 2.5. Landscape parameters and water surface area of the Lena basin

230 The physio-geographical characteristics of the 20 Lena tributaries sampled in this study and the
231 two points of the Lena main stem (upstream and downstream r. Aldan, **Table S1**) were determined by
232 applying available digital elevation model (DEM GMTED2010), soil, vegetation, lithological, and
233 geocryological maps. The landscape parameters were typified using TerraNorte Database of Land Cover
234 of Russia (Bartalev et al., 2020, <http://terranorte.iki.rssi.ru>). This included various type of forest
235 (evergreen, deciduous, needleleaf/broadleaf), grassland, tundra, wetlands, water bodies and other area.
236 The climate and permafrost parameters of the watershed were obtained from CRU grids data (1950-2016)
237 (Harris et al., 2014) and NCSCD data ([doi:10.5879/ecds/00000001](https://doi.org/10.5879/ecds/00000001), Hugelius et al., 2013), respectively,
238 whereas the biomass and soil OC content were obtained from BIOMASAR2 (Santoro et al., 2010) and
239 NCSCD databases. The lithology layer was taken from GIS version of Geological map of the Russian
240 Federation (scale 1 : 5 000 000, <http://www.geolkarta.ru/>). To test the effect of carbonate rocks on
241 dissolved C parameters, we distinguished acidic crystalline, terrigenous silicate rocks and dolostones and
242 limestones of upper Proterozoic, Cambrian and Ordovician age. We quantified river water surface area
243 using the global SDG database with 30 m^2 resolution (Pekel et al., 2016) including both seasonal and
244 permanent water for the open water period of 2016 and for the multiannual average (reference period

245 2000-2004). We also used a more recent GRWL Mask Database which incorporates first order wetted
246 streams (Allen and Pavelsky, 2018).

247 The Pearson rank order correlation coefficient (R_s , $p < 0.05$) was used to determine the
248 relationship between CO_2 concentrations and climatic and landscape parameters of the Lena River
249 tributaries. Further statistical treatment of CO_2 , DIC and DOC concentration drivers in river waters
250 included a Principal Component Analysis which allowed to test the effect of various hydrochemical and
251 climatic parameters on dissolved C pattern. For the PCA treatment, all variables were normalized as
252 necessary in the standard package of STATISTICA-7 (<http://www.statsoft.com>) because the units of
253 measurement for various components were different. The factors were identified via the Raw Data
254 method. To run the scree test, we plotted the eigen values in descending order of their magnitude against
255 their factor numbers. There was significant decrease in the PCA values between F1 and F2 suggesting
256 that a maximum of two factors were interpretable.

257

258 3. Results

259 3.1. CO_2 , CH_4 , DIC and DOC in the main stem and Lena tributaries and C emission fluxes

260 The main hydrological C parameters of the Lena River and its tributaries (pCO_2 , CH_4 , pH, DIC,
261 and DOC) are listed in **Tables 1 and 2**. Continuous pCO_2 measurements in the main stem (4285
262 individual data points) averaged for each 20 km interval over the full distance of the boat route
263 demonstrated a sizable increase (from ca. 380 to 1040 μatm) in pCO_2 northward (**Fig. 2**). There was a
264 positive correlation between the pCO_2 and distance from the head waters of the Lena River ($r = 0.625$, p
265 < 0.01 , **Fig. 3 A**). The CH_4 concentration was low (0.054 ± 0.023 and $0.061 \pm 0.028 \mu\text{mol L}^{-1}$ in the Lena
266 River and 20 tributaries, respectively) and did not change appreciably along the main stem and among
267 the 20 tributaries (**Fig. 3 B**). The DOC concentration did not demonstrate any systematic variations over
268 the main stem ($10.5 \pm 2.4 \text{ mg L}^{-1}$, **Fig. 3 C**), however it was higher and more variable in tributaries (15.8
269 $\pm 8.6 \text{ mg L}^{-1}$). The DIC concentration decreased about five-fold from the head waters to the middle course
270 of the Lena River (**Fig. 3 D**), and pH decreased by 0.8 units downstream (**Fig. 3 E**).

271 Generally, the concentrations of DOC measured in the present study during the peak of the spring
272 flood are at the highest range of previous assessments during summer baseflow (around 5 mg L⁻¹; range
273 of 2 to 12 mg L⁻¹, Cauwet and Sidorov, 1996; Lara et al., 1998; Lobbes et al., 2000; Kuzmin et al., 2009;
274 Kutscher et al., 2017). The DIC concentration in the main stem during spring flood was generally lower
275 than that reported during summer baseflow (around 10 mg L⁻¹; range of 5 to 50 mg L⁻¹) but consistent
276 with values reported in Yakutsk during May and June period (7 to 20 mg L⁻¹, Sun et al., 2018). A sizable
277 decrease in DIC concentration between the headwaters (first 500 km of the river) and the Lena River
278 middle course was also consistent with the alkalinity pattern reported in previous works during summer
279 baseflow (Pipko et al., 2010; Semiletov et al., 2011). For the Lena river tributaries, the most
280 comprehensive data set on major ions was acquired in July-August of 1991-1996 by Huh and Edmond's
281 group (Huh and Edmond, 1999; Huh et al., 1998a, b) and by Sun et al. (2018) in July 2012 and at the end
282 of June 2013. For most tributaries, the concentration of DIC was a factor of 2 to 5 lower during spring
283 flood compared to summer baseflow. This result can be explained by the strong dilution of carbonate-
284 rich groundwaters feeding the river in spring high flow compared to summer low flow.

285 The measured pCO₂ in the river water and published (Karlsson et al., 2021) gas transfer
286 coefficient (4.46 m d⁻¹) allowed for calculation of the CO₂ fluxes over the full length of the studied main
287 stem (2600 km) and the sampled tributaries. Calculated CO₂ fluxes of the main stem and tributaries
288 ranged from zero and slightly negative (uptake) values in the most southern part of the Lena River and
289 certain tributaries (N Katyma), to between 0.5-2.0 g C m⁻² d⁻¹ in the rest of the main stem and tributaries
290 (Tables 1, 2; Fig. 2 B). The largest part of the Lena River main stem, 1429 km from Kirenga to Tuolba,
291 exhibited quite stable flux of 1.1±0.2 g C m⁻² d⁻¹. In the last ~400 km part of the Lena River main stem
292 studied in this work, from Tuolba to Aldan, the calculated fluxes increased to 1.7±0.08 g C m⁻² d⁻¹.

293 The river water concentrations of dissolved CH₄ in the tributaries and the main channel
294 (0.059±0.006; IQR range from 0.025 to 0.199 μmol L⁻¹, Table 1, 2) did not exhibit any trend with
295 distance from headwaters or landscape parameters of the catchments. These values are consistent with
296 the range of CH₄ concentration in the low reaches of the Lena River main channel (0.03-0.085 μmol L⁻¹

297 ¹; Bussman, 2013) and are 100-500 times lower than those of CO₂. Consequently, diffuse CH₄ emissions
298 constituted less than 1 % of total C emissions and are not discussed in further detail.

299

300 *3.2. Diurnal (night/day) pCO₂ variations and spatial variations across the river transect*

301 The diel continuous CO₂ measurements of 3 tributaries (Kirenga, Tuolba and Aldan) and 14 sites
302 of the Lena River main channel showed generally modest variation with diurnal range within 10 % of
303 the average pCO₂ (Fig. 4 and Fig. S2). The observed variations in pCO₂ between day and night were not
304 linked to water temperature ($p > 0.05$), which did not vary more than 1-2 °C between the day and night
305 period.

306 The spatial variations of hydrochemical parameters were tested in the upper reaches of the Lena
307 main stem and its largest tributary - the Aldan River (Fig. S3). In the Lena River, over a lateral distance
308 of 550 m across the river bed, the pCO₂ and CH₄ concentrations were equal to 569±4.6 µatm and
309 0.0406±0.0074 µmol L⁻¹, respectively, whereas the DIC and DOC concentrations varied < 15% (n = 5).
310 In the Aldan River, over a 2700 m transect across the flow, the pCO₂ and CH₄ concentrations were equal
311 to 1035±95 µatm and 0.078±0.00894 µmol L⁻¹, respectively, whereas DIC and DOC varied within < 20%
312 (n = 4). Overall, these results supported our design of punctual (snap shot) sampling in the middle of the
313 river.

314

315 *3.3. Impact of catchment characteristics on pCO₂ in tributaries of the Lena River*

316 The CO₂ concentration in the Lena River main stem and tributaries increased from southwest to
317 northeast (Table 1, 2; Fig. 2), and this was reflected in a positive (R = 0.66) correlation between CO₂
318 concentration and continuous permafrost coverage and a negative (R = -0.76) correlation with MAAT
319 (Table 3). Among different landscape factors, C stock in upper 0-30 and 0-100 cm of soil, the proportion
320 of riparian vegetation and bare rocks, the coverage by deciduous needle-leaf forest, and coverage of river
321 watershed by water bodies (mostly lakes) exhibited significant ($p < 0.01$, $n = 19$) positive correlations
322 ($0.54 \leq R \leq 0.86$) with average pCO₂ of the Lena River tributaries (Fig. 5). The other potentially important
323 landscape factors of the river watershed (proportion of peatland and bogs, tundra coverage, total

324 aboveground vegetation, type of permafrost, annual precipitation) did not significantly impact the CO₂
325 concentration in the Lena River tributaries (**Table 3**).

326 Further assessment of landscape factor control on C parameters of the river water was performed
327 via a PCA. This analysis basically confirmed the results of linear regressions and revealed two factors
328 capable explaining only 12.5 and 3.5% of variability (**Fig. S4**). The F1 strongly acted on the sample
329 location at the Lena transect, the content of OC in soils, the watershed coverage by deciduous needle-
330 leaf forest and shrubs, riparian vegetation (a proxy for the width of the riparian zone), but also proportion
331 of tundra, bare rock and soils, water bodies, peatland and bogs (> 0.90 loading). The pCO₂ was
332 significantly linked to F1 (0.72 loading).

333

334 *3.4. Areal emission from the Lena River basin*

335 The areal emission of CO₂ from the lotic waters of the Lena River watershed were assessed based
336 on total river water coverage of the Lena basin in 2016 (28,197 km², of which 5,022 km² is seasonal
337 water, according to the Global SDG database). This value is consistent with the total river surface area
338 from the GRWL Mask database (22,479 km²). Given that the measurements were performed at the peak
339 of spring flood in 2016, we used the maximal water coverage of the Lena River basin.

340 Based on past calculated pCO₂ of the Lena River (400 - 1000 μatm, Semiletov, 1999; Semiletov
341 et al., 2011; Pipko et al., 2010) both the seasonality and spatial differences downstream are relatively
342 small. Indeed, for the lower reaches of the Lena River, from Yakutsk to the Lena Delta, Semiletov (1999)
343 and Semiletov et al. (2011) reported, for August-September 1995, the average pCO₂ of 538±96 μatm
344 (range 380-727 μatm). This value is very similar to the one obtained in July 2003 for the low reaches of
345 Lena (559 μatm, Pipko et al., 2010). Over the full length of the Lena River, from Ust-Kut to the Lena
346 mouth, Pipko et al. (2010) reported an average pCO₂ of 450 ± 100 μatm in June-July 2003. At the same
347 time, calculated pCO₂ from previous field campaigns are generally lower than the pCO₂ of the Lena
348 River main stem directly measured in this study: 700-800 μatm for the Ust Kut – Nuya segment (1331
349 km); 845 – 1050 μatm for the Nuya – Aldan segment (1050 km).

350 Thus, despite the absolute values of calculated pCO₂ involving uncertainties (our calculated:
351 measured pCO₂ in Lena River main channel and tributaries equaled 0.67±0.15 (n = 47)), this suggests
352 spatial and temporal stability of the pCO₂ in the Lena River waters and allows for extrapolation of the
353 measured pCO₂ in the Lena River from Yakutsk to Aldan to the lower reaches of the river. As for the
354 Lena tributaries, to the best of our knowledge there is no published information on pCO₂ concentration
355 and emission. Overall, the major uncertainty in estimation of the Lena River basin emission stems from
356 a lack of direct pCO₂ measurements in the northern part of the main channel over ca. 1000 km
357 downstream of the Aldan River including the large tributary Vilyi. Further, we noted that the largest
358 northern tributary, the Aldan River providing 70% of the spring time discharge of the Lena River (Pipko
359 et al., 2010), demonstrated sizably higher emissions compared to the Lena River main channel upstream
360 of Aldan (3.2±0.5 and 1.69±0.08 g C m⁻² d⁻¹, respectively).

361 For areal emission calculations, we used the range of CO₂ emissions from 1 to 2 g C m⁻² d⁻¹ which
362 covers full variability of both large and small tributaries and the Lena River main channel (Tables 1-2,
363 Figure 2 B). This estimation assumes lack of pCO₂ dependence on the size of the watershed in the Lena
364 basin as confirmed by our data (Fig. S5). For an alternative areal emission calculations, we explicitly
365 took into account the water area of the main stem (43% relative to the total water area of the Lena
366 catchment) and we introduced the partial weight of emission from the 3 largest tributaries (Aldan,
367 Olekma and Vitim) according to their catchment surface areas (43, 12 and 14% of all sampled territory,
368 respectively). We summed up contribution of the Lena river main stem and the tributaries and we
369 postulated the average emission from the main stem upstream of Aldan (1.25±0.30 g C m⁻² d⁻¹) as
370 representative of the whole Lena River. This resulted in an updated value of 1.65±0.5 g C m⁻² d⁻¹ which
371 is within the range of 1 to 2 g C m⁻² d⁻¹ assessed previously. Note that this value is most likely
372 underestimated because emissions from the main stem downstream of Aldan are at least 10 % higher
373 (Table 1, Fig. 1 B).

374 For the two months of maximal water flow (middle of May - middle of July), the C emission from
375 the whole Lena basin equates to 2.8 ± 0.85 Tg C which is 20 to 30% of the DOC and DIC lateral export
376 to the Arctic Ocean. Assuming six months of open water period and no emission during winter, this

377 yields $8.3 \pm 2.5 \text{ Tg C y}^{-1}$ of annual emission for the whole Lena River basin (2,490, 000 km²) with a total
378 lotic water area of 28,100 km². Considering the only 23,200 km² water area in July-October (and maximal
379 water coverage in May-June), these numbers decrease by 12% which is below the uncertainties
380 associated with our evaluation.

381

382

383 4. DISCUSSION

384 4.1. Possible driving factors of CO₂ pattern in the Lena River basin

385 Generally, the SW to NE increase in CO₂ concentrations and fluxes of the tributaries was
386 consistent with CO₂ pattern in the main stem (**Fig. 2; Tables 1, 2**), and thus can be considered as a
387 specific feature of CO₂ exchange between lotic waters and atmosphere in the studied part of the Lena
388 Basin. At the same time, there were sizable local variations (peaks) in CO₂ concentration of the main
389 stem along the sampling route (**Fig. 2 A**). Peaks shown on the diagram of the main stem are not
390 necessarily linked to CO₂-rich tributaries, but likely reflect local processes in the main stem, including
391 lateral influx from the shores and shallow subsurface waters, which is typical for permafrost regions of
392 forested Siberian watersheds (i.e., Bagard et al., 2011). Given that the data were averaged over ~20-km
393 distance, we believe that these peaks are not artifacts but reflect local heterogeneity of the pCO₂ pattern
394 in the main stem (turbulences, suprapermafrost water discharge, sediment resuspension and respiration).
395 Note that such a heterogeneity was not observed in the tributaries, at least at the scale of our spatial
396 coverage (see **Fig. S1 B, S3**).

397 The PCA demonstrated extremely low ability to describe the data variability (12% by F1 and only
398 3.5% by F2). We believe that the most likely reason of weak PCA capacity is the rather homogeneous
399 distribution of CO₂ and CH₄ among the tributaries, primarily linked to the specific hydrological period,
400 studied in this work - the spring flood. During this high flow period, the local lithological and soil
401 heterogeneities among tributaries or the segments of the main stem virtually disappear and surface flow
402 (via vegetation leaching) becomes the most important driver of riverine chemistry, as is known from

403 adjacent permafrost territories in Central Siberia (i.e., Bagard et al., 2011). Nevertheless, some specific
404 features of the data structure could be established. The first factor, significantly linked to pCO₂ (0.72
405 loading), strongly acted on the sample location at the Lena transect, the watershed coverage by deciduous
406 needle-leaf forest and shrubs, riparian vegetation, and also the proportion of tundra, bare rock and soils,
407 water bodies, peatland and bogs (> 0.90 loading). This is fully consistent with spatial variation of pCO₂
408 along the permafrost and climate gradient in the main channel and sampled tributaries. Positive loading
409 of riparian vegetation, peatlands and bogs on F1 (0.927 and 0.989, respectively) could reflect a
410 progressive increase in the feeding of the river basin by mire waters, an increase in the proportion of
411 needle-leaf deciduous trees, and in the width of the riparian zone from the SW to the NE direction.

412 Lack of sizable variation in pCO₂ between the day and night period or across the river bed
413 suggests quite low site-specific and diurnal variability. It may be indicative of a negligible role of primary
414 productivity in the water column given the low water temperatures, shallow photic layer of organic-rich
415 and turbid waters and lack of periphyton activity during high flow of the spring flood. The pCO₂
416 increased by a factor of 2 to 4 along the permafrost/temperature gradient from the southwest to the
417 northeast, for both the main channel and sampled tributaries. This may reflect progressive increase in the
418 feeding of the river basin by mire waters, increase in the proportion of needle-leaf deciduous forest, and
419 an increase in the width of the riparian zone. Another strong correlation is observed between the stock of
420 OC in soils (both 0-30 and 0-100 cm depth) and the pCO₂ of Lena tributaries. Organic-rich soils are
421 widely distributed in the central and northern part of the basin. The most southern part of the Lena basin
422 is dominated by carbonate rocks and crystalline silicates in generally mountainous terrain, where only
423 thin mineral soils are developed. The northern (downstream of the Olekma River) part of the basin
424 consists of soils developed on sedimentary silicate rocks as well as vast areas of easily eroded yedoma
425 soils. It is likely that both organic matter mineralization in OC-rich permafrost soils and lateral export of
426 CO₂ from these soils, together with particulate and dissolved OC export and mineralization in the water
427 column, are the main sources of CO₂ in the river water. Although some studies have demonstrated high
428 lability of DOM in arctic waters (Cory et al., 2014; Ward et al., 2017; Cory and Kling, 2018), others
429 suggest it is low and does not support the major part of CO₂ supersaturation in water (Shirokova et al.,

430 2019; Payandi-Rolland et al., 2020; Laurion et al., 2021). Note that we have not observed any significant
431 relationship between the DOC and pCO₂ in the Lena River and tributaries (**Fig. S6 A**). Lack of such a
432 correlation and absence of diurnal pCO₂ variations imply that in-stream processing of dissolved terrestrial
433 **OC** is not the main driver of CO₂ supersaturation in the river waters of the Lena River basin. **Furthermore,**
434 **a lack of lateral (across the river bed) variations in pCO₂ does not support a sizable input of soil waters**
435 **from the shore, although we admit that much higher spatial coverage along the river shore is needed to**
436 **confirm this hypothesis.**

437 The role of underground water discharge in regulating pCO₂ pattern of the tributaries is expected
438 to be most pronounced in the SW part of the basin (Lena headwaters), where carbonate rocks of the
439 basement would provide sizable amounts of CO₂ discharge in the river bed. However, there was no
440 **relationship between** the proportion of carbonate rocks on the watershed and the pCO₂ in the tributaries
441 (**Fig. S6 B**). **Furthermore,** for the Lena River main stem, the lowest CO₂ concentrations were recorded in
442 the upper reaches (first 0-800 km) **where carbonate** rocks dominate. Altogether, this **makes the** impact of
443 CO₂ from underground carbonate reservoirs on river water CO₂ concentrations **unlikely**. **This is further**
444 **illustrated by a lack of correlation between pCO₂ and DIC or pH (Fig. S7 A of the Supplement).** The pH
445 **did not control the CO₂ concentration in the main stem and only weakly impacted the CO₂ in the**
446 **tributaries (Fig. S7 B).** The latter could reflect an increase in pCO₂ in the northern tributaries which
447 **exhibited generally lower pH compared to the SW tributaries hosted within the carbonate rocks. Overall,**
448 **such low correlations of CO₂ with DIC and pH reflected a generally low predictive capacity to calculate**
449 **pCO₂ from measured pH, temperature and alkalinity (see section 3.4).**

450 Therefore, other sources of riverine CO₂ may include **particulate organic carbon** processing in the
451 water column (Attermeyer et al., 2018), river sediments (Humborg et al., 2010) and within the riparian
452 zone (Leith et al., 2014, 2015) which require further investigation. **In addition,** although there was no
453 sizable variation in pCO₂ between the day and night period or across the river bed, the flux could show
454 different spatial and temporal patterns if *k* shows larger variability (cf., Beaulieu et al., 2012). This calls
455 for a need of direct flux measurements in representative rivers and streams of the Lena River basin.
456 **Overall, the present study demonstrates highly dynamic and non-equilibrium behavior of CO₂ in the river**

457 waters, with possible *hot spots* from various local sources. For these reasons, *in-situ*, high spatial
458 resolution measurements of CO₂ concentration in rivers—such as those reported for the Lena Basin in
459 this study—are crucially important for quantifying the C emission balance in lotic waters of high
460 latitudes.

461

462 4.2. Areal emission from the Lena River basin vs lateral export to the Arctic Ocean

463 The estimated CO₂ emissions from the Lena River main channel over 2600 km distance (0.8 –
464 1.7 g C m⁻² d⁻¹) are comparable to values directly measured in rivers and streams of the continuous
465 permafrost zone of western Siberia (0.98 g C m⁻² d⁻¹, Serikova et al., 2018), the Kolyma River (0.35 g C
466 m⁻² d⁻¹ in the main stem; 2.1 g C m⁻² d⁻¹ for lotic waters of the basin), and the Ob River main channel
467 (1.32±0.14 g C m⁻² d⁻¹ in the permafrost-free zone, Karlsson et al., 2021). At the same time, the Lena
468 River flux (FCO₂) values are lower than typical emissions from running waters in the contiguous United
469 States (3.1 g C m⁻² d⁻¹, Hotchkiss et al., 2015), small mountain streams in Northern Europe (3.3 g C m⁻²
470 d⁻¹, Rocher-Ros et al., 2019), and small streams of the northern Kolyma River (6 to 7 g C m⁻² d⁻¹, Denfeld
471 et al., 2013) and Ob River in the permafrost-affected zone (3.8 to 5.4 g C m⁻² d⁻¹, Karlsson et al., 2021).
472 In contrast to the main stem, the range of FCO₂ in the tributaries is larger (0.2 to 3.2 g C m⁻² d⁻¹) and
473 presumably reflects a strong variability in environmental conditions across a sizable landscape and
474 climate transect.

475 Total C emissions from other major Eastern Eurasian permafrost-draining rivers (i.e. sum of
476 Kolyma, Lena and Yenisei rivers) based on indirect estimates (40 Tg C y⁻¹, Raymond et al., 2013) are
477 generally supportive of the estimations of the Lena River in this study (5 to 10 Tg C y⁻¹). At the same
478 time, the C emission from the Lena river basin (28,100 km² water area) are lower than those of the lotic
479 waters of western Siberia (30 Tg C y⁻¹ for 33,389 km² water area, Karlsson et al., 2021). The latter drain
480 through thick, partially frozen peatlands within the discontinuous, sporadic and permafrost-free zones,
481 which can cause high OC input and processing and, thus, enhanced C emissions (Serikova et al., 2018).

482 Despite the high uncertainty on our regional estimations [due to lack of directly measured gas
483 transfer values and low seasonal resolution] we believe that these estimations are conservative and can

484 be considered as first order values pending further improvements. In order to justify extrapolation of our
485 data to all seasons and the entire area of the Lena basin, we analyzed data for spatial and temporal
486 variations in pCO₂ of the Lena River main stem from available literature. From the literature there were
487 three important findings. First, based on published data, the seasonal and spatial variabilities of pCO₂
488 across the majority of the Lena River main stem are not high during open water period, although the low
489 reaches of the Lena River may exhibit higher emissions compared to the middle and upper course (see
490 section 3.4). Second, although small mountainous headwater streams of the tributaries may exhibit high
491 *k* due to turbulence, this could be counteracted by lower CO₂ supply from low OC in mineral soil, lack
492 of riparian zone and scarce vegetation. Third, although these small streams (watershed area < 100 km²)
493 may represent > 60% of total watershed surfaces of the Lena basin (Ermolaev et al., 2018), their
494 contribution to the total water surface is < 20% (19% from combined analysis of DEM GMTED2010 and
495 16% from the GRWL or Global SDG database as estimated in this study). Therefore, given that (i) within
496 the stream-river continuum, the CO₂ efflux increases only two-fold demonstrating a discharge decrease
497 by a factor of 10,000 (from 100 to 0.01 m³ s⁻¹, Hotchkiss et al., 2015), and (ii) the watershed area had no
498 impact on pCO₂ in the river water (Fig. S5), this uncertainty is likely less important. As such, instead of
499 integrating indirect literature data, we used the pCO₂ values measured in the present study to calculate
500 the overall CO₂ emission from all lotic waters of the Lena basin.

501 The C evasion from the Lena basin assessed in the present work is comparable to the total
502 (DOC+DIC) lateral export by the Lena River to the Arctic Ocean (10 Tg C y⁻¹ by Semiletov et al. (2011),
503 or 11 Tg C y⁻¹ (5.35 Tg DIC y⁻¹ + 5.71 Tg DOC y⁻¹ by Cooper et al. (2008)). Moreover, the C evasion
504 strongly exceeds sedimentary C input to the Laptev Sea by all Siberian rivers (1.35 Tg C y⁻¹, Rachold et
505 al. (1996) and Dudarev et al. (2006)), the Lena River annual discharge of particulate organic carbon (0.38
506 Tg y⁻¹, Semiletov et al., 2011), and OC burial on the Kara Sea Shelf (0.37 Tg C y⁻¹, Gebhardt et al., 2005).

507 Typical concentrations of CH₄ in the Lena tributaries and the main channel are 100 to 500 times
508 lower than those of CO₂. Given that the global warming potential (GWP) of methane on a 100-year scale
509 is only 25 times higher than that of CO₂, the long-term role of diffuse methane emission from the Lena
510 River basin is still 4 to 20 times lower than that of CO₂. However, on a short-term scale (20 years), the

511 GWP of methane can be as high as 96 (Alvarez et al., 2018) and its role in climate regulation becomes
512 comparable to that of the CO₂. This has to be taken into account for climate modeling of the region.

513 The follow up studies of this large heterogenous and important system should include CO₂
514 measurements in 1) the low reaches of the Lena River, downstream of Aldan, notably large organic-rich
515 tributaries such as Vilyi (454,000 km²) and where the huge floodzone (20-30 km wide) with large number
516 of lakes and wetlands is developed, and 2) highly turbulent eastern tributaries of the Lena River
517 downstream of Aldan, which drain the Verkhoyansk Ridge and are likely to exhibit elevated gas transfer
518 coefficients.

519

520 5. Conclusions

521 Continuous pCO₂ measurements over 2600 km of the upper and middle part of the Lena River
522 main channel and 20 tributaries during the peak of spring flood allowed to quantify, for the first time, in-
523 situ pCO₂ variations which ranged from 500 to 1700 μatm and exhibited a 2 to 4-fold increase in CO₂
524 concentration northward. There was no major variation in pCO₂ between the day and night period or
525 across the river bed which supports the chosen sampling strategy. The northward increase in pCO₂ was
526 correlated with an increased proportion of needle-leaf deciduous trees, the width of the riparian zone and
527 the stock of organic C in soils. Among the potential drivers of riverine pCO₂, changes in the vegetation
528 pattern (northward migration of larch tree line in Siberia; Kruse et al., 2019) and soil OC stock are likely
529 to be most pronounced during ongoing climate warming and thus the established link deserves further
530 investigation. The total C emission from the lotic waters of the Lena River basin ranges from 5 to 10 Tg
531 C y⁻¹ which is comparable to the annual lateral export (50% DOC, 50 % DIC) by the Lena River to the
532 Arctic Ocean. However, these preliminary estimations of C emission should be improved by direct flux
533 measurements across seasons in different types of riverine systems of the basin, notably in the low
534 reaches of the Lena River.

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543

544 **Authors contribution.**

545 SV and OP designed the study and wrote the paper; SV, YK and OP performed sampling, analysis and
546 their interpretation; MK performed landscape characterization of the Lena River basin and calculated
547 water surface area; JK provided analyses of literature data, transfer coefficients for FCO₂ calculations
548 and global estimations of areal emission vs export.

549

550 **Competing interests.**

551 The authors declare that they have no conflict of interest.

552

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890 **Table 1.** Measured water temperature, pCO₂, calculated CO₂ flux, CH₄, DOC, and DIC concentrations
 891 and pH in the Lena River main stem (average ± s.d.; (n) is number of measurements).

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River transect	T _{water} , °C	pCO ₂ , µatm	FCO ₂ , g C m ⁻² d ⁻¹ k = 4.464
Lena upstream of Kirenga (0-578 km)	12.65±0.22 (99)	714±22 (99)	0.849±0.061 (99)
Lena Kirenga – Vitim (579-1132 km)	9.17±0.15 (87)	806±8.8 (87)	1.19±0.024 (87)
Lena Vitim -Nuya (1132-1331 km)	8.10±0.115 (27)	797±22 (27)	1.22±0.072 (27)
Lena Nuya – Tuolba (1331-2008 km)	9.61±0.09 (95)	846±12 (95)	1.29±0.034 (95)
Lena Tuolba – Aldan (2008-2381 km)	10.6±0.21 (52)	1003±28 (52)	1.69±0.081 (5)

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	CH ₄ , µmol L ⁻¹	DOC, mg L ⁻¹	DIC, mg L ⁻¹	pH
Lena upstream of Kirenga (0-578 km)	0.068±0.003 (6)	13.9±1.4 (6)	20.0±1.2 (6)	8.12±0.203 (7)
Lena Kirenga – Vitim (579-1132 km)	0.040±0.002 (12)	7.55±0.246 (14)	6.30±0.485 (14)	7.77±0.040 (14)
Lena Vitim -Nuya (1132-1331 km)	0.038±0.003 (5)	9.02±0.29 (3)	4.55±0.70 (3)	7.69±0.063 (3)
Lena Nuya – Tuolba (1331-2008 km)	0.037±0.002 (6)	10.4±0.78 (2)	5.09±1.157 (2)	7.62±0.052 (2)
Lena Tuolba – Aldan (2008-2381 km)	0.088±0.034 (5)	11.6±0.27 (5)	5.24±0.102 (5)	7.49±0.044 (5)

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917 **Table 2.** Measured water temperature, pCO₂, calculated CO₂ flux, CH₄, DOC, DIC concentration and
 918 pH in the tributaries (average ± s.d.; (n) is number of measurements).

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Tributary	T _{water} , °C	pCO ₂ , µatm	FCO ₂ , g C m ⁻² d ⁻¹
№4 Orlinga (208 km)	8.0±0.0 (13)	515±2.9 (13)	0.347±0.01 (13)
№5 Nijnaya Kitima (228 km)	6.8±0.0 (11)	462±9.4 (11)	0.193±0.03 (11)
№8 Taiur (416 km)	8.5±0.0 (10)	575±31 (10)	0.523±0.095 (10)
№10 Bol. Tira (529 km)	11.9±0.0 (15)	788±12 (15)	1.04±0.03 (15)
№12 Kirenga (579 km)	10.2±0.0 (323)	448±4 (323)	0.131±0.01 (323)
№25 Thcayka (1025 km)	8.6±0.01 (8)	856±13 (8)	1.37±0.04 (8)
№28 Tchuya (1110 km)	5.9±0.0 (5)	751±5.7 (5)	1.16±0.019 (5)
№29 Vitim (1132 km)	6.8±0.0 (10)	654±10 (10)	0.812±0.03 (10)
№32 Ykte (1265 km)	4.9±0.0 (11)	676±4.8 (11)	0.943±0.02 (11)
№34 Kenek (1312 km)	7.60±0.0 (11)	710±2.6 (11)	0.964±0.01 (11)
№36 Nuya (1331 km)	11.8±0.0 (10)	752±6.0 (10)	0.947±0.02 (10)
№38 Bol. Patom (1670 km)	6.9±0.0 (5)	730±12 (5)	1.05±0.04 (5)
№39 Biriuk (1712 km)	14.2±0.0 (5)	929±19 (5)	1.32±0.05 (5)
№40 Olekma (1750 km)	6.4±0.0 (11)	802±14 (11)	1.30±0.05 (11)
№43 Markha (1948 km)	17.5±0.0 (15)	844±15 (15)	0.998±0.03 (15)
№44 Tuolba (2008 km)	12.3±0.0 (305)	1181±6 (305)	2.08±0.02 (305)
№46 Siniaya (2118 km)	18.5±0.0 (24)	894±19 (24)	1.08±0.04 (24)
№48 Buotama (2170 km)	18.5±0.0 (24)	1160±25 (24)	1.66±0.06 (24)
№52-54 Aldan (2381 km)	14.8±0.02 (316)	1715±12 (316)	3.23±0.03 (316)

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934 **Table 2**, continued.

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	CH₄, μmol L⁻¹	DOC, mg L⁻¹	DIC, mg L⁻¹	pH
№4 Orlinga (208 km)	0.064	13.4	27.9	8.64
№5 Nijnaya Kitima (228 km)	0.033	16.7	13.1	8.48
№8 Taiur (416 km)	0.079	10.0	11.2	8.36
№10 Bol. Tira (529 km)	0.084	22.7	14.9	8.13
№12 Kirenga (579 km)	0.036	5.13	6.86	7.97
№25 Thcayka (1025 km)	0.066	16.7	22.5	8.30
№28 Tchuya (1110 km)	0.037	7.08	3.44	7.57
№29 Vitim (1132 km)	0.057	10.1	2.18	7.70
№32 Ykte (1265 km)	0.037	5.49	15.3	7.86
№34 Kenek (1312 km)	0.053	21.1	16.0	8.12
№36 Nuya (1331 km)	0.048	26.6	11.7	7.80
№38 Bol. Patom (1670 km)	0.026	6.99	4.56	7.76
№39 Biriuk (1712 km)	0.047	29.2	11.3	7.87
№40 Olekma (1750 km)	0.046	13.3	3.3	7.53
№43 Markha (1948 km)	0.088	27.4	10.9	8.00
№44 Tuolba (2008 km)	0.035	14.5	14.7	7.98
№46 Siniaya (2118 km)	0.113	33.2	7.73	7.97
№48 Buotama (2170 km)	0.124	12.2	31.6	8.45
№52-54 Aldan (2381 km)	0.088 (4)	9.07±0.75 (4)	6.67±0.13 (4)	7.59±0.02 (4)

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937 Footnote : in all tributaries except Aldan, there was only one measurement of CH₄, DOC, DIC and pH

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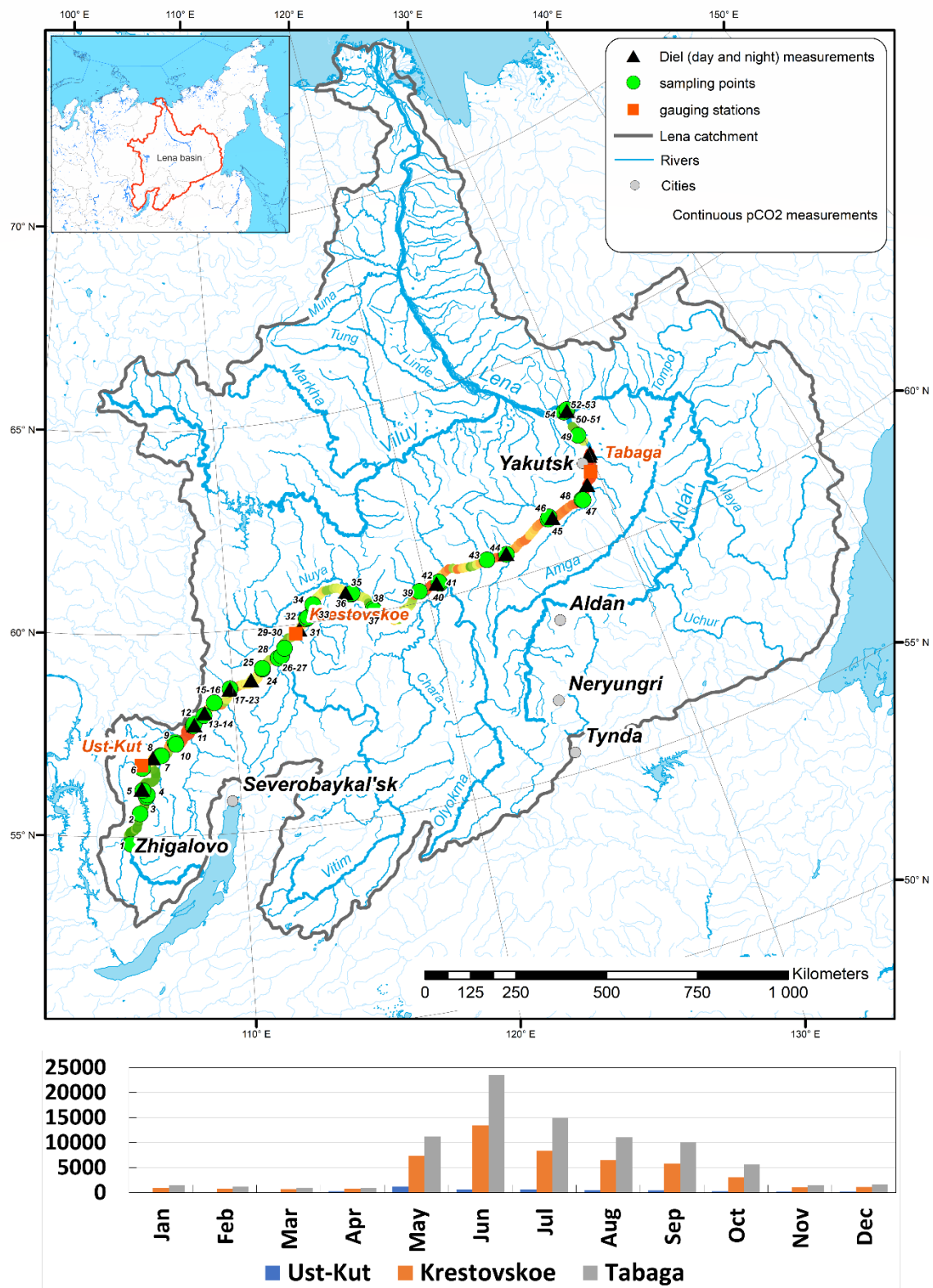
939 **Table 3.** Pearson correlations between pCO₂ and landscape parameters of the Lena tributaries.
 940 Significant correlations ($p < 0.05$) are marked by asterisk. Methane concentration did not exhibit any
 941 significant correlation with all tested parameters.

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% coverage of the watershed and climate	R_{Pearson}
Broadleaf Forest	0.04
Humid Grassland	-0.52*
Shrub Tundra	-0.05
Riparian Vegetation	0.87*
Croplands	-0.31
Bare Soil and Rock	0.54*
Evergreen Needle-leaf Forest	-0.59*
Deciduous Broadleaf Forest	-0.14
Mixed Forest	-0.34
Deciduous Needle-leaf Forest	0.56*
Bogs and marches	0.44
Palsa bogs	0.29
Recent burns	-0.25
Water bodies	0.63*
Aboveground biomass	-0.55*
Soil C stock, 0-30 cm	0.54*
Soil C stock, 0-100 cm	0.65*
Carbonate rocks	0.20
Continuous permafrost	0.66*
Discontinuous permafrost	-0.27
Sporadic permafrost	-0.43
Isolated permafrost	-0.19
Mean annual air temperature	-0.76*
Mean annual precipitation, mm	0.10

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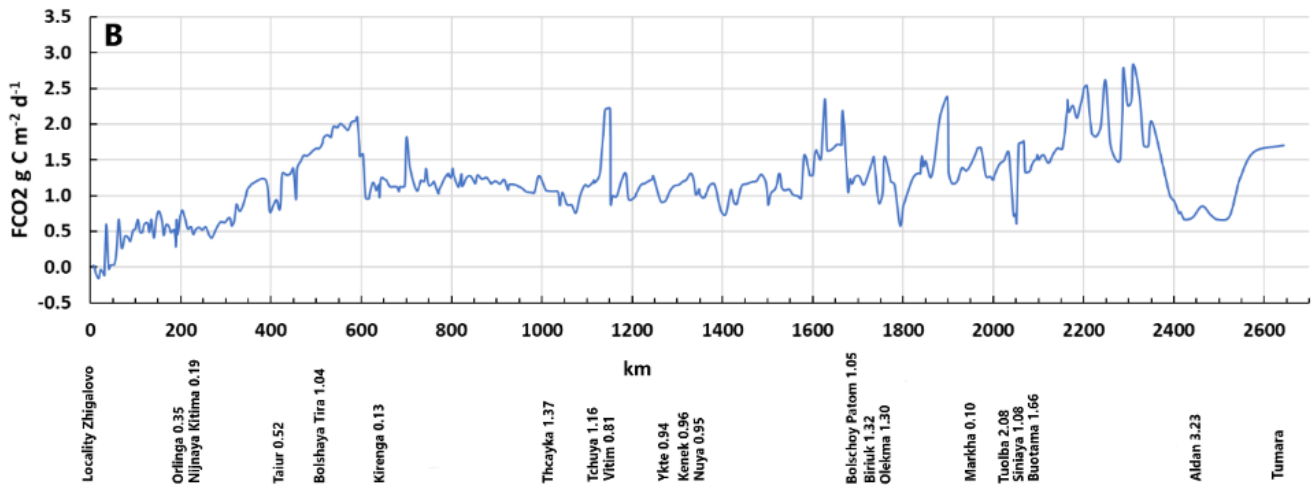
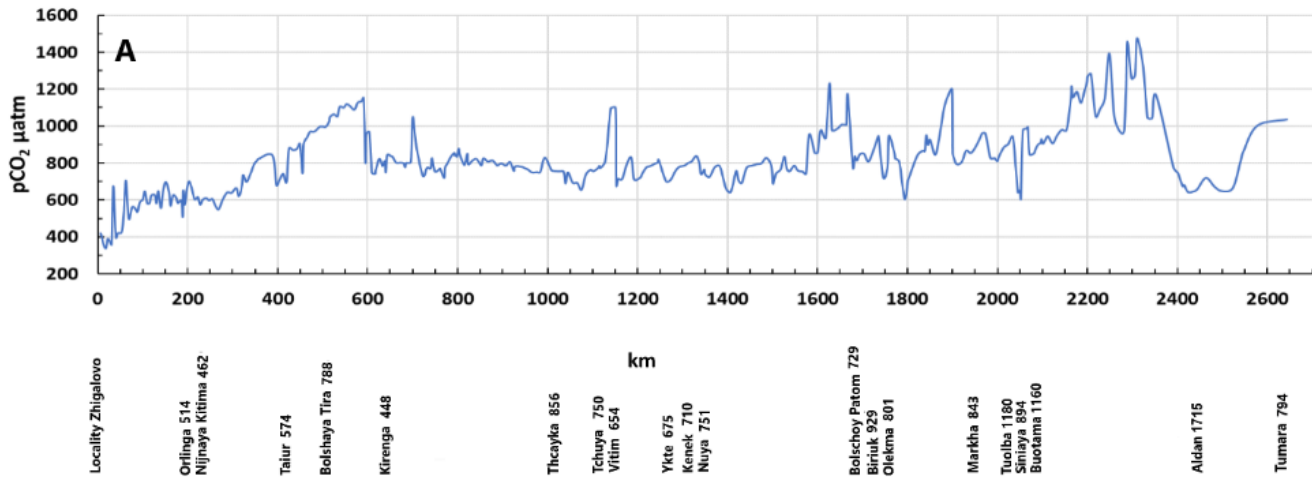


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947 **Fig. 1.** Map of the studied Lena River watershed with continuous pCO₂ measurements in the main
 948 stem. Mean multi-annual hydrographs of Ust-Kut, Krestovskoe and Tabaga station (labelled in red on
 949 the map) are provided in the insert.

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959 **Figure 2.** A 20-km averaged pCO₂ profile (A) and calculated CO₂ fluxes (B) of the Lena River main
 960 stem of over 2600 km distance, from Zhigalovo to the Tumara River. The average pCO₂ (µatm) and
 961 fluxes (g C m⁻² d⁻¹) of the main sampled tributaries are provided as numbers below X axes. Note that
 962 peaks of CO₂ concentration at the main stem are not linked to conflux with tributaries.

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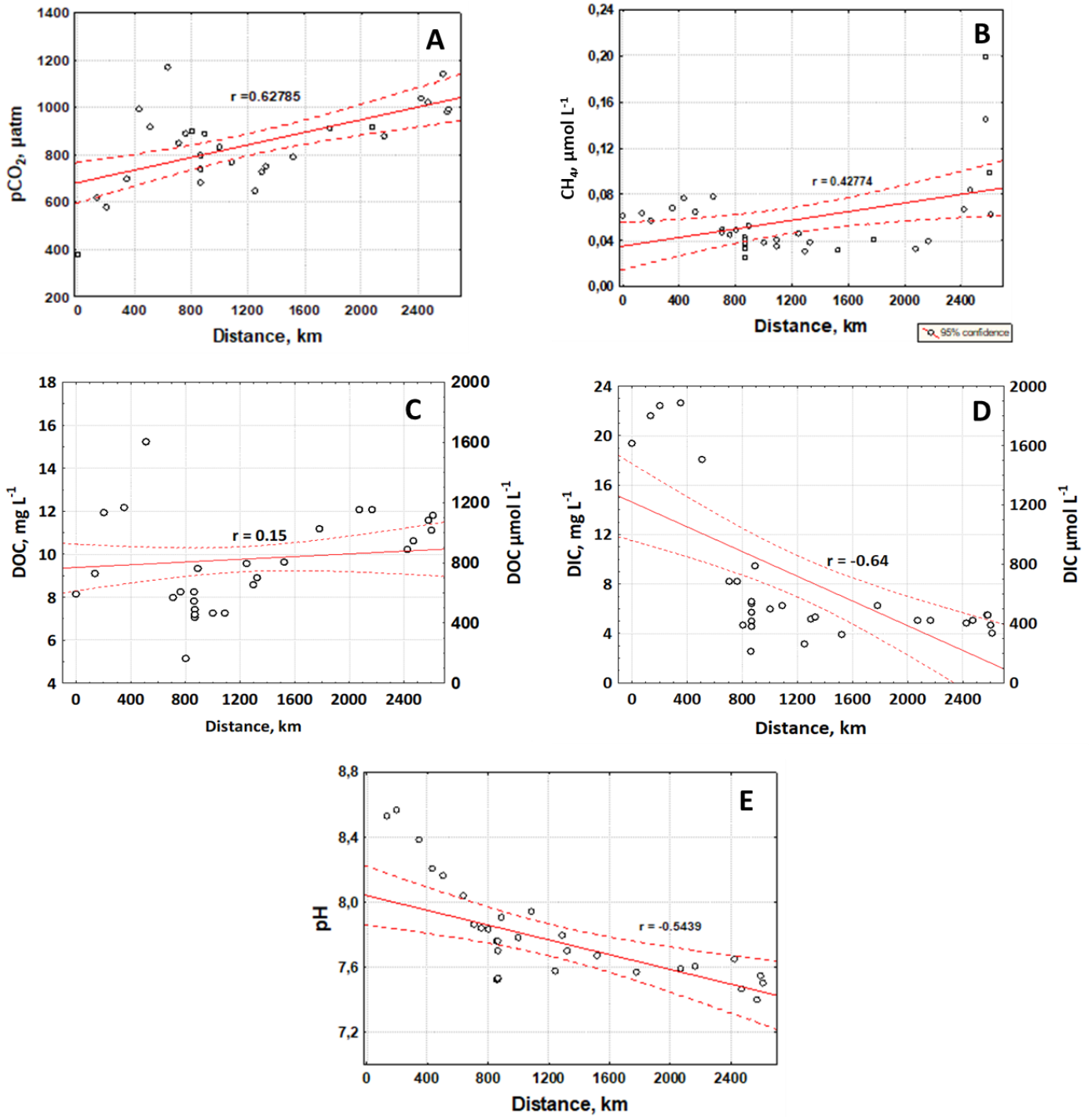
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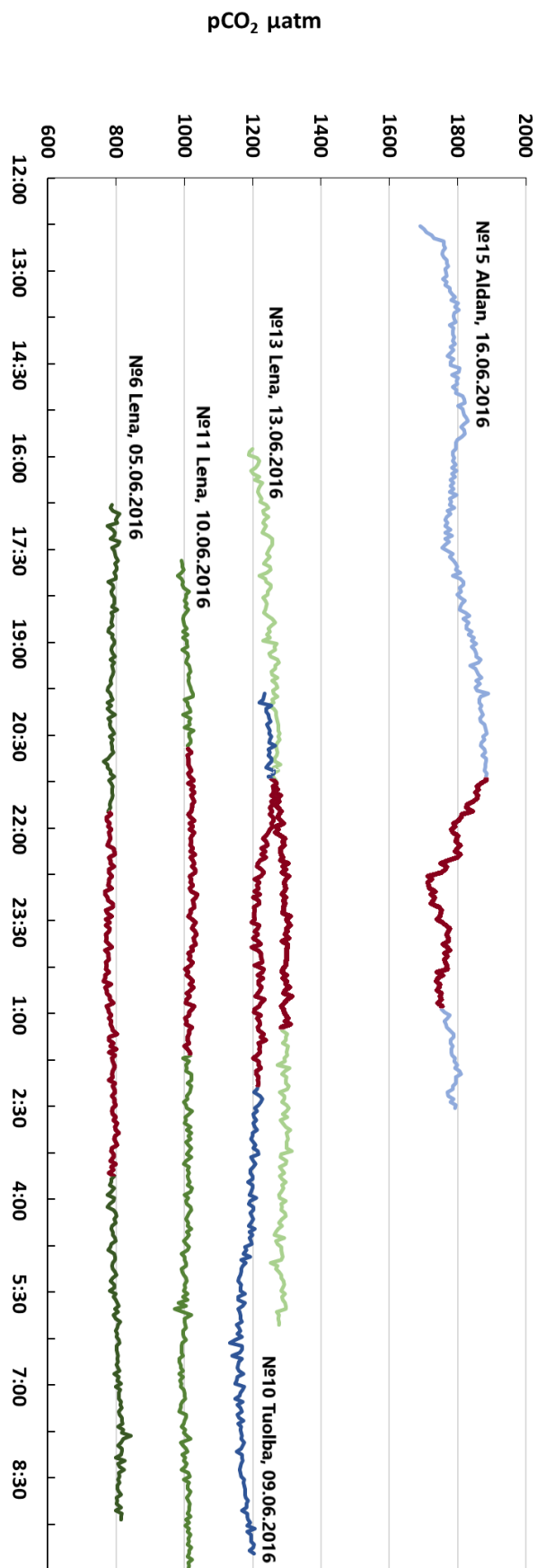
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973 **Figure 3.** Averaged (over 20-km distance) CO₂ (A), CH₄ (B), DOC (C), DIC (D) and pH (E)

974 concentration over the distance of the boat route at the Lena River, from the south-west to north-east.

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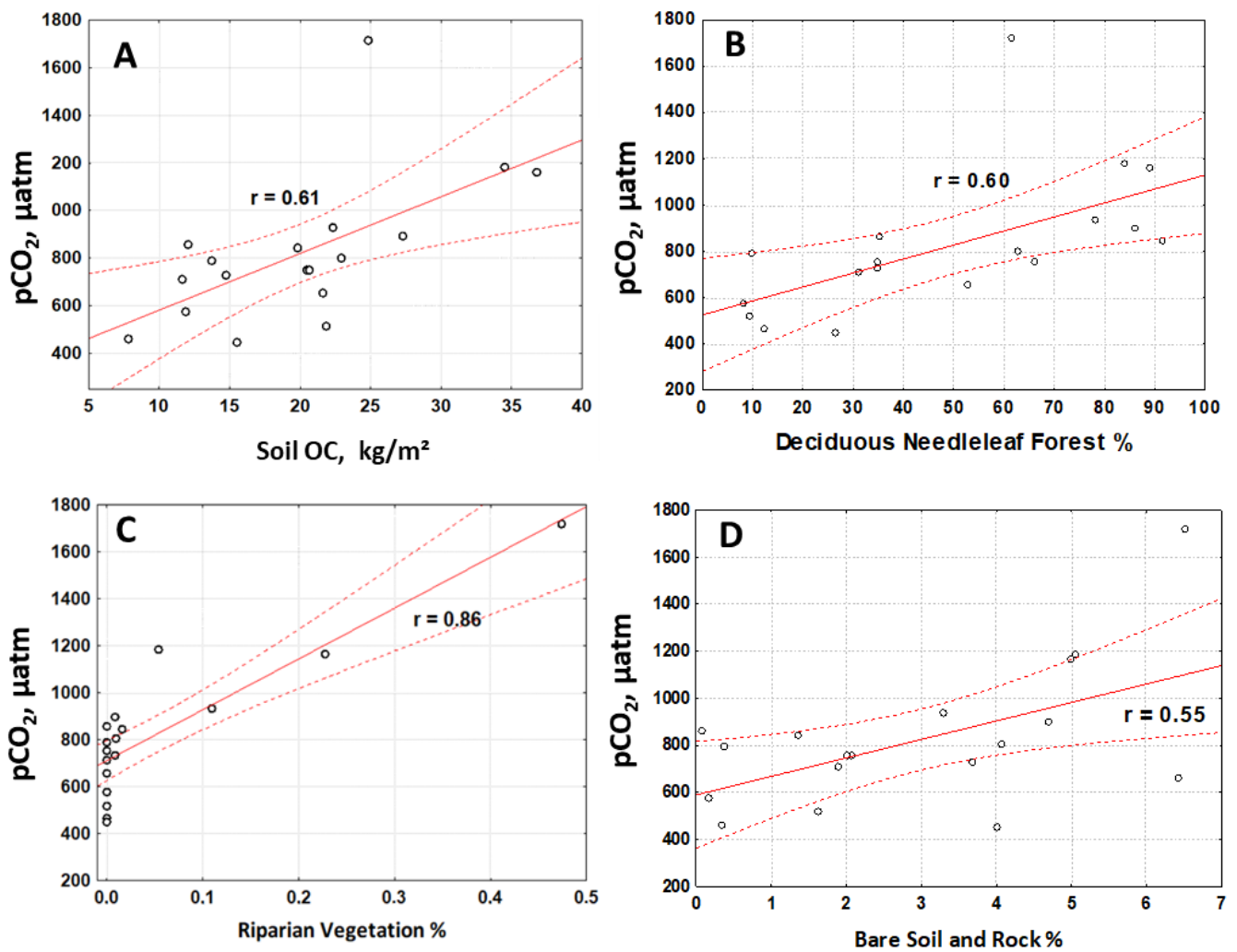
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978 **Figure 4.** Continuous pCO₂ concentration in the Lena River and two tributaries from late afternoon to
 979 morning next day. Red part of the line represents night time. Variations of water temperature did not
 980 exceed 2 °C.

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994 **Figure 5.** Significant ($p < 0.05$) positive control of landscape parameters – OC stock in 0-100 cm of
995 soil (A), and proportion of deciduous needle-leaf forest (B), riparian vegetation (C) and bare soil and
996 rock (D) in the watershed on pCO₂ in the Lena River tributaries.