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

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Abstract

Greenhouse gas (GHG) emissions from inland waters of permafrost-affected regions is one of the key factor 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 pCO_2 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 pCO_2 in Lena (range 400-1400 µatm) and tributaries (range 400-1600 µatm) was oversaturated and remained generally stable (within ca. 20 %) over the night/day period and

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across the river channels. The pCO_2 in tributaries increased northward with mean annual temperature decrease and permafrost increase; this change was positively correlated with C stock in soil and the proportion of deciduous needle-leaf forest and riparian vegetation. Based on gas transfer coefficients obtained from rivers of the Siberian permafrost zone, we calculated CO_2 emission for the main stem and tributaries. Typical fluxes ranged from 1 to 2 g C m^{-2} d^{-1} (>99% CO_2, < 1 % CH_4) which is comparable with CO_2 emission measured in Kolyma, Yukon and Mackenzie and permafrost-affected rivers in western Siberia. The areal C emissions from lotic waters of the Lena watershed were quantified via taking into account the total area of permanent and seasonal water of the Lena basin (28,000 km²). Assuming 6 months of the year of open water period and no emission under ice, the annual C emissions from the whole Lena basin range from 5 to 10 Tg C yr^{-1}, which is comparable to the DOC and DIC lateral export to the Arctic Ocean.

Introduction

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

Araujo et al., 2015; Kutscher et al., 2017; Griffin et al., 2018) and general hydrochemistry (Gordeev and Sidorov, 1993; Cauwet and Sidorov, 1996; Huh et al., 1998a,b; Huh and Edmond, 1999; Wu and Huh, 2007; Kuzmin et al., 2009; Pipko et al., 2010; Georgiadi et al., 2019; Juhls et al., 2020) including novel isotopic approaches for nutrients (Si, Sun et al., 2018) and trace metals such as Li (Murphy et al., 2019) and Fe (Hirst et al., 2020). This interest is naturally linked to the Lena River location within the forested continuous permafrost/taiga zone covered by organic-rich yedoma soil. Under on-going climate warming, the soils of the Lena River watershed are subjected to strong thawing and active (seasonally unfrozen) layer deepening (Zhang et al., 2005) accompanied by overall increase in river water discharge (McClelland et al., 2004; Ahmed et al., 2020), flood intensity and frequency (Gautier et al., 2018). The Lena River exhibits the highest DOC concentration among all great Arctic rivers (i.e., Holmes et al., 2013) which may reflect weak DOC degradation in the water column and mobilization of both contemporary and ancient OC to the river from the watershed (Feng et al., 2013; Wild et al., 2019). In contrast to rather limited works on CO_2 and CH_4 emissions from water surfaces of Eastern Siberia (Semiletov, 1999; Denfeld et al., 2013), extensive studies were performed on land, in the polygonal tundra of the Lena River Delta (Wille et al., 2008; Bussman, 2013; Sachs et al., 2008; Kutzbach et al., 2007) and the Indigirka Lowland (van der Molen et al., 2007). Finally, there have been several detailed studies of sediment and particular matter transport by the Lena River to the Laptev Sea (Rachold et al., 1996; Dudarev et al., 2006) together with detailed research of the Lena River Delta (Zubrzycki et al., 2013; Siewert et al., 2016).

Surprisingly, despite such an extensive research on C transport, storage, and emission in Eastern Siberian landscapes, C emissions of the Lena River main steam and tributaries remain virtually unknown, compared to relatively good knowledge of that in Yukon (Striegl et al., 2012; Stackpoole et al., 2017), Mackenzie (Horan et al., 2019), Ob (Karlsson et al., 2021; Pipko et al., 2019) and Kolyma (Denfeld et al., 2013). The only available estimates of C emission from inland waters of the Lena basin are based on few indirect (calculated gas concentration and modelled fluxes) snapshot data with very low spatial and temporal resolution (Raymond et al., 2013). Similar to other regions, this introduce uncertainties and cannot adequately capture total regional C emissions (Abril et al., 2015; Denfeld et al., 2018; Klaus et

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al., 2019; Klaus and Vachon, 2020; Karlsson et al., 2021). In particular, no detailed studies at the peak of spring flood have been performed and the understanding of various contrasting tributaries of the Lena River remain quite poor. As a result, reliable estimations of magnitude and controlling factors of C emission are poorly understood for the Lena River and its tributaries. The present work represents a first assessment of CO_2 and CH_4 concentration and fluxes of the main stem and tributaries during the peak of spring flow, calculating C emissions, and relating these data to river hydrochemistry and GIS-based landscape parameters. This should allow identifying environmental factors controlling GHG concentrations and emission in the Lena River watershed in order to use this knowledge for foreseeing future changes in C balance of the largest permafrost-affected Arctic river.

2. Study Site, Materials and Methods

2.1. Lena river and its tributaries

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

The peak of annual discharge depends on the latitude (**Fig. 1**) and occurs in May in the south (Ust-Kut) and June in the middle and low reaches of the Lena River (Yakutsk, Kysyr). From May 29 to June 17, 2016, we moved downstream the Lena River by boat with an average speed of 30 km h^{-1} (Gureyev, 2016). As such, we followed the progression of spring and moved from the south-west to the north-east, thus collecting the river water at approximately the same stage of maximal discharge. Note that transect sampling is a common way to assess river water chemistry in extreme environments (Huh and Edmond, 1999; Spence and Telmer, 2005), and generally, a single sampling during high flow season provides the best agreement with time-series estimates (Qin et al., 2006). Regular stops each 80-100 km along the Lena River allowed sampling for major hydrochemical parameters and CH_4 at the main stem. We also moved 500-1500 m upstream of selected tributaries to record pCO_2 for at least 1 h and to sample for river hydrochemistry; see examples of spatial coverage in **Fig. S1 B**. From late afternoon/evening to the next morning, we stopped for sleep but continued to record pCO_2 in the Lena River main stem (15 sites, evenly distributed over the full 2600 km transect) and two tributaries (Aldan and Tuolba).

2.2. CO_2 and CH_4 concentrations

Surface water partial pressure of CO_2 (pCO_2) was measured continuously, *in-situ* by deploying from the boat a portable infrared gas analyzer (IRGA, GMT222 CARBOCAP® probe, Vaisala®; accuracy ± 1.5%) of two ranges (2 000 and 10 000 ppm). The probe was enclosed within a waterproof and gas-permeable membrane and placed into a tube which was submerged 0.5 m below the water surface. This system was mounted on a small boat in a perforated steel pipe ~0.5 m below water surface. The tube had two necessary opening of different diameter, which allowed free water flow with a constant rate during the moving of the boat. A Campbell logger was connected to the system allowing continuous recording of the pCO_2 (ppm), water temperature (°C) and pressure (mbar) every minute during 5 minutes at 15 minutes interval yielding 4,285 individual pCO_2, water temperature and pressure measurements in total. The pCO_2 values in the Lena River tributaries were measured over the first 500-2000 m distance upstream the tributary mouth, and comprised between 5 and 34 measurements in case of day-time visit and between 305 and 323 individual pCO_2 readings for each tributary in case of day and night time monitoring.

Sensor preparation was conducted in the lab following method described by Johnson et al. (2009). The measurement unit (MI70, Vaisala®; accuracy ± 0.2%) was connected to the sensor allowing instantaneous readings of $p\mathrm{CO}_2$. The sensors were calibrated in the lab against standard gas mixtures (0, 800, 3 000, 8 000 ppm; linear regression with $R^2 > 0.99$) before and after the field campaign. The sensors' drift was 0.03-0.06% per day and the overall error was 4-8% (relative standard deviation, RSD). Following calibration, post-measurement correction of the sensor output induced by changes in water temperature and barometric pressure was done by applying empirically derived coefficients following Johnson et al. (2009). These corrections never exceeded 5% of the measured values.

For CH_4 analyses, unfiltered water was sampled in 60-mL Serum bottles and closed without air bubbles using vinyl stoppers and aluminum caps and immediately poisoned by adding 0.2 mL of saturated $HgCl_2$ via a two-way needle system. In the laboratory, a headspace was created by displacing approx. 40% of water with N_2 (99.999%). Two 0.5-mL replicates of the equilibrated headspace were analyzed for their concentrations of CH_4, using a Bruker GC-456 gas chromatograph (GC) equipped with flame ionization and thermal conductivity detectors. After every 10 samples, a calibration of the detectors was performed using Air Liquid gas standards (i.e. 145 ppmv). Duplicate injection of the samples showed that results were reproducible within ±5%. The specific gas solubility for CH_4 (Yamamoto et al., 1976) were used in calculation of total CH_4 content in the vials and then recalculated to μmol L^{-1} of the initial waters.

2.3. Flux calculation

CO_2 flux (F_{CO_2}) was calculated using Equation 1:

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

where K_h is the Henry's constant corrected for temperature and pressure (mol L^{-1} atm^{-1}), k_{CO_2} is the gas exchange velocity at a given temperature, C_{water} is the water CO_2 concentration, and C_{air} is the CO_2

concentration in the ambient air. Temperature-specific solubility coefficients were used to calculate respective CO_2 concentrations in the water following Wanninkhof et al. (1992).

The k_{CO_2} was fixed at 4.6 m day^{-1} following numerous measurements by floating chambers conducted by our group on permafrost-affected rivers in western Siberia (Serikova et al., 2018; Karlsson et al., 2021). This value is consistent with the k_{CO_2} reported for the Kolyma River and its large tributaries ($k = 3.9 \pm 2.5$ m d^{-1}, Denfeld et al., 2013), tributaries and main stem of the Yukon river basin ($k_{600} = 4.9$ - 7.6 m d^{-1}, Striegl et al. 2012), large rivers in the Amazon and Mekong basins ($k_{600} = 3.5 \pm 2.1$ m d^{-1}, Alin et al., 2011) and with modelling results of k for large rivers across the world ($k = 3$ - 4 m d^{-1}, Raymond et al., 2013).

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

2.4. Landscape parameters and water surface area of the Lena basin

The physico-geographical characteristics of the 20 Lena tributaries sampled in this study and the two points of the Lena main stem (upstream and downstream r. Aldan, **Table 1**) were determined by applying available digital elevation model (DEM GMTED2010), soil, vegetation, lithological, and geocryological maps. The landscape parameters were typified using TerraNorte Database of Land Cover of Russia (Bartalev et al., 2020, http://terranorte.iki.rssi.ru). This included various type of forest (evergreen, deciduous, needleleaf/broadleaf), grassland, tundra, wetlands, water bodies and other area. The climate and permafrost parameters of watershed were obtained from CRU grids data (1950-2016) (Harris et al., 2014) and NCSCD data (doi:10.5879/ecds/00000001, Hugelius et al., 2013), respectively, whereas the biomass and soil organic C content were obtained from BIOMASAR2 (Santoro et al., 2010) and NCSCD databases. The lithological composition of bedrocks was taken from State Geological Map of Russia with a resolution of 1:1,000,000 (VSEGEI, http://www.geolkarta.ru/). To test the effect of carbonate rocks on dissolved C parameters, we distinguished acidic crystalline, terrigenous silicate rocks and dolostones and limestones of upper Proterozoic, Cambrian and Ordovician age. We quantified river water surface area using the global SDG database with 30 m² resolution (Pekel et al., 2016) including both seasonal and permanent water for the open water period of 2016 and for the multiannual average (reference period 2000-2004). We also used a more recent GRWL Mask Database which incorporates first order wetted streams (Allen and Pavelsky, 2018).

The Pearson rank order correlation coefficient (Rs) ($p < 0.05$) was used to determine the relationship between CO_2 concentrations and climatic and landscape parameters of the Lena River tributaries. Further statistical treatment of CO_2, DIC and DOC concentration drivers in river waters included a Principal Component Analysis which allowed to test the effect of various hydrochemical and climatic parameters on behavior of DOC and GHG.

3. Results

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

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

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The measured pCO_2 in the river water and published (Karlsson et al., 2021) gas transfer coefficient (4.46 m day^{-1}) allowed to calculate the CO_2 fluxes over the full length of the studied main stem (2600 km) and the sampled tributaries. The calculated CO_2 fluxes of the main stem and tributaries ranged from zero and slightly negative (uptake) values in the most southern part of the Lena River and 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 (**Tables 1, 2; Fig. 2 B**). The largest part of the Lena River main stem, 1429 km from Kirenga to Tuolba 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 studied in this work, from Tuolba to Aldan, the calculated fluxes increased to 1.7±0.08 g C m^{-2} d^{-1}.

The river water concentrations of dissolved CH_4 in the tributaries and the main channel (0.059±0.006; IQR range from 0.025 to 0.199 µmol L^{-1}, **Table 1, 2**) did not exhibit any trend with the distance or landscape parameters of the cathcments. These values are consistent with the range of CH_4 concentration in the low reaches of the Lena River main channel (0.03-0.085 µmol L^{-1}; Bussman, 2013) and 100-500 times lower than those of CO_2. Consequently, diffuse CH_4 emissions constituted less than 1 % of total C emissions and are not discussed in further details.

3.2. Diurnal (night/day) pCO_2 variations and spatial variations across the river transect

The diel continuous CO_2 measurements of 3 tributaries (Kirenga, Tuolba and Aldan) and 14 sites of the Lena River main channel showed generally modest variation with diurnal range within 10 % of average pCO_2 (**Fig. 4** and **Fig. S2**). The observed variations in pCO_2 between day and night were not linked to water temperature ($p > 0.05$), which did not vary more than 1-2 °C between the day and night period.

The spatial variations of hydrochemical parameters were only tested in the upper reaches of the Lena main stream and its largest tributary - the Aldan River (**Fig. S3**). In the Lena River, over a lateral distance of 550 m across the river bed, the pCO_2 and CH_4 concentrations were equal to 569±4.6 µatm and 0.0406±0.0074 µmol L^{-1}, respectively, whereas DIC and DOC varied < 15% (n = 5). In the Aldan River, over a 2700 m transect, the pCO_2 and CH_4 concentrations were equal to 1035±95 µatm and

0.078±0.00894 µmol L^{-1}, respectively, whereas DIC and DOC varied within < 20% (n = 4). Overall, these results supported our design of punctual (snap shot) sampling in the middle of the river.

3.3. Impact of catchment characteristics on pCO_2 in Lena River basin

The CO_2 concentration in the Lena River main stem and tributaries increased from south-west to north-east (**Table 1, 2; Fig. 2**), and this was reflected in a positive (R = 0.66) correlation between CO_2 concentration and continuous permafrost coverage and a negative (R = -0.76) correlation with MAAT (**Table 3)**. Among different landscape factors, C stock in upper 0-30 and 0-100 cm of soil, the proportion of riparian vegetation and bare rocks, the coverage by deciduous needle-leaf forest, and coverage of river watershed by water bodies (mostly lakes) exhibited significant ($p < 0.01$, $n = 19$) positive correlations ($0.54 \leq R \leq 0.86$) with average pCO_2 of the Lena River tributaries (**Fig. 5**). The other potentially important landscape factors of the river watershed (proportion of peatland and bogs, tundra coverage, total aboveground vegetation, type of permafrost, annual precipitation) did not significantly impact the CO_2 concentration in the Lena River tributaries (**Table 3**).

Further assessment of landscape factor control on C parameters of the river water was performed via a PCA. This analysis basically confirmed the results of linear regressions and revealed two factors capable explaining only 12.5 and 3.5% of variability (**Fig. S4**). The F1 strongly acted on the sample location at the Lena transect, the content of OC in soils, the watershed coverage by deciduous needle-leaf forest and shrubs, riparian vegetation, but also proportion of tundra, bare rock and soils, water bodies, peatland and bogs (> 0.90 loading). The pCO_2 was significantly linked to F1 (0.72 loading).

3.4. Areal emission from the Lena River basin

The areal emission of CO_2 from the lotic waters of the Lena River watershed were assessed based on total river water coverage of the Lena basin in 2016 (28,197 km², of which 5,022 km² is seasonal water, according to the Global SDG database). This value is consistent with the total river surface area from the GRWL Mask database (22,479 km²). Given that the measurements were performed at the peak of spring flood in 2016, we used the maximal water coverage of the Lena River basin.

Based on past calculated pCO_2 of the Lena River (400 - 1000 µatm, Semiletov, 1999; Semiletov et al., 2011; Pipko et al., 2010) both the seasonality and spatial differences downstream are relatively minor. Indeed, for the low reaches of the Lena River, from Yakutsk to the Lena Delta, Semiletov (1999) and Semiletov et al. (2011) reported, for August-September 1995, the average pCO_2 of 538±96 µatm (range 380-727 µatm). This value is very similar to the one obtained in July 2003 for the low reaches of Lena (559 µatm, Pipko et al., 2010). Over the full length of the Lena River, from Ust-Kut to the Lena mouth, Pipko et al (2010) reported an average pCO_2 of 450 ± 100 µatm in June-July 2003. At the same time, calculated pCO_2 from previous field campaigns are generally lower than the pCO_2 of the Lena River main stem directly measured in this study: 700-800 µatm for the Ust Kut – Nuya transect (1331 km); 845 – 1050 µatm for the Nuya – Aldan part (1050 km).

Thus, despite the absolute values of calculated pCO_2 involve uncertainties (our calculated: measured pCO_2 in Lena River main channel and tributaries equaled 0.67±0.15 (n = 47)), this suggest spatial and temporal stability of the pCO_2 in the Lena River waters and allow to extrapolate the measured pCO_2 in the Lena River from Yakutsk to Aldan to the low reaches of the river. As for the Lena tributaries, for the best of our knowledge there is no published information on pCO_2 concentration and emissions. Overall, the major uncertainty in estimation of the Lena River basin emission stems from lack of direct pCO_2 measurements in the northern part of main channel over ca. 1000 km downstream the Aldan River including the large tributary Vilyi. Indeed, we noted that the largest northern tributary, the Aldan River which provides 70% of spring time discharge of the Lena River (Pipko et al., 2010), exhibited sizably higher emissions compared to the Lena River main channel upstream of Aldan (3.2±0.5 and 1.69±0.08 g C m^{-2} d^{-1}, respectively).

For areal emission calculations, we used the range of CO_2 emissions from 1 to 2 g C m^{-2} d^{-1} which covers full variability of both large and small tributaries and the Lena River main channel (**Tables 1-2, Figure 2 B**). This estimation assumes lack of pCO_2 dependence on the size of the watershed in the Lena basin as confirmed by our data (**Fig. S5**). For the two months of maximal water flow (middle of May - middle of July), the C emission from the whole Lena basin ranges from 1.7 to 3.4 Tg C yr^{-1} which is 20 to 30% of the the DOC and DIC lateral export to the Arctic Ocean. Assuming six months of open water

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period and no emission during winter, this yields between 5 and 10 Tg C y^{-1} of annual emission for the whole Lena River basin (2,490, 000 km²) with a total lotic water area of 28,100 km².

4. DISCUSSION

4.1. Possible driving factors of CO_2 pattern in the Lena River basin

Lack of sizable variation in pCO_2 between the day and night period or across the river bed suggests quite low site-specific and diurnal variability. It may be indicative of negligible role of primary productivity in the water column, given low water temperatures, shallow photic layer of organic-rich and turbid waters and lack of periphyton activity during high flow of the spring flood. The pCO_2 increased by a factor of 2 to 4 along the permafrost/temperature gradient from the south-west to the north-east, for both the main channel and sampled tributaries. This may reflect progressive increase in the feeding of the river basin by mire waters, increase in the proportion of needle-lead deciduous trees, and an increase in the width of the riparian zone. Another strong correlation is observed between the stock of organic C in soils (both 0-30 and 0-100 cm depth) and the pCO_2 of Lena's tributaries. Organic-rich soils are widely distributed in the central and northern part of the basin. The most southern part of the Lena basin is dominated by carbonate rocks and crystalline silicates in generally mountainous terrain, where only thin mineral soils are developed. The northern (downstream of the Olekma River) part of the basin consist of soils developed on sedimentary silicate rocks as well as vast areas of easily eroded yedoma soils. It is likely that both OM mineralization in OC-rich permafrost soils and lateral export of CO_2 from these soils, together with particulate and dissolved OC export and mineralization in the water column, are the main sources of CO_2 in the river water. Although some studies demonstrated high lability of DOM in arctic waters (Cory et al., 2014; Ward et al., 2017; Cory and Kling, 2018), other suggested it is low and do not support major part of CO_2 supersaturation in water (Shirokova et al., 2019; Payandi-Rolland et al., 2020; Laurion et al., 2021). Note that we have not observed any significant relationship between the DOC and pCO_2 in the Lena River and tributaries (**Fig. S6 A**). Lack of such a correlation and absence of diurnal

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pCO_2 variations imply that in-stream processing of dissolved terrestrial organic C is not the main driver of CO_2 supersaturation in the river waters of the Lena River basin.

The role of underground water discharge in regulating pCO_2 pattern of the tributaries is expected to be most pronounced in the SW part of the basin (Lena headwaters), where carbonate rocks of the basement would provide sizable amount of CO_2 discharged in the river bed. However, there was no relationship ($p < 0.05$) between the proportion of carbonate rocks on the watershed and the pCO_2 in the tributaries (**Fig. S6 B**), whereas for the Lena River main stem, the lowest CO_2 concentrations were recorded in the upper reaches (first 0-800 km) where the carbonate rocks dominate. Altogether, this makes unlikely the impact of underground CO_2 from carbonate reservoirs on river water CO_2 concentrations. Therefore, other sources of riverine CO_2 may include POC processing in the water column (Attermeyer et al., 2018), river sediments (Humborg et al., 2010) and within the riparian zone (Leith et al., 2014, 2015) which require further investigation. Besides, although there was no sizable variation in pCO_2 between the day and night period or across the river bed, the flux could show different spatial and temporal patterns if *k* shows larger variability (cf., Beaulieu et al., 2012). This calls for a need of direct flux measurements in representative rivers and streams of the Lena River basin.

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

The estimated CO_2 emissions from the Lena River main channel over 2600 km distance (0.8 – 1.7 g C m^{-2} d^{-1}) are fairly well comparable with values directly measured in rivers and streams of continuous permafrost zone of western Siberia (0.98 g C m^{-2} d^{-1}, Serikova et al., 2018), the Kolyma River (0.35 g C 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 (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 River FCO_2 values are lower than typical emissions from running waters in the contiguous Unites 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} d^{-1}, Rocher-Ros et al., 2019), small streams of the northern Kolyma River (6 to 7 g C m^{-2} d^{-1}, Denfeld et al., 2013) as well as the Ob River in the permafrost-affected zone (3.8 to 5.4 g C m^{-2} d^{-1}, Karlsson et al., 2021). In contrast to the main steam, the range of FCO_2 in the tributaries is larger (0.2 to 3.2 g C m^{-2} d^{-1})

and presumably reflects strong variability of environmental conditions across a sizable landscape and climate transect.

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

Despite high uncertainty on our regional estimations (lack of directly measured gas transfer values and seasonal resolution), we believe that these estimations are conservative and can be considered as first order values pending further improvements. In order to justify extrapolation of our data to all seasons and entire area of the Lena basin, we analyzed spatial and temporal variations in pCO_2 in the Lena river main stem from available literature data. First, based on published data, the seasonal and spatial variabilities of pCO_2 across the majority of the Lena River main stem are not high during open water period, although the low reaches of the Lena River may exhibit higher emissions compared to middle and upper course (see **section 3.4**). Second, although small mountainous headwater streams of the tributaries may exhibit high k due to turbulence, this could be counteracted by lower CO_2 supply due to low OC in mineral soil, lack of riparian zone and scarce vegetation. Moreover, although these small streams (watershed area < 100 km²) may represent > 60% of total watershed surfaces of the Lena basin (Ermolaev et al., 2018), their contribution to the total water surface is < 20% (19% from combined analysis of DEM GMTED2010 and 16% from the GRWL or Global SDG database as estimated in this study). Therefore, given that within the stream-river continuum, the CO_2 efflux increases only two-fold with a discharge decrease by a factor of 10,000, from 100 to 0.01 $m^3\ s^{-1}$ (Hotchkiss et al., 2015), and that the watershed area had no impact on pCO_2 concentration in the river water (**Fig. S5**), this uncertainty is likely less important. As such, instead of integrating indirect literature data, we used the pCO_2 values measured in the present study to calculate the overall CO_2 emission from all lotic waters of the Lena basin.

Overall, the follow up studies of this large heterogenous and important system should include CO_2 measurements in 1) the low reaches of the Lena River, downstream of Aldan, notably large organic-rich tributaries such as Vilyi (454,000 km²) and where the huge floodzone (20-30 km wide) with large number of lakes and wetlands is developed, and 2) highly turbulent eastern tributaries of the Lena River downstream of Aldan, which drain the Verkhoyansk Ridge and are likely to exhibit elevated gas transfer coefficients.

The C evasion from the Lena basin assessed in the present work is comparable to the total (DOC+DIC) lateral export by the Lena River to the Arctic Ocean (10 Tg C yr^{-1}, Semiletov et al., (2011), or 11 Tg C yr^{-1} (5.35 Tg DIC yr^{-1} + 5.71 Tg DOC yr^{-1} by Cooper et al., 2008). Moreover, the C evasion strongly exceeds sedimentary C input to the Laptev Sea by all Siberian rivers (1.35 Tg C y^{-1}, Rachold et al. (1996) and Dudarev et al. (2006)), the Lena River annual discharge of particulate organic carbon (0.38 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).

Conclusions

Continuous pCO_2 measurements over 2600 km of the upper and middle part of the Lena River main channel and 20 tributaries during the peak of spring flood allowed to quantify for the first time, in-situ pCO_2 variations which ranged from 500 to 1700 µatm and exhibited a 2 to 4-fold increase in pCO_2 concentration northward. There was no major variation in pCO_2 between the day and night period or across the river bed which supports the chosen sampling strategy. The northward increase in pCO_2 was correlated with increased proportion of needle-lead deciduous trees, the width of the riparian zone and the stock of organic C in soils. Among the potential drivers of riverine CO_2, changes in the vegetation pattern (northward migration of larix treeline in Siberia; Kruse et al., 2019) and soil OC stock are likely to be most pronounced during ongoing climate warming and thus the established link deserves further investigation. The total C emission from the lotic waters of the Lena River basin ranges from 5 to 10 Tg C y^{-1} which is comparable to the annual lateral export (50% DOC, 50 % DIC) by the Lena River to the

Arctic Ocean. However, these preliminary estimations of C emission should be improved by direct flux measurements across seasons in different types of riverine system of the basin, notably in the low reaches of the Lena River.

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Authors contribution.

SV and OP designed the study and wrote the paper; SV, YK and OP performed sampling, analysis and their interpretation; MK performed landscape characterization of the Lena River basin and calculated water surface area; JK provided analyses of literature data, transfer coefficients for FCO_2 calculations and global estimations of areal emission vs export.

Competing interests.

The authors declare that they have no conflict of interest.

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Table 1. Measured water temperature, pCO_2, calculated CO_2 flux, CH_4, DOC, and DIC concentrations and pH in the Lena River main stem (average ± s.d.; (n) is number of measurements).

River transect	T_{water}, °C	pCO_2, µatm	FCO_2, g C m^{-2} d^{-1}	CH_4, µmol L^{-1}
Lena upstream of Kirenga (0-578 km)	12.65±0.22 (99)	714±22 (99)	0.849±0.061 (99)	0.068±0.003 (6)
Lena Kirenga – Vitim (579-1132 km)	9.17±0.15 (87)	806±8.8 (87)	1.19±0.024 (87)	0.040±0.002 (12)
Lena Vitim -Nuya (1132-1331 km)	8.10±0.115 (27)	797±22 (27)	1.22±0.072 (27)	0.038±0.003 (5)
Lena Nuya – Tuolba (1331-2008 km)	9.61±0.09 (95)	846±12 (95)	1.29±0.034 (95)	0.037±0.002 (6)
Lena Tuolba – Aldan (2008-2381 km)	10.6±0.21 (52)	1003±28 (52)	1.69±0.081 (5)	0.088±0.034 (5)

	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)

Table 2. Measured water temperature, pCO_2, calculated CO_2 flux, CH_4, DOC, DIC concentration and pH in the tributaries (average ± s.d.; (n) is number of measurements).

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)

Table 2, continued.

	CH_4, 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)

Footnote : in all tributaries except Aldan, there was only one measurement of CH_4, DOC, DIC and pH

Table 3. Pearson correlations between pCO_2 and landscape parameters of the Lena tributaries. Significant correlations ($p < 0.05$) are marked by asterisk. Methane concentration did not exhibit any significant correlation with all tested parameters.

% 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

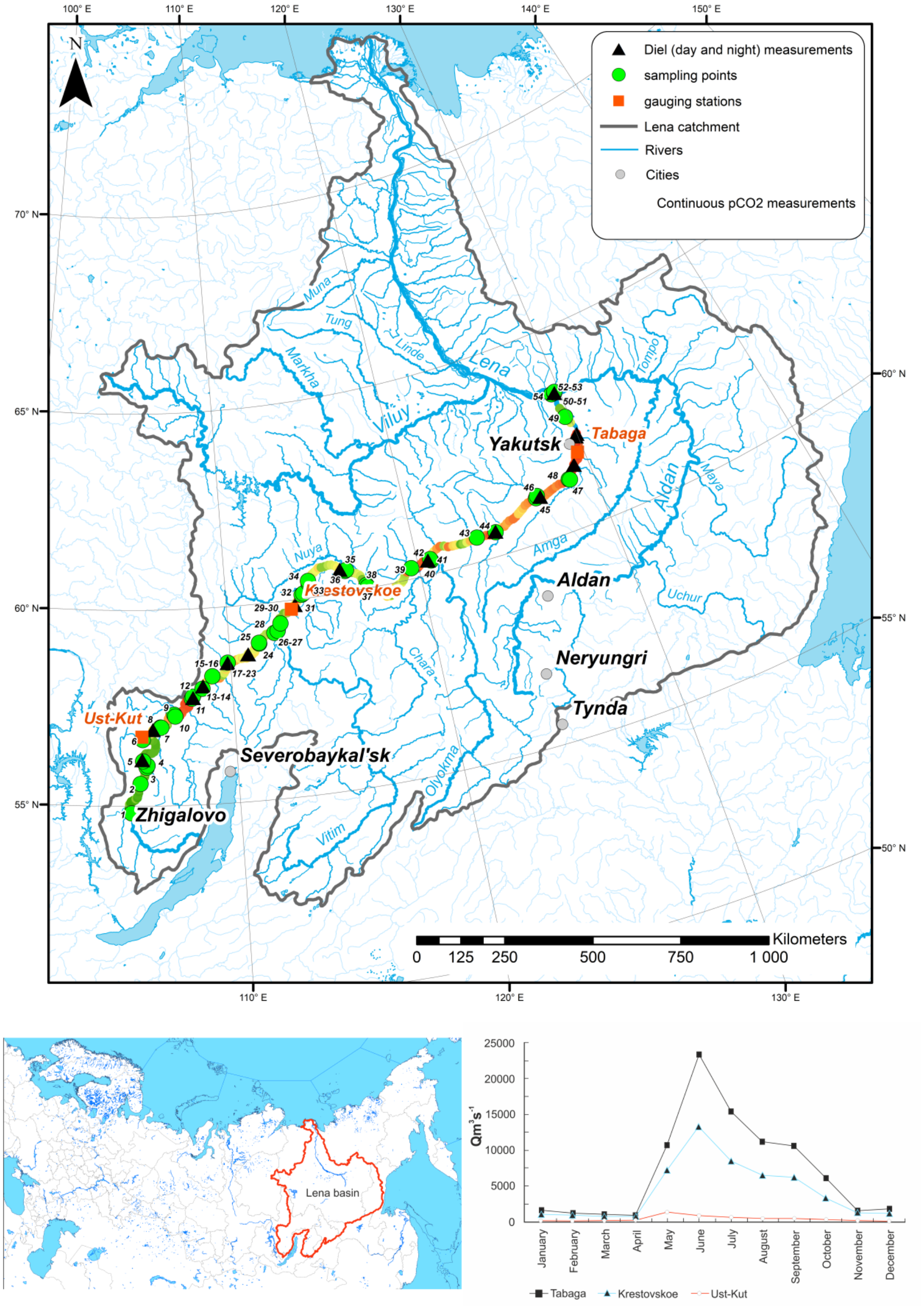

Fig. 1. Map of the studied Lena River watershed with continuous pCO_2 measurements in the main stem. Mean multi-annual hydrographs of Ust-Kut, Krestovskoe and Tabaga station (labelled in red on the map) are provided in the insert.

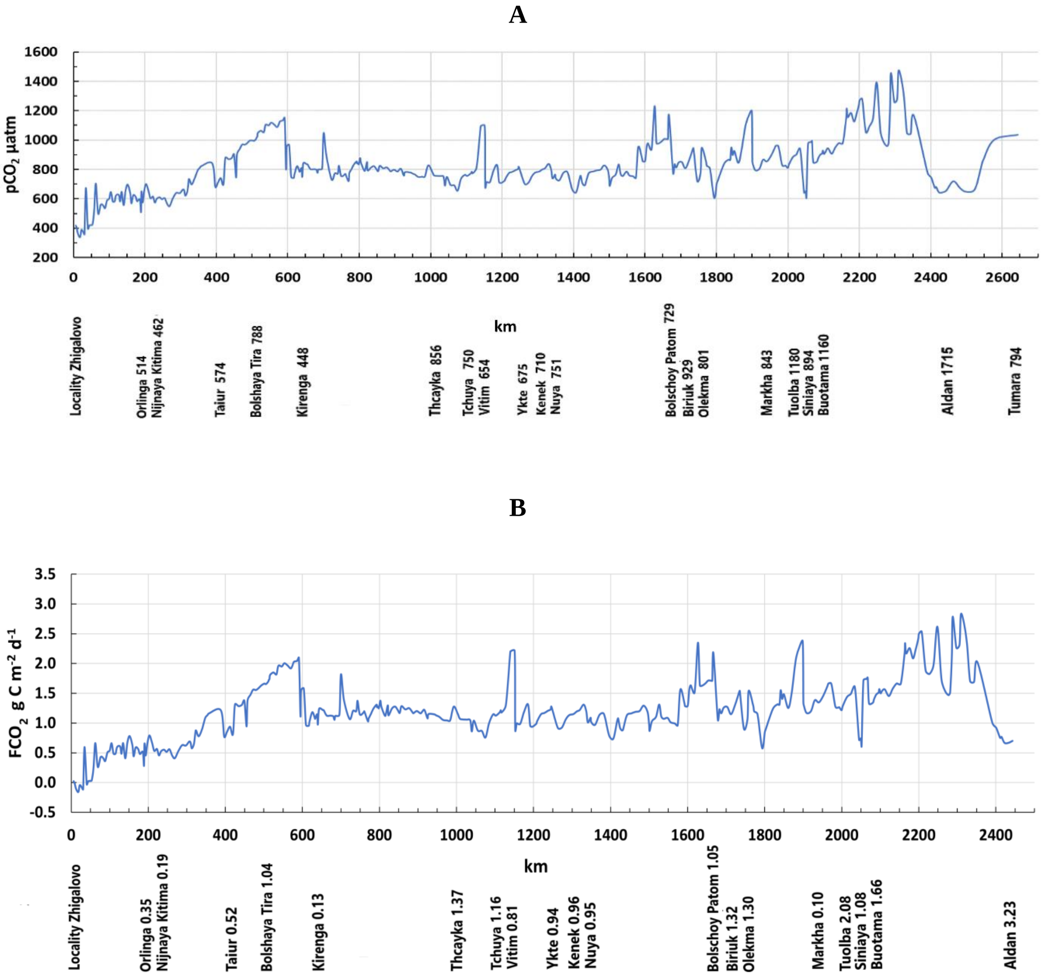

Figure 2. A 20-km averaged pCO$_2$ profile (A) and calculated CO$_2$ fluxes (B) of the Lena River main stem over 2600 km distance, from Zhigalovo to the Tumara River. The average pCO$_2$ (µatm) and fluxes (g C m^{-2} d^{-1}) of the main sampled tributaries are provided below X axes.

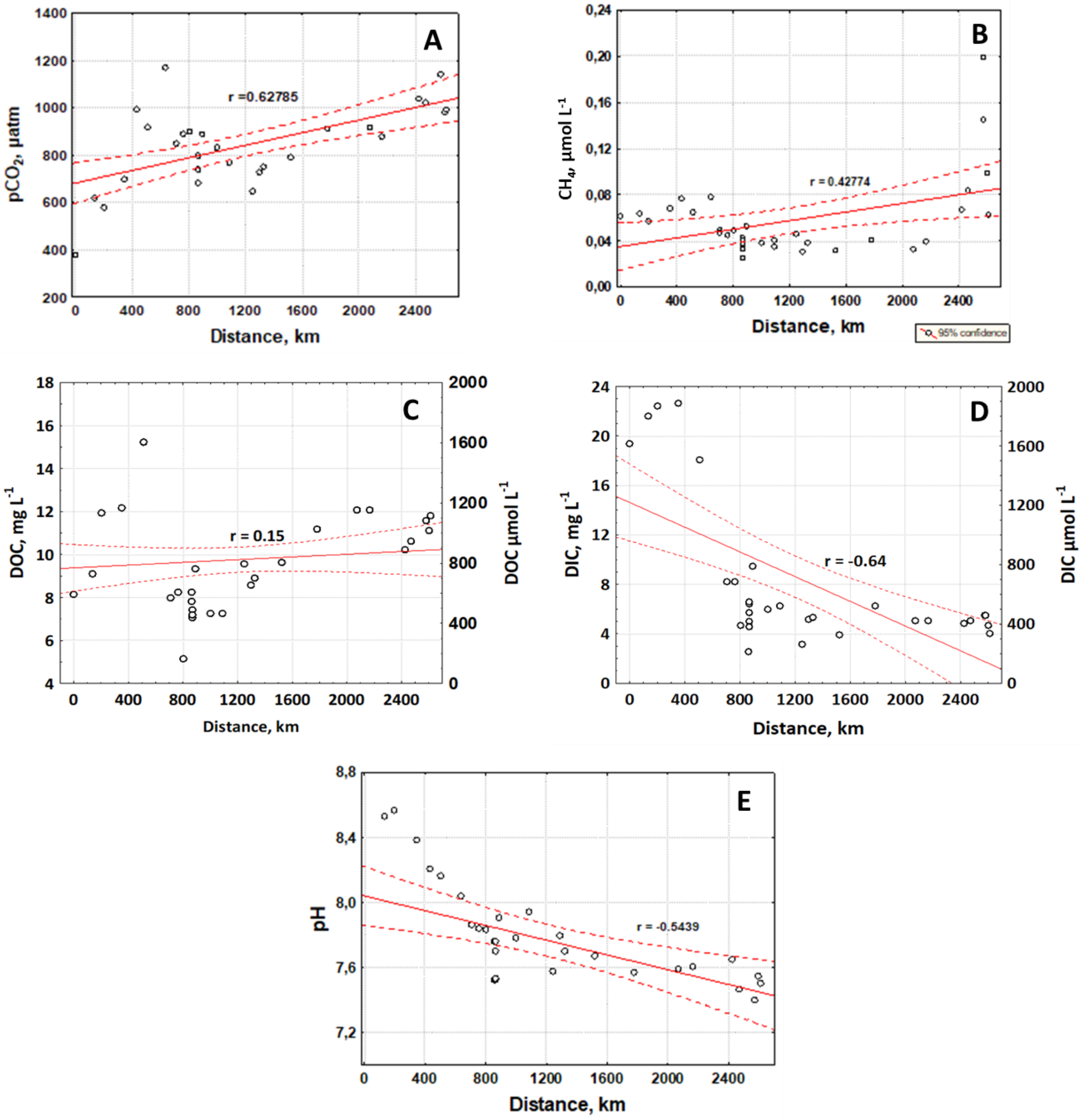

Figure 3. Averaged (over 20-km distance) CO_2 (A), CH_4 (B), DOC (C), DIC (D) and pH (E) concentration over the distance of the boat route at the Lena River, from the south-west to north-east.

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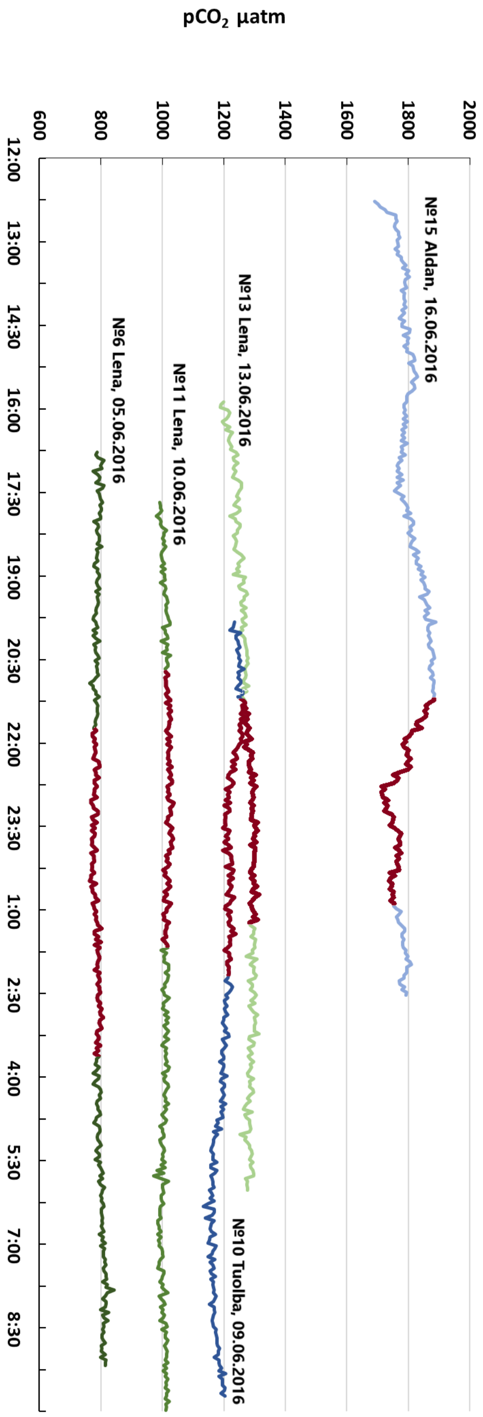

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843 **Figure 4.** Continuous pCO_2 concentration in the Lena River and two tributaries from the late afternoon 844 to the morning of the next day. Red part of the line represents the night time. The variations of water 845 temperature did not exceed 2 °C.

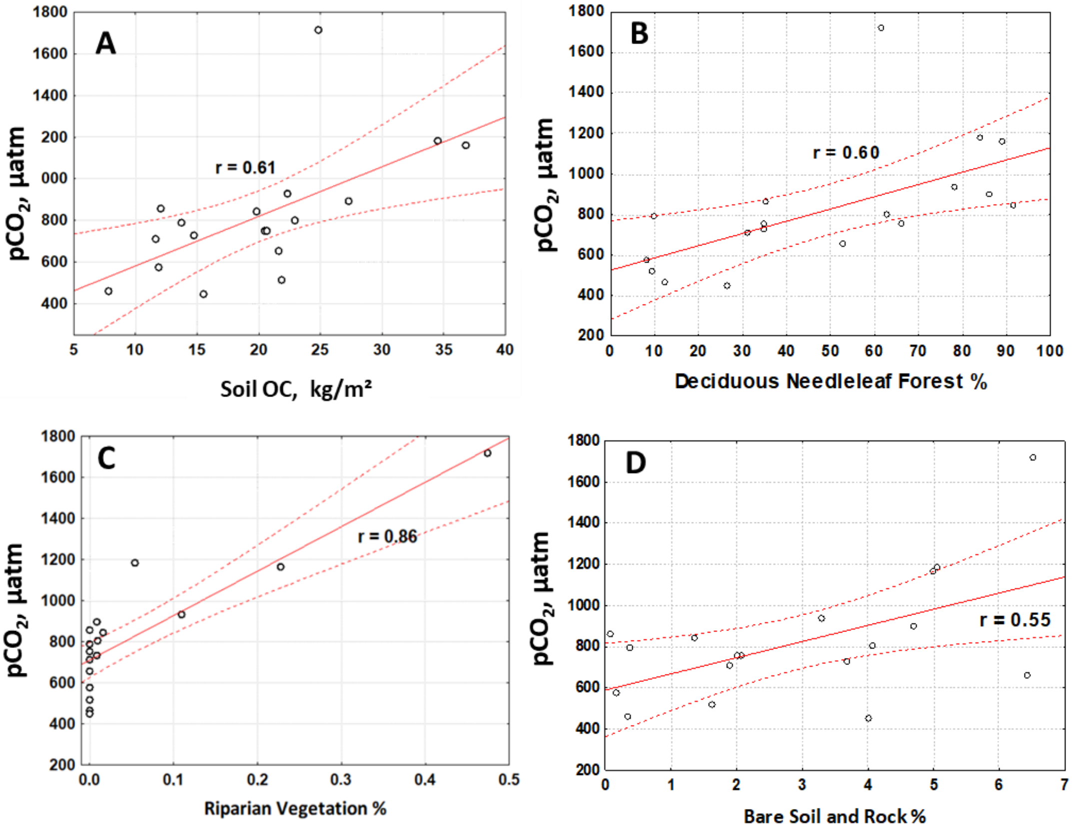

Figure 5. Significant ($p < 0.05$) positive control of landscape parameters – OC stock in 0-100 cm of soil (A), and proportion of deciduous needle-leaf forest (B), riparian vegetation (C) and bare soil and rock (D) in the watershed on pCO_2 in the Lena River tributaries.