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Article

Landscape, soil, lithology, climate and permafrost control on dissolved carbon, major and trace elements in the Ob River, western Siberia

Iurii Kolesnichenko[1], Larisa G. Kolesnichenko[1], Sergey N. Vorobyev[1], Liudmila S. Shirokova[2,3], Igor P. Semiletov[4,5,6,7], Oleg V. Dudarev[4], Rostislav S. Vorobev[1], Ulyana Shavrina[1], Sergey N. Kirpotin[1], and Oleg S. Pokrovsky[1,2,3*]

[1] BIO-GEO-CLIM Laboratory, Tomsk State University, Lenin pr., 36, Tomsk, Russia
[2] Geosciences and Environment Toulouse, UMR 5563 CNRS, 14 Avenue Edouard Belin 31400 Toulouse, France
[3] N. Laverov Federal Center for Integrated Arctic Research, Russian Academy of Sciences, Arkhangelsk, Russia
[4] Laboratory of Arctic Research, V.I. Il'ichev Pacific Oceanological Institute, Far Eastern Branch of Russian Academy of Sciences, 43 Baltic street, Vladivostok, 690041, Russia
[5] Tomsk Polytechnic University, Tomsk, Russia
[6] Tomsk State University, Tomsk, Russia
[7] Department of Chemistry, Moscow State University, Moscow, Russia
*Corresponding: oleg.pokrovsky@get.omp.eu

Abstract: In order to foresee possible changes in elementary composition of Arctic river waters, complex studies with extensive spatial coverage, including gradients in climate and landscape parameters, are needed. Here we used the unique position of the Ob River, draining through the vast partially frozen peatlands of western Siberia Lowland and encompassing a sizable gradient of climate, permafrost, vegetation, soils and Quaternary deposits, to assess a snap-shot (8-23 July 2016) concentration of all major and trace elements in the main stem (~ 3000 km transect from the Tom River confluence in the south to Salekhard in the north) and its 11 tributaries. During studied period, corresponding to the end of the spring flood-summer baseflow, there was a systematic decrease, from the south to the north, of Dissolved Inorganic Carbon (DIC), Specific Conductivity, Ca and some labile trace elements (Mo, W and U). In contrast, Dissolved Organic Carbon (DOC), Fe, P, divalent metals (Mn, Ni, Cu, Co, Pb) and low mobile trace elements (Y, Nb, REEs, Ti, Zr, Hf and Th) sizably increased their concentration northward. The Irtysh River, the main Ob's tributary, delivered enhanced amount of Cl, SO_4, Na, K but also DOC to the Ob River as the concentrations of these elements in the main stem increased 2 to 3-fold after the confluence with Irtysh.

The observed latitudinal pattern in element concentrations can be explained by progressive disconnection of groundwaters from the main river and its tributaries due to a northward increase in the permafrost coverage. A northward increase in bog versus forest coverage and an increase in DOC and Fe export enhanced the mobilization of insoluble, low mobile elements which were present in organo-ferric colloids (1 kDa - 0.45 µm) as confirmed by in-situ dialysis size fractionation procedure. The chemical composition of sampled mainstream and tributaries demonstrated significant ($p < 0.01$) control of latitude of the sampling point, permafrost coverage, proportion of bogs, lakes, floodplain coverage, lacustrine and fluvio-glacial Quaternary deposits of the watershed. This impact was mostly visible on DOC, Fe, P, divalent metals (Mn, Co, Ni, Cu, Pb), Rb and low mobile lithogenic trace elements (Al, Ti, Cr, Y, Zr, Nb, REEs, Hf, Th). The pH and concentrations of soluble, highly mobile elements (DIC, SO_4, Ca, Sr, Ba, Mo, Sb, W, U) positively correlated with the proportion of forest, loesses, eluvial, eolian, and fluvial Quaternary deposits on the watershed. Consistent with these correlations, a Principal Component Analysis demonstrated two main factors explaining the variability of major and trace element concentration in the Ob River main stem and tributaries. The DOC, Fe, divalent metals and trivalent and tetravalent trace elements were presumably controlled by a northward increase in permafrost, floodplain, bogs, lakes and lacustrine

deposits on the watersheds. The DIC and labile alkaline-earth metals, oxyanions (Mo, Sb, W) and U were impacted by southward-dominating forest coverage, loesses, eluvial and fertile soils.
Assuming that climate warming in the WSL will lead to a northward shift of the forest and permafrost boundaries, a "substituting space for time" approach predicts an increase in concentration of DIC and labile major and trace elements and a decrease of the transport of DOC and low soluble trace metals in the form of colloids in the main stem of the Ob River. However, an unknown factor is the change in hydrochemistry of the largest southern tributary, the Irtysh River, which is impacted by permafrost-free steppe and forest-steppe zone. Overall, seasonally-resolved transect studies of large riverine systems of western Siberia are needed to assess the hydrochemical response of this environmentally-important territory to on-going climate change.

Keywords: river, forest, bog, permafrost, carbon, major ions, iron, colloids, trace element

1. Introduction

Studies on hydrochemistry of large Arctic rivers are at the forefront of climate warming research due to their high importance in carbon (C) and greenhouse gases (GHG) regulation at the planetary scale and their high vulnerability to on-going environmental changes [1, 2]. Presently, researchers achieved rather good understanding of hydrological fluxes and river water hydrochemistry, including both suspended and particulate load, at the terminal (gauged) stations across the Arctic, thanks to systematic work of State Hydrological Surveys of main Artic countries [3], and more recently, concerted works of various international programs [4, 5, 6, 7, 8, 9]. In contrast, the knowledge of spatial variations of major and trace components of the river water along the main stem of Arctic rivers and their tributaries, which is necessary for understanding the environmental controls and export mechanisms, remains rather limited.

The Ob River, which is the largest Arctic river in terms of its watershed area (2,975,000 km^2), is an important vector of carbon, nutrients, major and trace element transfer to the Kara Sea [10, 11]. It drains highly vulnerable discontinuous and sporadic permafrost (20% in average), which is extremely rich in organic C (OC) due to the dominance of peat soils [12]. Most recent hydrological studies demonstrated that, over past 80 years, the Ob River discharge increased by ca 7.7% [13], at a rate of 384 and 173 m$^{(3)}$/s (10 year$^{(-1)}$)), in spring and winter respectively, which was linked to a rapid increase in both warming and wetting of the territory [14]. This, together with its unique geographical situation and landscape setting, render the Ob River watershed among the key targeting regions for biogeochemical studies in the Arctic [2]. Interestingly, that the number of publications per year on the Ob River increased from < 10 prior to 1995 to 10-20 in 1996-2014 and 50-80 over the past 6 years.

Several detailed studies of Dissolved Organic Carbon (DOC) and major elements were conducted at the terminal gauging station of the river, near the Salekhard city [4, 5, 6]. These included DOC time series observation by molecular-level techniques [15] and via remote sensing [16] and quantification of particulate organic matter export [7]. In contrast, spatial coverage of the river main stem and tributaries is rather low, with just a few studies of the dissolved carbon and related CO_2 and CH_4 emissions [17, 18] and one study of molecular composition of DOC [19]. Large amount of data are available from systematic State Rosgidromet monitoring of OC and major ions on four gauging stations of the Ob River (Salekhard, Belogor'e, Aleksandrovskoe and Kolpashevo) during 1970-2010 and measurements by Tomsk Politechnical University as summarized in ref. [20]. The OC-controlled export of Hg by the Ob River is considered by Mu et al. [21] and Sonke et al. [22]. A snapshot study of ^{137}Cs was performed at the scale of the Ob Basin [23]. Much less is known about other major and trace elements, especially their variations among the tributaries.

An important feature of the Ob River basin is the dominance of wetlands and mires which contain huge amount of OC and provide sizable input of DOM and relevant metals such as Fe to the Ob River main stem and tributaries due to strong hydrological connectivity (i.e., [24, 25, 26]. As a result of this enhanced input of Fe and DOC to the Ob River, its waters are likely to contain high

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concentration of colloids. However, this aspect has never been tested for the Ob River, although organic and organo-mineral colloids can be important carriers of a number of trace elements as it is known in small rivers [27] and surface waters [28, 29] across the western Siberia.

The novelty of the present work is to acquire a snap-shot picture of DOC, major and trace elements (TE), including their colloidal forms, in the main stem and several tributaries of the Ob River, and to test, for the first time for this territory, a landscape control, including vegetation, type of soil and Quaternary deposits, on chemical composition of the river water. The latter becomes now possible due to significant progress in digitalizing the available vegetation, permafrost and climate maps of large territories that allows straightforward landscape-based interpretation of river water chemistry (see examples in ref. [30, 31, 32]). In this study, we hypothesized a dominant control of bog and permafrost coverage on the concentrations of DOC, major and trace elements in the main stem. In particular, in accord with previous studies of small WSL rivers [10, 30, 31, 33, 34, 35, 36], we expected a northward increase in DOC and decrease in soluble alkaline-earth metals. In order to reveal the mechanisms of element transport in the Ob River and its tributaries, we assessed the colloidal and truly dissolved (low molecular weight) concentration of carbon, all major and trace elements in selected samples. We hypothesized drastic change in organic colloids after the confluence of the Ob and Irtysh rivers, following recent study of DOM pattern in the Ob River main stem [19]. We anticipate that acquiring this new knowledge on riverine major and trace elements over large climate, permafrost and vegetation gradient of the Ob River basin will allow empirical but straightforward prediction of future changes in river water chemistry, following a well-established substituting space for time approach.

2. Study site and methods

2.1. The Ob River of the WSL and its sampling

The Ob River, which delivers 15% of the total freshwater flow to the Arctic Ocean (404 km^3 y^{-1}), combines the features of southern rivers, draining steppe and forest-steppe regions, via its longest tributary, the Irtysh River, and the western Siberia Lowland rivers draining through a mixture of forest and mires. Further in the north, the Ob River is influenced by permafrost peatlands. The largest tributary of the Ob River is Irtysh which drains a territory of 1,643,000 km^2 and exhibit the annual discharge of 88.371 km^3 which is 37.5 % of that of the Ob River upstream their confluence at Khanty-Mansiisk. During the open water high flow period of 2016, the contribution of Irtysh amounted to 49% of the Ob River discharge upstream the confluence.

In this work, we used a ship ('OM-341' vessel, see ref. [19]) route sampling and collected 21 main stem water samples going from the north to the south between 8 July 2016 and 23 July 2016 (**Fig. 1**). We covered in total 2,952 km of the river length which encompassed a sizable gradient in the latitude (from 66.79 °N in the most northern site Salemal to 56.91 °N in the most southern site Kozjulyno, **Table S1**). In addition, 11 tributaries of the Ob River were collected. The summer of 2016 was rather cold (-3.1° below normal (defined as prior to 2000) for July) and slightly humid (130% of normal precipitation for July).

The river water was collected from the surface (0.5 m depth) in the central part of the main stem or minimum 500 m upstream the tributary via a pre-cleaned polypropylene 1-L container and immediately filtered (< 0.45 μm regenerated cellulose filter) using a pre-cleaned 250-mL polysulfone Nalgene filter unit combined with a vacuum pump. First, 250 mL of MilliQ water was filtered and the first portion of the river water filtrate (250 mL) was discarded. There was no decrease in the rate of filtration for sampled volumes (typically less than 500 mL) so we do not expect any artifacts linked to filter clogging.

In addition to traditional 0.45 μm filtration, 10 selected samples of the main stem and tributaries were processed for 1 kDa (~0.0013 nm) dialysis, allowing to quantify the nominal low molecular weight (LMW$_{<1\ kDa}$) and colloidal (1 kDa - 0.45 μm) fractions. For this, large volumes of the river water were collected into thoroughly cleaned and rinsed 5-L plastic container via filtration through 20 μm

Nylon net, to avoid large particles, zooplankton and insects. Dialysis experiments were performed using 50-ml pre-cleaned dialysis bags placed in the river water during 3 to 5 days as described elsewhere [28, 37]. The plastic container was kept in darkness at the temperature similar to that of the river water and gently agitated due to ship movement and manually. The stability of river water chemical composition during the full length of dialysis procedure was verified by comparison of dissolved (< 0.45 µm) concentrations of all solutes before and after the exposure; the difference did not exceed 10% (at $p < 0.01$). As such, even if some microbial activity could occur during dialysis, it did not modify the colloidal composition of the river water.

2.2. Analytical techniques

All filtered and dialysed samples were stored in the refrigerator 1 month prior the analyses. Major anion concentrations (Cl- and SO_4^{2-}) were measured by ion chromatography (Dionex 2000i) with an uncertainty of 2%. The dissolved organic carbon (DOC) and dissolved inorganic carbon (DIC) was determined using a Shimadzu TOC-VSCN Analyzer with an uncertainty of 3% and a detection limit of 0.3 mg/L [38]. In samples with low DOC (< 10 mg/L), the DIC was also analyzed via potentiometric alkalinity titration procedure with an uncertainty of ±1% and a detection limit of 5×10^{-5} mol L^{-1}; the difference with results of Shimadzu did not exceed 5%.

Major and trace elements were measured by quadrupole ICP-MS (7500ce, Agilent Technologies). Indium and rhenium were used as internal standards at their concentrations of approximately 3 µg L^{-1}. The international geostandard SLRS-5 (Riverine Water Reference Material for Trace Metals certified by the National Research Council of Canada) was used to check the validity and reproducibility of each analysis (see ref. 37 for analytical details). There was good agreement between our replicated measurements of SLRS-5 and the certified values (relative difference < 15 %). Elements lacking the certified values in the SLRS sample or those having high intrinsic uncertainty of measurements (> 20%) are not discussed in this work (Be, Sn, Te). For all major and most trace elements, analyzed by ICP MS, the concentrations in the blanks were below analytical detection limits (≤ 0.1-1 ng/L for Cd, Ba, Y, Zr, Nb, REE, Hf, Pb, Th, U; 1 ng/L for Ga, Ge, Rb, Sr, Sb; ~ 10 ng/L for Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As). Further details of analyses are provided elsewhere [27, 28, 31].

2.3. Landscape parameters of tributaries and the main stem

The landscape parameters of the 9 tributaries and 23 points of the Ob River main stem were determined by digitalizing available soil, vegetation, lithological, and geocryological maps (**Figure 1, Table S2**). The landscape and soil parameters were typified using United State Database of Soil Resources (http://egrpr.soil.msu.ru/); the borders were verified and corrected according to available Landsat images. The permafrost parameters of watershed were obtained from CRU grids data (1950-2016) and NCSCD data. The type of Quaternary deposits were taken from State Geological Map of Russia with a resolution of 1:1,000,000 (http://www.geolkarta.ru/).

The Pearson rank order correlation coefficient (Rs) ($p < 0.05$) was used to determine the relationship between each major and trace element concentrations and the latitude, climatic, lithological and landscape parameters of the watersheds. Further statistical treatment of element concentration drivers in river waters included a Principal Component Analysis with a variance estimation method and a scree test to minimize the number of governing factors. This analysis allowed to test the effect of various environmental parameters (landscape, soil, vegetation, permafrost and type of Quaternary deposits) of the watershed on spatial of riverine solutes in both the Ob River main stem and the tributaries.

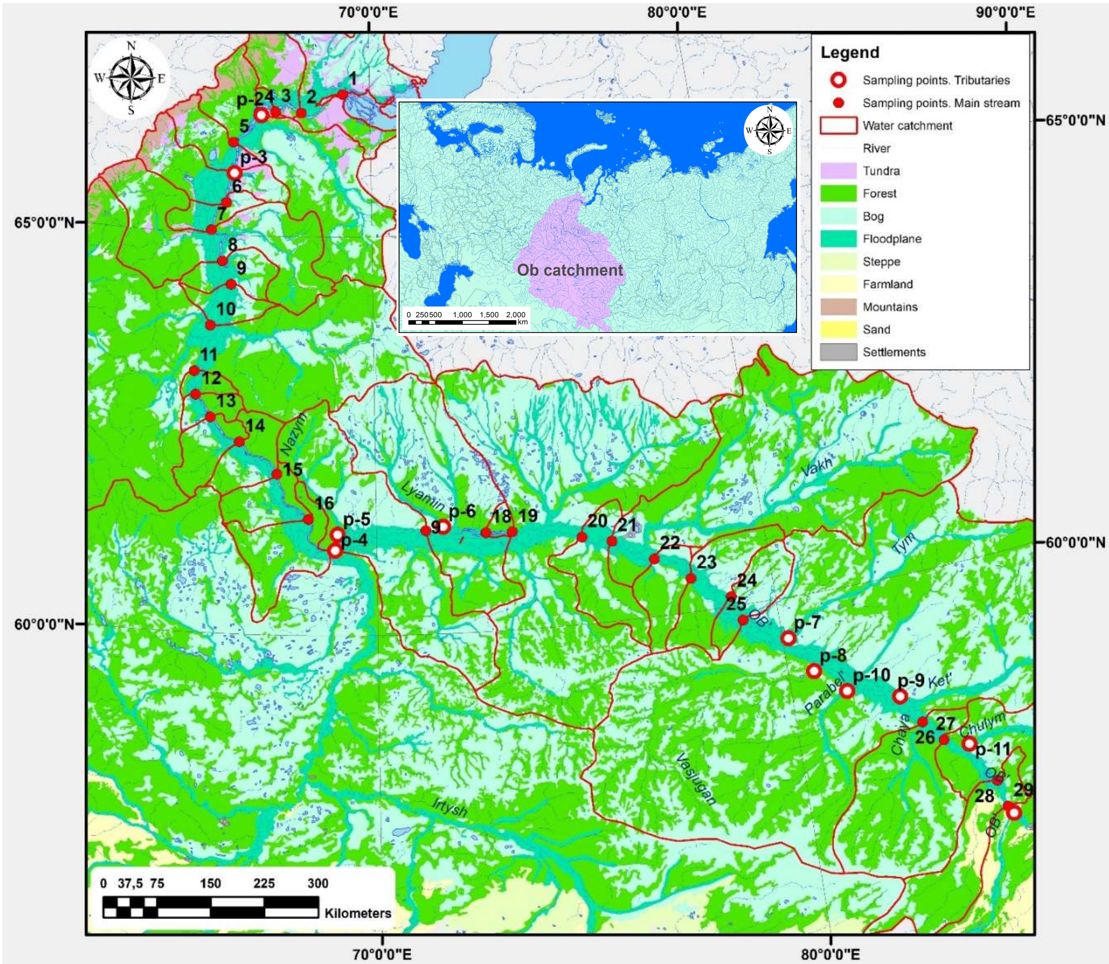

Fig 1. The Ob River and tributaries sampling points (solid and open circles, respectively) showing the dominant landscapes of the Ob River basin.

3. Results

3.1. Impact of the latitude on element concentration in the main stem of the Ob River

According to major and trace element behavior along the water course of the main stem [39], from the south to the north transect, three main families were distinguished as illustrated in **Figures 2 and 3**. Dissolved Inorganic Carbon, alkaline-earth metals (Ca, Sr), Mo, W, and U sizably (by a factor of 2.0) decreased their concentrations northward (**Fig. 2**).

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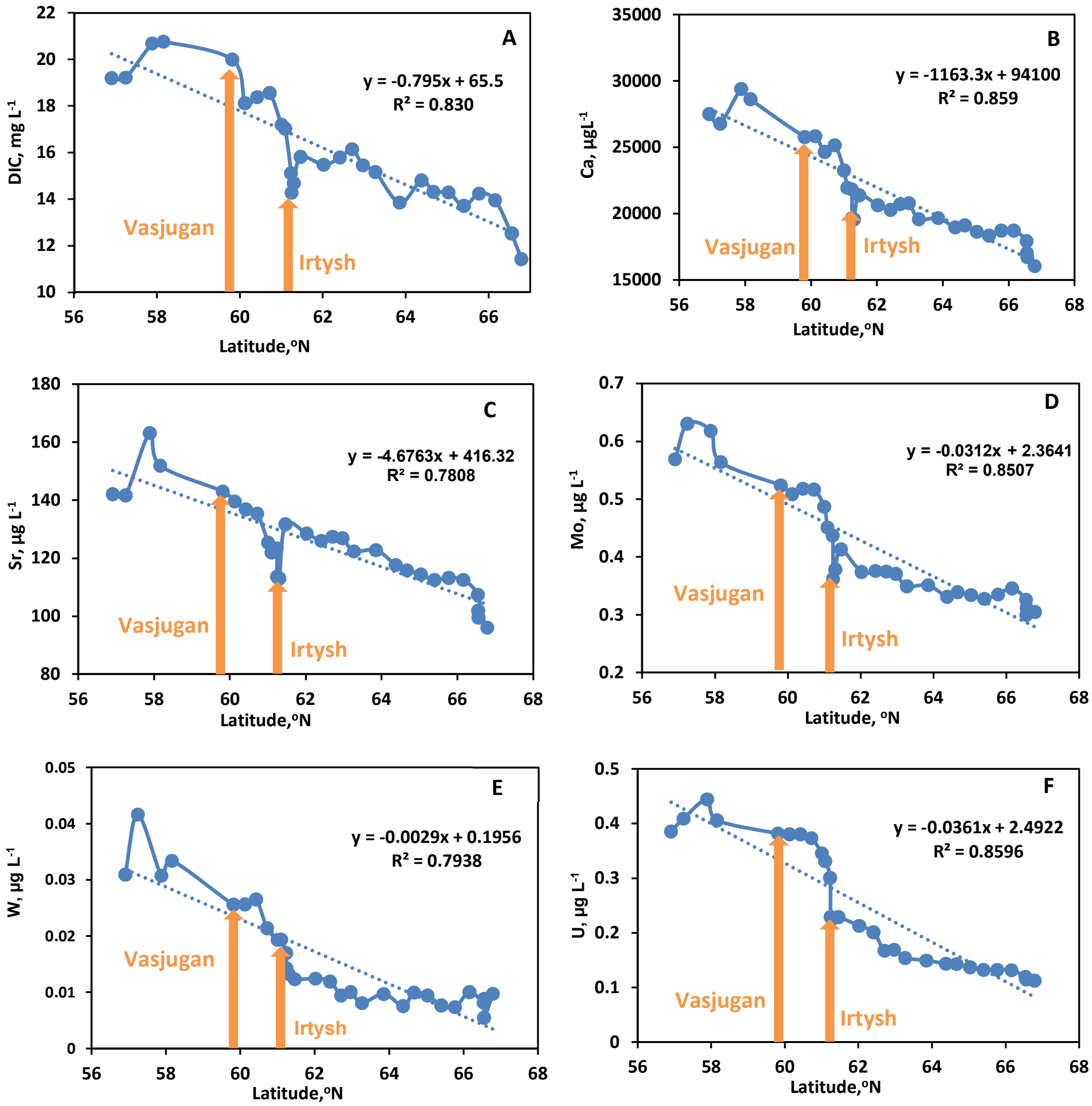

Figure 2. Latitudinal trends of labile major and trace elements decreasing their concentrations in the main stem of the Ob River from the south to the north: DIC (A), Ca (B), Sr (C), Mo(D), W (E) and U (F). The two main tributaries likely to control DOC and trace metal delivery, rivers Vasjugan and Irtysh, are shown by arrows.

The 2nd group of elements included DOC, P, Mn, Al, Fe, Ti, Ni, Cu, Co, Rb, Pb, Y, Zr, Nb, REE, Hf and Th which strongly (by a factor of 2 to 10) increased their concentration from the south to the north (**Fig. 3**). Sharp increase of these element concentration in the main stem of the Ob River occurred after the confluence first with Vasjugan, and then, Irtysh; the impact of Irtysh was especially seen for major anions (Cl, SO_4) and alkali (Na, K), and Pb (**Fig. S1** of the Supplement). Finally, a group of elements did not exhibit any sizable (> 1.5 x) change in concentration over the main stem (Si, Li, V, Ba) or the evolution of their concentration did not follow any specific pattern (B, Mg, Cr, Zn, Cd, Ga, Ge, Cs, Tl) as illustrated in **Fig. S2**. Note some peaks in concentrations detected for Zn, Cd and Pb which can be tentatively linked to local pollution sources.
Because the Vasyugan River, draining the largest world mire [40], can substantially modify the Ob River chemical composition, the degree of element enrichment in the northern part of the Ob River (downstream of Irtysh) was compared to its intermediate part (between Vasyugan and Irtysh) and the

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southern part (between Tom and Vasyugan) was assessed via comparison of average concentrations in these three segments of the main stem (**Table 1**). It can be seen that the elements mostly enriched (by a factor of 2 to 10) in the northern part of the river are DOC, Cl, P, Mn, Fe, some divalent metals (Mn, Ni, Pb) and insoluble lithogenic elements -Al, Ti, Nb, Y, REE, Ti, Zr, Hf, Th).

Table 1. Mean (±SD) concentration (µg L^{-1}) of major and trace elements in in three distinct parts of the Ob River main stem upstream and downstream its confluence with Vasyugan and Irtysh.

Descritions	Tom - Vasyugan (n = 4)	Vasyugan - Irtysh (n = 11)	Irtysh - Salemal (n = 16)
Cl	1740±510	1660±430	7240±530
SO_4	7070±1340	5106±888	8480±392
DOC	4600±930	7200±2100	11500±400
UV_{245}^*	0.13±0.45	0.28±0.12	0.49±0.02
DIC	18500±5300	16690±2180	15100±766
Li	2.63±0.92	1.91±0.17	2.74±0.19
B	13.9±2.5	10.0±0.63	17.7±1.64
Na	5866±1471	4533±296	8635±566
Mg	5086±803	4092±308	4788±236
Al	6.84±1.96	11.56±3.02	18.67±2.28
Si	2987±934	2905±797	2440±75
P	30.7±14.7	49.030±19.5	75.4±5.7
K	1115±183	972±137	1571±98
Ca	29664±3822	22677±2683	19978±914
Ti	3.27±1.20	5.46±1.84	8.80±1.04
V	1.67±0.3	1.37±0.08	1.52±0.03
Cr	0.14±0.11	0.27±0.12	0.30±0.12
Mn	2.0±1.48	18.0±49.0	4.5±0.5
Fe	59±62	440±475	664±58
Co	0.04±0.02	0.07±0.1	0.07±0.01
Ni	0.6±0.2	0.9±0.5	1.9±0.1
Cu	1.7±0.5	1.7±0.2	2.3±0.2
Zn	4.8±3.2	3.0±1.5	5.5±3.2
Ga	0.01±0.004	0.01±0.002	0.01±0.002
Ge	0.01±0.003	0.007±0.002	0.009±0.001
As	1.4±0.2	1.3±0.5	1.5±0.07
Rb	0.6±0.09	0.8±0.5	1.2±0.09
Sr	165±33	126±13	123±6
Y	0.04±0.03	0.1±0.05	0.3±0.01
Zr	0.02±0.02	0.06±0.03	0.1±0.01
Nb	0.002±0.002	0.007±0.005	0.02±0.002
Mo	0.7±0.08	0.4±0.1	0.4±0.03
Cd	0.01±0.01	0.009±0.009	0.01±0.006
Sb	0.1±0.02	0.1±0.02	0.09±0.004

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Descritions	Tom - Vasyugan (n = 4)	Vasyugan - Irtysh (n = 11)	Irtysh - Salemal (n = 16)
Cs	0.001±0.00	0.001±0.000	0.002±0.000
Ba	24±3	22±0.9	24±0.7
La	0.03±0.02	0.1±0.04	0.2±0.01
Ce	0.04±0.03	0.2±0.08	0.4±0.02
Pr	0.01±0.01	0.03±0.01	0.06±0.003
Nd	0.03±0.03	0.1±0.04	0.2±0.02
Sm	0.01±0.01	0.03±0.01	0.06±0.004
Eu	0.004±0.002	0.008±0.002	0.015±0.001
Gd	0.01±0.01	0.03±0.01	0.06±0.004
Tb	0.001±0.001	0.004±0.001	0.008±0.000
Dy	0.006±0.004	0.02±0.008	0.05±0.003
Ho	0.001±0.001	0.004±0.002	0.009±0.001
Er	0.004±0.003	0.01±0.004	0.03±0.002
Tm	0.000±0.000	0.002±0.001	0.004±0.000
Yb	0.003±0.002	0.01±0.004	0.02±0.002
Lu	0.000±0.000	0.002±0.001	0.003±0.000
Hf	0.002±0.001	0.008±0.004	0.02±0.002
W	0.03±0.01	0.02±0.007	0.01±0.002
Tl	0.002±0.001	0.001±0.000	0.002±0.000
Pb	0.07±0.1	0.2±0.1	0.2±0.02
Th	0.001±0.001	0.007±0.004	0.02±0.002
U	0.4±0.06	0.3±0.1	0.2±0.03

* units are cm^{-1}

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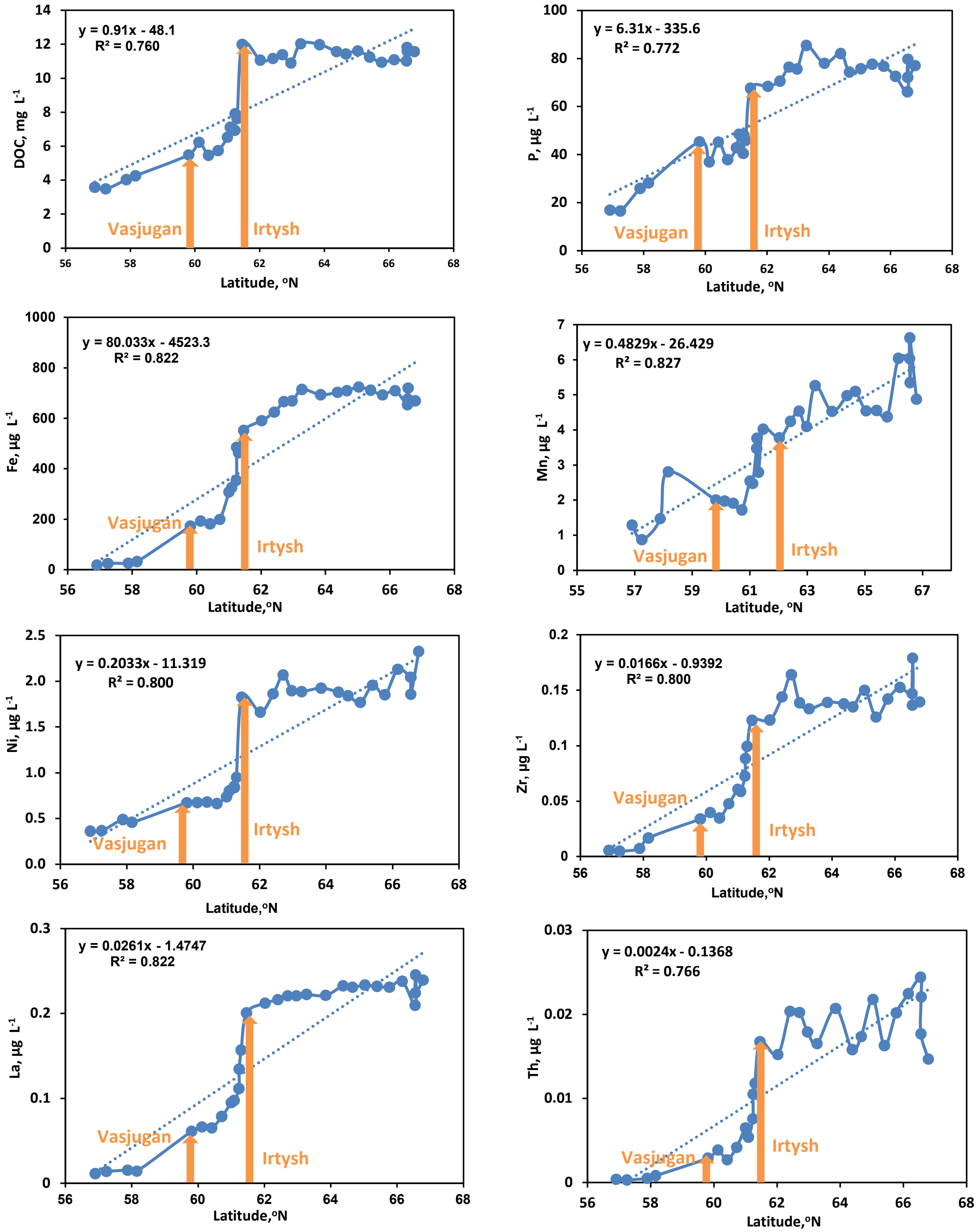

Figure 3. Latitudinal trends of DOC, nutrients (P, Fe, Mn, Ni) and insoluble trace elements (Zr, La, Th) showing strong increase in concentration in the Ob main stem after the confluence with the Vasyugan and Irtysh River (shown by arrows).

The distinction of riverine solutes into three major groups depending on their latitudinal pattern was also confirmed by a Pearson correlation coefficients between the element concentration and the latitude (**Table S3** of the Supplement). The elements of the first group exhibited quite strong (≤ -0.90, $p < 0.005$) negative correlations with latitude, whereas the elements of second group had $R_{Pearson} > 0.80$. The elements with weak or non-systematic pattern showed statistically significant correlations ($p < 0.05$) but the $|R_{Pearson}|$ was typically below 0.8.

3.2. Major and trace elements in the tributaries

The 11 tributaries sampled in this study exhibited highly contrasting behavior in both major and trace element concentrations. Similar to the main stem, this allowed revealing distinct group of elements according to their latitudinal pattern. The southern tributaries (upstream of Irtysh) were enriched in DIC, major anions, alkali and alkaline-earth metals and U, whereas the northern tributaries exhibited much higher concentrations of Mn, Fe, Co (> 5 times) and were sizably enriched in DOC, Cr, P, Zn, Cd, Nb, trivalent and tetravalent low soluble trace elements (**Fig. S3**).

The Irtysh River played a governing role in the concentration pattern of the main stem, as it presented sizable addition of many elements, mostly labile anions and oxyanions (DIC, Cl, SO_4, B, Mo, Sb, W), alkalis and alkaline-earth metals (Li, Na, K, Rb, Mg, Ca, Sr, Ba), some divalent transition metals (Mn, Co, Ni), U and DOC to the main stem, given its high discharge (49 % of the Ob River at the confluence in 2016) and most importantly, elevated concentration of elements (**Fig. 4**). Some elements however, exhibited higher concentration in the northern part of the Ob River compared to the Irtysh River: Al, Fe, Cr, Zn, Ga, Nb, Ti, Zr, Hf, REEs (**Fig. 4**). Presumably, these elements are additionally delivered to the Ob River main stem via multiple small-size tributaries, which were not sampled in the present work.

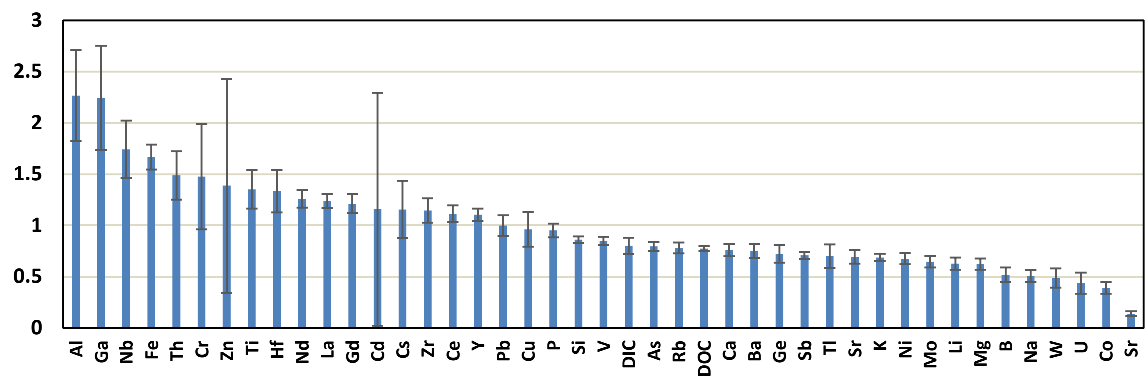

Figure 4. The ratio of average (± s.d.) element concentration of element in the Ob River main stem downstream the Irtysh (till the Ob mouth) to the concentration in the Irtysh River. Values above 1 indicate sizable enrichment of the main stem in a given element by small tributaries in the north.

3.3. Colloidal status of major and trace elements in the Ob River and tributaries

The dialysis procedure allowed a first-order assessment of the colloidal fraction (defined as the % of the element in the 1 kDa - 0.45 μm fraction divided by its total dissolved concentration in the < 0.45 μm fraction). The percentage of colloidal fraction ranged from 0-10% (Cl, SO_4, DIC, Na, K, Li, B, Si, Mg, Ca Mo) to 80-90% (trivalent and tetravalent hydrolysates) as listed in **Table S4**. In the northern part of the basin (downstream of Irtysh), the distribution of colloidal forms of elements among the tributaries and the main stem was quite homogeneous. This allowed identification three main groups of solutes with respect to their colloidal status (**Fig. 5)**: (1) alkalis and alkaline-earth metals, major anions and Si, trace oxyanions (Mo, Sb, Ge) presenting between 0 and 20 % of colloidal forms; (2) DOC, divalent transition metals (No, Cu, Cd), V, Cr, Cs, Tl, W and U which were sizably impacted

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by colloids (20-70 %), and (3) P, Fe, Mn, Co and all trivalent and tetravalent hydrolysates (Al, Ga, Y, REEs, Ti, Zr, Hf, Th) which were present essentially (> 70-80 %) in the colloidal form.

The DOC exhibited remarkably homogeneous proportion of colloidal forms (42±3 %) in the main stem and most tributaries including Irtysh. However, two most southern tributaries sampled for colloids (Ket' and Tom') exhibited much lower proportion of colloidal DOC as well as of Fe, Co, Ni. Other colloid-affected elements (Al, Ti, P, V, Cr, As, Ga, Y, Zr) also demonstrated the lowest proportion of colloids in southern tributaries (Irtysh, Ket and Tom). Uranium exhibited quite particular colloidal pattern, with gradual decreasing of colloidal fraction from the north (70- 90 %) to the middle course including Irtysh (20 to 50%) and further decreasing in two most southern tributaries Ket' and Tom' (3%).

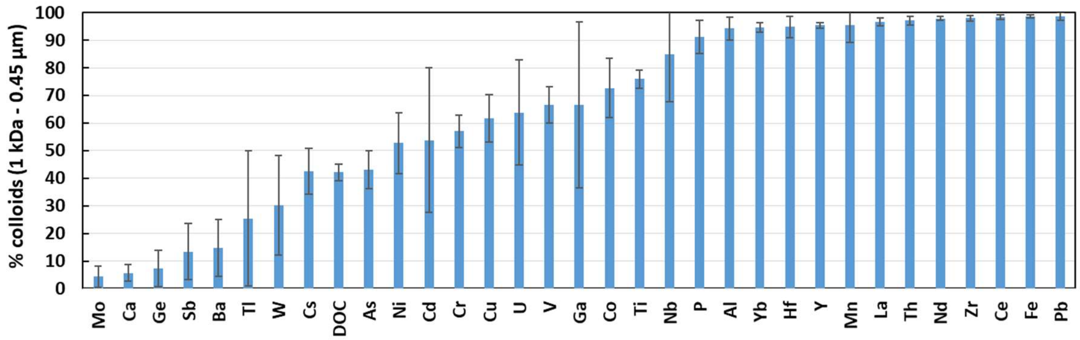

Figure 5. Proportion of colloidal fraction of elements in the Ob River downstream of Irtysh (4 points) and 3 tributaries (Pasaydeyakha, Poluy and Pitljar). Alkalis and alkaline-earth metals and oxyanions exhibit < 10% of colloids and not shown here.

Noteworthy that the role of the largest tributary (Irtysh) in colloidal pattern of the Ob River was not that significant: for most elements, their colloidal fraction in Irtysh was not statistically different from that of the main stem and downstream (northern) tributaries (**Fig. S4**).

3.4. Testing the landscape, soil and Quaternary deposits control on element concentraion

The main stem of the Ob River and its tributaries sampled in this study exhibited strong variability in main landscape parameters such as watershed size, MAAT, permafrost, forest, lake, bog and floodplain coverage as well as soils and Quaternary deposits. The main environmental parameters of the Ob River basin progressively evolved from the south to the north, which allowed testing the impact of climate and landscape on element concentration in several selected points of the main stem. The latitude of the sampling point, permafrost coverage and proportion of bogs, lakes, floodplain, lacustrine and fluvio-glacial Quaternary deposits of the watershed strongly correlated ($p < 0.05$) with DOC, Fe, P, divalent metals (Mn, Co, Ni, Cu, Pb), Rb and low mobile lithogenic trace elements (Al, Ti, Cr, Y, Zr, Nb, REEs, Hf, Th) concentrations (**Table S3 A, B; Fig. 6 and Fig. S5**). The pH and concentrations of soluble, highly mobile elements (DIC, SO_4, Ca, Sr, Ba, Mo, Sb, W, U) positively correlated with the proportion of forest, loess and fertile soils, eluvial, eolian, and fluvial Quaternary deposits on the Ob River watershed (**Fig. 6** and **Fig. S5** where for a number of most important parameters such as forest coverage (pH, Ca, Mo and U), floodplain area (DOC, P, Fe, Th) and watershed coverage by fluvio-glacial deposits (DOC, Al, Fe, La) are illustrated). Other landscape factors were of secondary importance for element control yet exhibited sizable correlations with particular elements. Examples are podzol soil coverage of the watershed that was positively linked to concentration of Al, Cr, Ga and Th, and saline soils that positively impacted the concentration of Cl, SO_4, Li, B, Na, K, As and Rb in the river water.

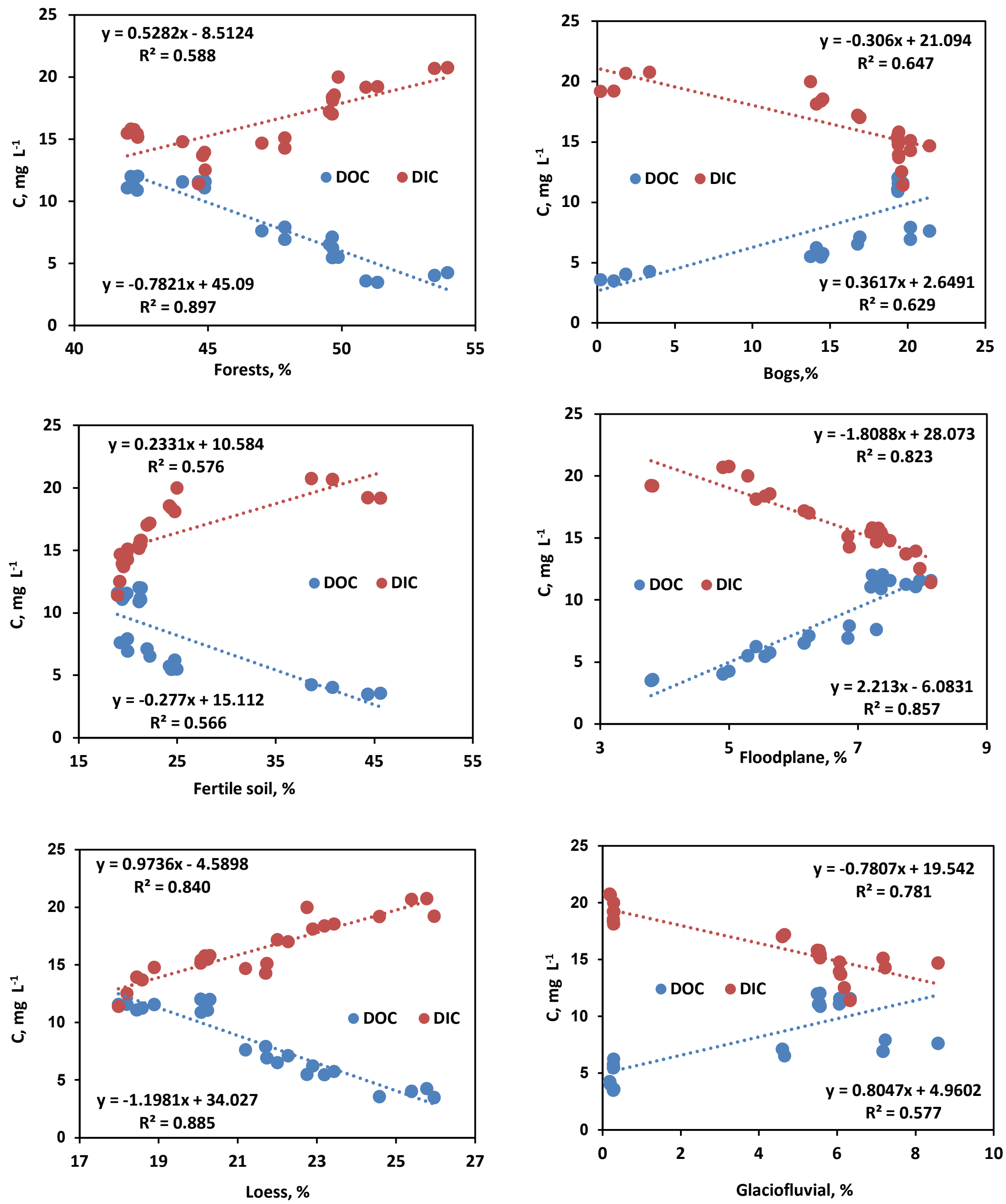

Figure 6. Examples of landscape factors, soil and Quaternary deposit control on DOC and DIC concentration in the Ob River. See examples of other factors and elements in **Fig. S5**. A correlation matrix of environmental parameters of the watershed and riverine solutes is provided in Table S3.

Pairwise correlations of riverine element concentration with landscape parameters of the watershed were further developed via multi-parametric statistical approach (Principal Component Analysis). Considering both the main stem of the Ob River and its tributaries, two main factors were revealed, accounting for 41% and 13% of total variability, respectively (**Fig. 7**). The F1 was presumably controlled by a northward increase in permafrost, floodplain, bogs, lakes and lacustrine deposits on the watersheds and acted on the DOC, Al, P, Ti, Fe, divalent metals, Rb, Cs, Nb, and trivalent and tetravalent trace elements concentrations. The second factor (F2) included the Specific Conductivity, SO_4^{2-}, Li, B, Na, labile alkaline-earth metals .Mg,

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Sr, Ba), oxyanions (Mo, Sb, W) and oxyanions (As, Sb) which were impacted by southward-dominating forest coverage, loesses, eluvial and fertile soils. This factorial structure was found to be highly stable and preserved in general features for both the Ob River main stem and tributaries, if treated separately (**Fig. S6**).

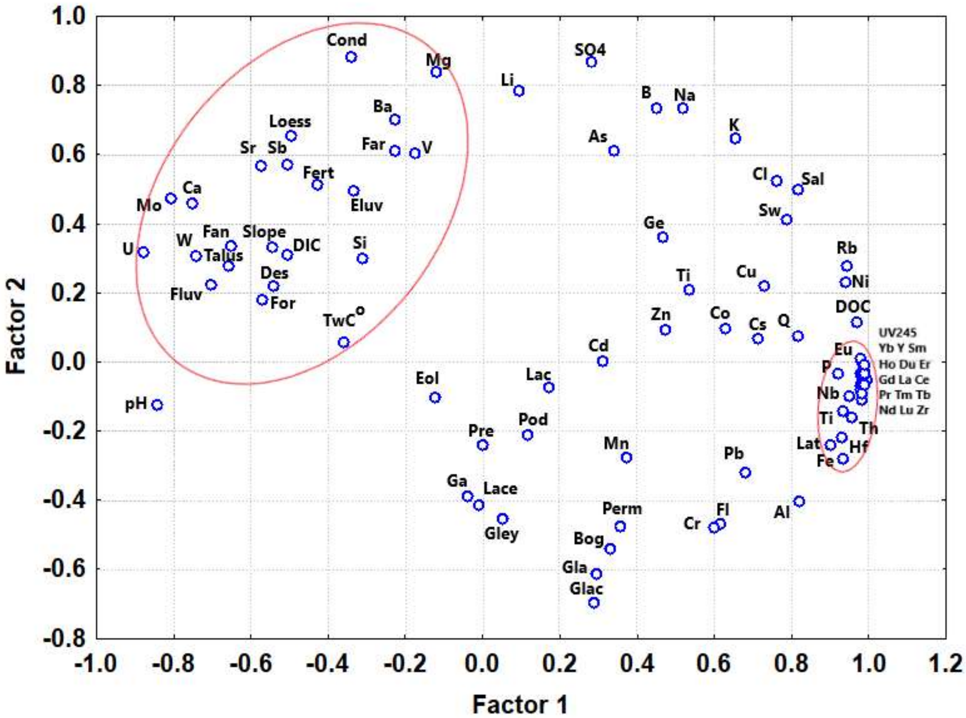

Figure 7. Results of the PCA treatment of all dataset (the Ob River main stem and tributaries) including elementary composition of the river water and landscape parameters of the watersheds. The landscape parameters (% of watershed coverage) are abbreviated as following: For, Forest; Fl, floodplain; Perm, permafrost; Far, farmland; Pre, pre-Quaternary deposits; Glac, glacial deposits; Slope, slopewash deposits, Des, deserptium (rock stream deposits); Lac, lacustrine and glacio-lacustrine deposits; Fan, fan-alluvial deposits; Gla, glacial deposits; Eol, eolian deposits; Pod, podsols; Sal, saline soils; Fert, fertile soils; TwC°, Water temperature; Cond, Specific Conductivity.

4. Discussion

4.1. Contrasting spatial pattern of DOC, major and trace elements between northern and southern river segment is due to latitudinal patetrn of landscape, soil and Quaternary deposits.

The hydrochemical composition of the Ob River basin demonstrated several distinct groups of elements defined according to the latitudinal patterns of their concentrations, similar to what was reported for small rivers of the WSL [31, 36]. The DOC, organically-bound metals (V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd and Pb) and the majority of low soluble TE (Al, REEs, Nb, Ti, Zr, Hf, and Th) exhibited a minimal concentration in the upper reaches of Ob and a maximal concentration in the northern, permafrost-affected zone. The northward increase in concentration of these elements in both main stem and tributaries may originate from multiple factors. First, enhanced input of lithogenic low mobility elements in the northern part of the Ob River (permafrost zone) may occur due to a decrease of river connectivity with deep and shallow groundwater and an increase in the surface flow. The surface drainage through forest litter together with surface runoff from surrounding bogs led to river water enrichment in organic and organo-ferric, organo-aluminum (DOC, Fe, Al) colloids [30,34]. These colloids act as main carriers of divalent metals, insoluble trivalent and tetravalent trace elements, Pb, Cr, V in western Siberian rivers [27, 28].

Another possible explanation for a northward increase in concentration of lithogenic and low mobile elements and a decrease in concentration of soluble labile elements lays in dynamics of peat formation / decay across the territory as recently suggested for small rivers of the WSL [36]. Because of on-going recovery from last glaciation, the WSL terrestrial ecosystems are not at the stationary stage: the growth of mires in the south leads to active accumulation of peat [40, 41] whereas the permafrost peatlans in the north thaw and degrade [42-44]. Given that the WSL peat preferentially accumulates heavy metals (V, Cr, Zn, Pb), trivalent (TE^{3+}) and tetravalent (TE^{4+}) trace elements (Al, Y, REEs, Ti, Zr, Hf, Th) and is depleted in alkalis and alkaline-earths metals, As, and Mo [45], it is possible that riverine concentration of trace elements inherits the element cycling between the growing/decaying peat and surface hydrological network.

At the same time, the obtained results did not confirm completely the initial hypothesis. An expected northward decrease of concentration of ground-water originated soluble, highly mobile anions, alkalis and alkaline earth metals was not observed for Cl, SO_4, Na and K. Presumably, these elements bear the influence of salt soils of the Irtysh River which flows through partially salty steppe and forest-steppe landscapes. Further to the north, the Ob River drains through paleo-marine deposits containing salt minerals (based on sedimentary cores available from extensive drilling of the territory [46]). This may enrich the main stem in highly mobile Na, Cl and SO_4.

It is important to note that, unlike it was reported for water pCO_2 and molecular composition of riverine DOM (i.e., 17, 18, 19], the Irtysh River does not play a regulatory role of the lower Ob River water chemistry in terms of concentrations of other major and trace elements including organo-mineral colloids. Instead, the hydrochemical conditions of the Ob River are shaped by integration of multiple tributaries those chemical composition gradually changes along the permafrost and landscape gradient, as it is known for other river basins of western Siberia such as Taz and Pur [36].

The concentration of riverine solutes was found to be strongly controlled by the abundance of bogs and lakes on the watershed and its floodplain coverage, for both the main stem and tributaries. The group of elements, positively correlated with wetlands, reflects a leading role of both organic-rich soils and sediments of wetlands (unified here as bogs, lakes and floodplains) in DOC, P, K, divalent metals and insoluble elements mobilization (notably trivalent and tetravalent hydrolysates) from the watershed to the river. This presents a prominent contrast to very small boreal catchments described in Northern Sweden, where wetlands decreased the fluxes of metals from boreal forests to downstream [47, 48, 49]. According to these authors, such a decrease occurred due to a combination of low weathering in peat soils and accumulation of organophilic metals in peat. The contrast between northern Scandinavian and western Siberian settings is consistent with a possibility of peat degradation rather than accumulation in northern part of the Ob River basin (downstream of the confluence with Irtysh). This would lead to enhanced release of trace metals to the streams and rivers. It is also possible that the presence of permafrost in the northern part of the WSL greatly shortens the water and element pathways between wetlands and rivers, compared to that in the non-permafrost regions of northern Sweden. The transport of trace metals released from mineral soils under the forest towards the river is therefore strongly enhanced by allochthonous organic matter originated from peat bogs and generally wetlands of the region.

Another landscape parameter is the presence of podzol soils at the watershed. Similar to glacial quaternary deposits, these soils led to enrichment of the river water in low mobile, lithogenic elements such as Al, Cr, Ga, REE, Hf, Th (see **Table S3**). These elements mark the presence of primary silicate minerals, often developed on felsic moraine as it is shown in European boreal regions (i.e., ref. [50]).

In contrast to metals and low mobile trace elements, the DIC, alkaline-earth metals, oxyanions (Mo, As, W) and U, those concentration decreased northward, were positively affected by the presence of forest, loess, eluvial, fluvial and fertile soils. From the one hand, this can reflect a northward decrease of the connectivity between groundwaters (enriched in these elements; see ref.

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[51]) and surface waters due to increase in the permafrost coverage (i.e., ref. [33, 34]). From the other hand, a southward increase in watershed coverage by soils which are rich in these labile elements (loess and eluvial and eolian soils, containing carbonate concretions and fauna) may explain preferential enrichment in these elements in the southern part of the Ob River watershed. Note that eolian solid aerosol deposits in western Siberia are also enriched in these soluble elements [52]. It is not excluded that both factors (soils and groundwaters) are pronounced at the scale of such a large watershed.

Finally, the presence of saline soils in the southern part of the Ob basin produced sizable enrichment of the river water in anions (Cl, SO_4), B, alkalis (Li, Na, K) presumably originated from local surface salt deposits and evaporates.

4.2. Riverine transport of trace element occurs in the form of organo-ferric colloids

All major and trace elements in the Ob River and its tributaries presented a colloidal pattern which was similar to the one established for boreal and permafrost-affected zones of high latitudes [28, 37, 53, 54, 55, 56, 57]. Soluble, highly labile alkalis, anions and oxyanions were negligibly affected by colloids as they do not interact with organic matter or colloidal Fe/Al hydroxides and thus present in the form of simple ions or neutral molecules. The second group of colloidally-bound trace elements reflected their capacity to either complex with colloidal organic matter (divalent transition metals), or co-precipitate with organo-ferric colloids (trivalent and tetravalent hydrolysates).

A prominent feature of the major part of the Ob Basin is that spatial variability of colloid distribution among the tributaries and the main stem was rather low, and only two southern tributaries (Ket, Tym) presented remarkable contrast to the rest of the Ob River basin. This presumably reflects homogeneous physico-geographical and landscape parameters of the majority of Ob River and its tributaries north of Irtysh. The two southern tributaries exhibit less amount of bogs and lakes in their watersheds and contain carbonate minerals in their base rocks (clays and silts with carbonate concretions, ref. 30). In accord with previous studies of small rivers of the WSL [58], we hypothesize that the presence of carbonate rocks and decreasing the proportion of wetlands in the southern part of the Ob basin lead to 1) a decrease in concentration of DOC and Fe which can serve as main carriers of trace metals; 2) an increase in DIC concentration and enhanced delivery of ionic form of elements from the groundwater. The latter is facilitated by much stronger connectivity between DOC-poor, DIC-rich groundwater and surface waters in permafrost-free part of the Ob Basin [34].

An interesting and unexpected result is extremely high proportion of colloidal Mn (83 to 100% in all samples except Ket (23%)). Typical colloidal fraction of Mn in boreal and permafrost-affected surface waters (rivers, lakes, bog waters) including peat porewater is between 80 and 20 % [28, 29], and the summer period usually accounts for the lowest proportion of colloidal Mn, due to the dominance of low molecular weight (< 1 k Da) exometabolites that are capable to complex divalent metals [27]. However, the lowest proportion of Mn colloids is observed only in the most southern tributary which also exhibits a low proportion of colloidal OC. In these sites, Mn is likely to be present as a divalent cation originated from the groundwater or complexed with autochthonous low molecular weight DOM. In contrast, the majority of Mn in all other tributaries and the main stem was tightly bound to colloids. We therefore hypothesize the transport of Mn in the form of high molecular weight, Fe-rich organic colloids in the main part of the Ob Basin. It is possible that high proportion of bogs on the watersheds of all rivers north of the Vasyugan River provides such an enhanced colloidal transport. These Mn-rich colloids are likely to be generated at the interface of anoxic and oxic surface waters of the bogs and then delivered to the river via surface flow. Note that enhanced riverine Mn transport in mire-affected regions is reported in Northern Europe [55, 59].

Another striking example of speciation control on element migration in the river water presents uranyl (UO_2^{2+}) ion. Uranium (VI) speciation in surface waters rich in DOM and Fe is largely controlled by high molecular weight organo-ferric colloids [28, 60]. In non-permafrost zone, these colloids are formed at the riparian/hyporheic zone of the river where the Fe(II) and U(VI)-rich groundwaters mix

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with oxygenated, organic-rich surface waters [37]. In the permafrost zone, these colloids originate from surface flow over forest floor and mire waters. Thus, in the northern part of the Ob River basin, elevated concentrations of both Fe and DOC provide suitable conditions for U transport in the form of colloids (70-90% in the 1 kDa - 0.45 µm form). Mires, which are highly abundant in the Ob basin north of Ket tributary, are also capable mobilizing uranium to the river water as it is known from other boreal setting [61]. In contrast, in southern part of the basin, the groundwater contacting with carbonate rocks of the basement or the loess soils are enriched in HCO_3^- which renders uranyl into soluble, non-colloidal carbonate complexes. Such a mechanism is fairly well known for other boreal regions which are impacted by carbonate rocks [62]. As a result, the proportion of colloidal form of UO_2(VI) progressively decreases southward and becomes as low as 3 % in two most southern tributaries (Ket and Tom) which exhibit the highest DIC, Ca and the lowest DOC and Fe concentration. Overall, progressive southward decrease of colloidal status of U reflects the increase of connectivity between surface and groundwaters of the WSL (i.e., ref. 34, 63) and a decrease of bog coverage of river watershed in the same direction.

4.3. Possible impact of landscape changes on element concentration in the Ob River and its tributaries.

Unique geographical position of the Ob River, which traverses, from the south to the north, several distinct landscape zones and encompasses a large permafrost, MAAT and vegetation gradient, allows to use a substituting space for time approach (i.e., ref. [64]) for foreseeing possible future changes in river water hydrochemistry based on contemporary pattern of riverine solutes. This approach has been efficiently used for small rivers of the WSL [30, 31, 33, 34, 35, 58], but, to the best of our knowledge, never attempted for the Ob River main stem. The restrictions of this approach are the following: lack of accounting for the time scale, necessary for the northern ecosystem to reach the new "more southern" state; ignoring possible shift in the structure of vegetation and soil microbial community, and change in hydrologic seasons. Considering these restrictions, only preliminary, first-order assessment of possible changes can be made.

Given rather high similarity of riverine solutes in the Ob River and tributaries across different permafrost zones, north of Irtysh (excluding the most southern part, south of Vasyugan), the shift in permafrost boundaries or the increase in the thickness of the active layer [65, 66, 67, 68], are not expected to sizably impact the hydrochemical composition of rivers in the Ob basin, unless the connectivity between deep underground and surface waters is modified (i.e., 10, 30). The latter may lead to enhanced concentration of soluble highly mobile elements in the north, as it is demonstrated for the case of small WSL rivers of the Pur and Taz watersheds [27].

At the same time, the elements affected by possible adsorption on clay minerals underlying the peat deposits, first of all, DOC (see [69, 70, 71, 72, 73] may modify their concentration in the northern part of the Ob basin. Given that wetlands (bogs, lakes and floodplain zone) were the main controlling factor of element concentration in the main stem and tributaries across the studied spatial gradient, we believe that global-level changes in WSL landscape such as forestation of the northern part and decrease in the proportion of bogs, all induced by changes in atmospheric precipitation, terrestrial productivity and duration of open-water seasons [i.e., 74]. However, quantification of the impact of these factors requires extensive ecosystem-level regional modeling which goes beyond the scope of this study.

5. Conclusions

Based on the 3000 km sampling transect of the Ob River and its 11 tributaries performed during end of the spring flood – beginning of the summer baseflow period, this snapshot study of large Arctic river dissolved (< 0.45 µm) load revealed the dominant physio-geographical control on the hydrochemistry of the river water.

The latitudinal pattern of element concentration in the main stem was strongly pronounced and distinguished two main groups of solutes. The spatial variations of main stem hydrochemistry were generally consistent with previous studies on carbon in the Ob River and on major and trace elements

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in small rivers of the WSL. The first group included mobile elements bearing the influence of groundwaters, connected to the river waters in the southern part of the WSL. These are DIC, Ca, Mo, W, and U which sizably (by a factor of 2.0) decreased their concentrations northward. The 2nd group of elements included DOC, P, Mn, Al, Fe, Ti, Ni, Cu, Co, Rb, Pb, Y, Zr, Nb, REE, Hf and Th which strongly (by a factor of 2 to 10) increased their concentration in the Ob River main stem from the south to the north. For these elements, the increase in concentrations occurred after confluence with Vasyugan River draining through the world's largest mire of peatland-lake complex.

The land cover approach allowed testing the control of main physio-geographical parameters of the Ob River watersheds on element concentration along the main steam and among the tributaries. Bogs, floodplain and lake coverage and permafrost presence on the watershed were found to be the main factors of northward increasing in concentration of DOC and low-soluble trace element, which were present in the form of organic and organo-mineral colloids (Fe, Mn, Al, V, Cr, Co, Ni, Cu, Zn, Cd and Pb and lithogenic trivalent and tetravalent TE (Al, Ti, Zr, Hf, Th) and REEs. These elements were also positively affected by the presence of glacial, lacustrine and fluvio-glacial Quaternary deposits. In contrast, soluble highly mobile alkalis, alkaline-earth metals, DIC, SO_4, B, Mo, Sb, As, W and U were positively affected by the presence of forest developed on loesses, fertile and saline soils, eluvial, fluvial and eolian Quaternary deposits given that these substrates could contain soluble carbonate minerals and salt inclusions.

We revealed high homogeneity in both element concentration and colloidal status in the main stem of the Ob River and its tributaries located north of the confluence with Irtysh. Presumably, the distribution of wetlands (bogs, floodplains, lakes and lacustrine deposits) was the dominant factor defining elementary pattern in waters of the Ob basin whereas the permafrost exhibited a subordinary control. As such, in case of drastic environmental changes in the WSL territory (permafrost boundary shift and active layer depth increase, vegetation coverage, precipitation regime), the changes of bogs, lakes and forest distribution rather than permafrost thaw might become the governing factors of modifications in the river hydrochemical regime.

Supplementary Materials: The following are available online at www.mdpi.com/xxx/s1, Figure S1: Latitudinal dependence of Cl, SO_4, Na, K and Pb concentration in the main stem of the Ob River; Figure S2: Main stem concentration of elements which do not show any particular pattern with latitude: Si, Mg, Cu, V, As and Sb; Figure S3: A histogram of elemental ratio in 2 northern tributaries (downstream of Irtysh) to 8 southern tributaries (upstream of Irtysh) of the Ob River; Figure S4: Proportion of colloidal (1 kDa - 0.45 μm) fraction of DOC, Ca, Fe, Mn, Al, Cu, U, La and Th in the main stem (blue box plot column) and tributaries of the southern (blue circles) and northern (red circles) part of the Ob Basin; Fig. S5. Examples of major and trace element concentration with landscape parameters of the Ob River main stem and tributaries; Fig. S6. Results of PCA treatment of the solute data and watershed characteristics (separately Ob main stem and tributaries). Table S1: List of sampled sites at the main stem of the Ob River and its tributaries. Table S2: Main landscape parameters and genetic type of Quaternary deposits (% of the watershed coverage) of the tributaries and several key points at the Ob River main stem; Table S3: A correlation matrix of element concentration in the Ob main stem tributaries and landscape coverage (%) of the watersheds; Table S4: Proportion of colloidal fraction of elements in the Ob River downstream of Irtysh (4 points) and 7 tributaries.

Author Contributions: YK and OP designed the study and wrote the paper; YK, LK, SV performed sampling, analysis of major cations and their interpretation; IP was the leader of the ship expedition and provided data interpretation; LS was in charge of DOC, DIC and anion measurements and their interpretation; LK and YK provided GIS-based interpretation, mapping and identification of river watersheds. Each co-author have seen and approved the final paper and contributed to writing the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

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SUPPLEMENTARY MATERIAL

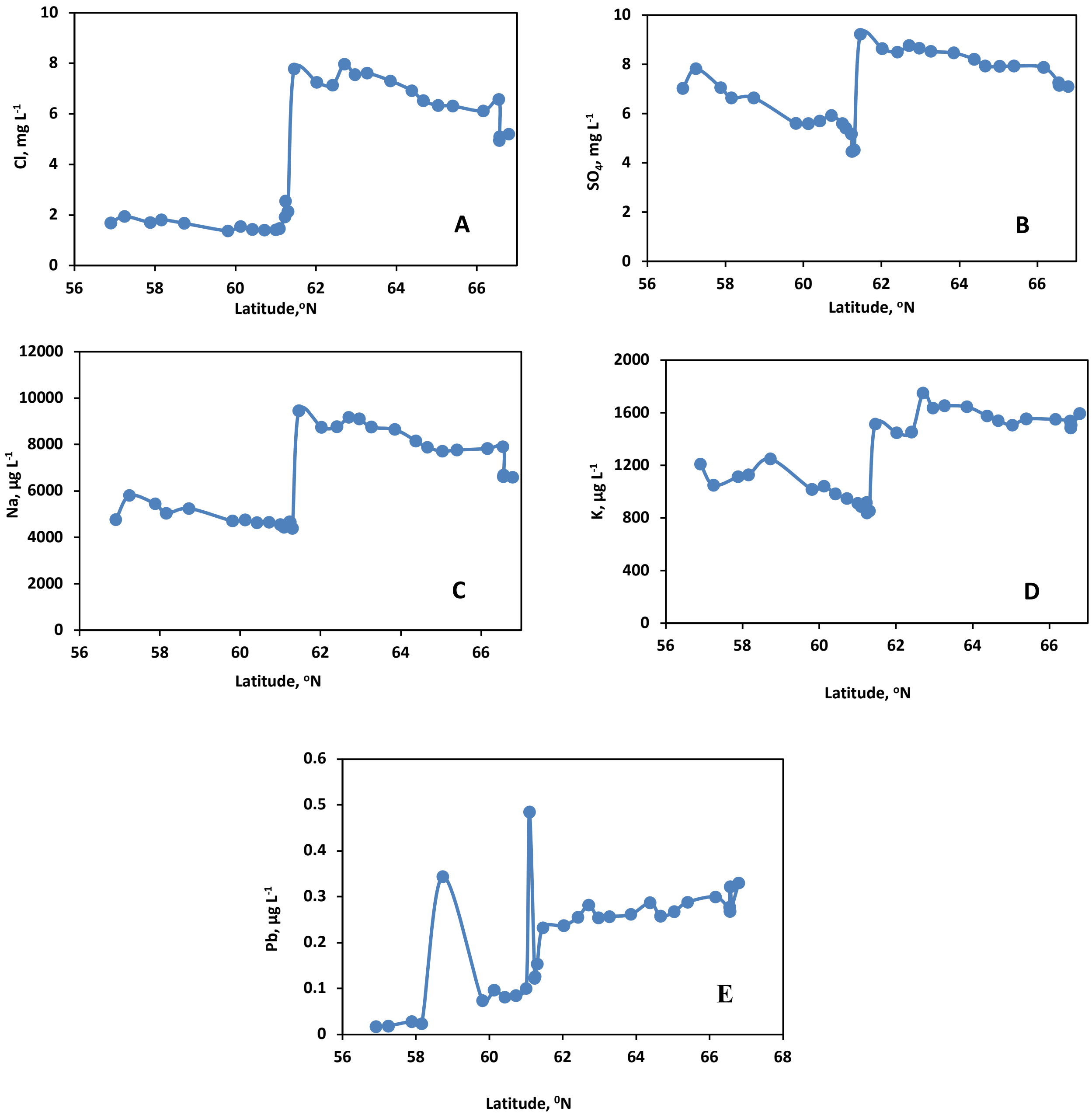

Figure S1. Latitudinal dependence of Cl (A), SO_4 (B), Na (C), K(D) and Pb (E) concentration in the main stem of the Ob River. A sharp increase visible at approx. 61°N is the confluence with the Irtysh River.

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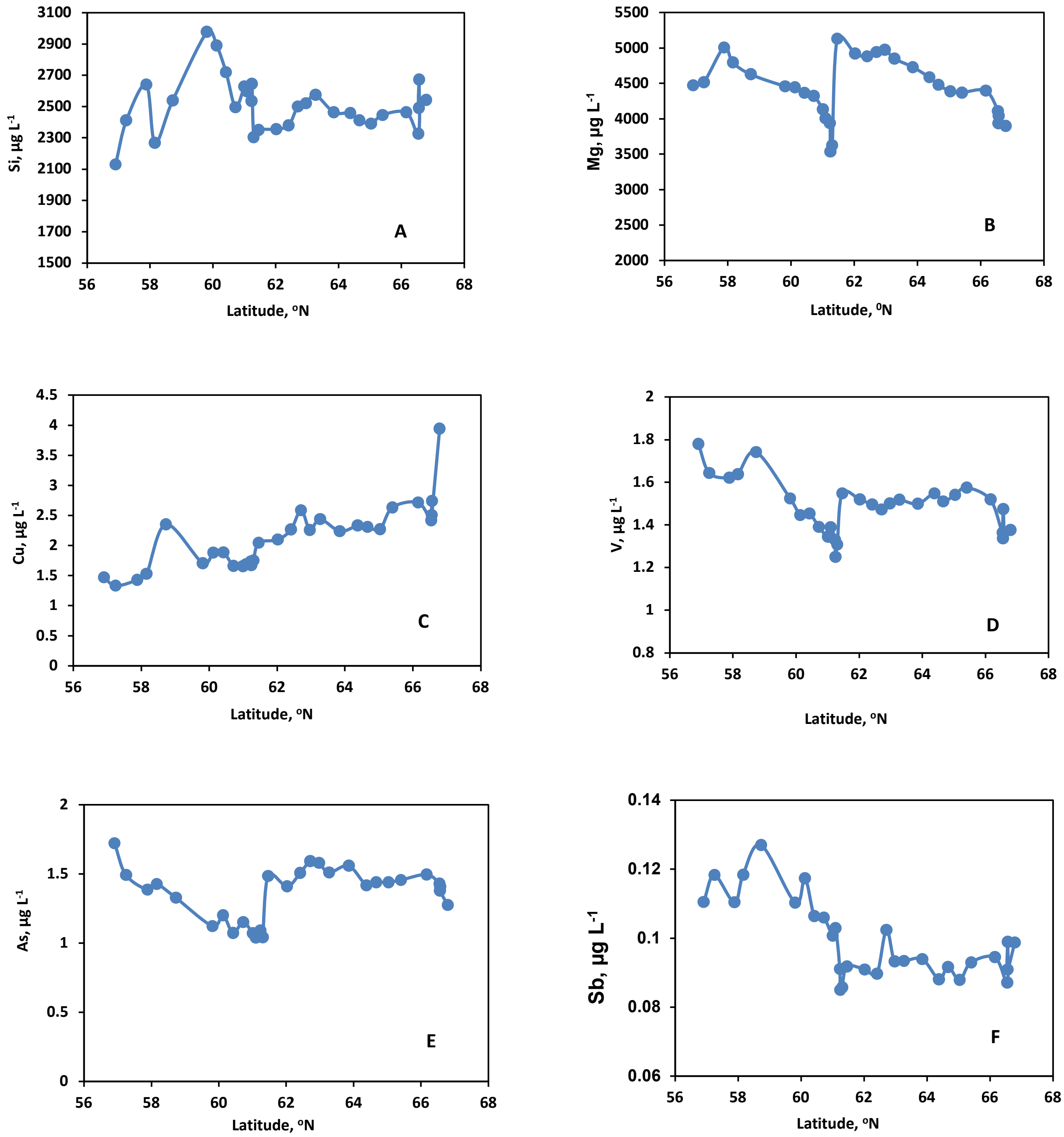

Figure S2. Main stem concentration of elements which do not show any particular pattern with latitude: Si (A), Mg (B), Cu (C), V (D), As (E) and Sb (E).

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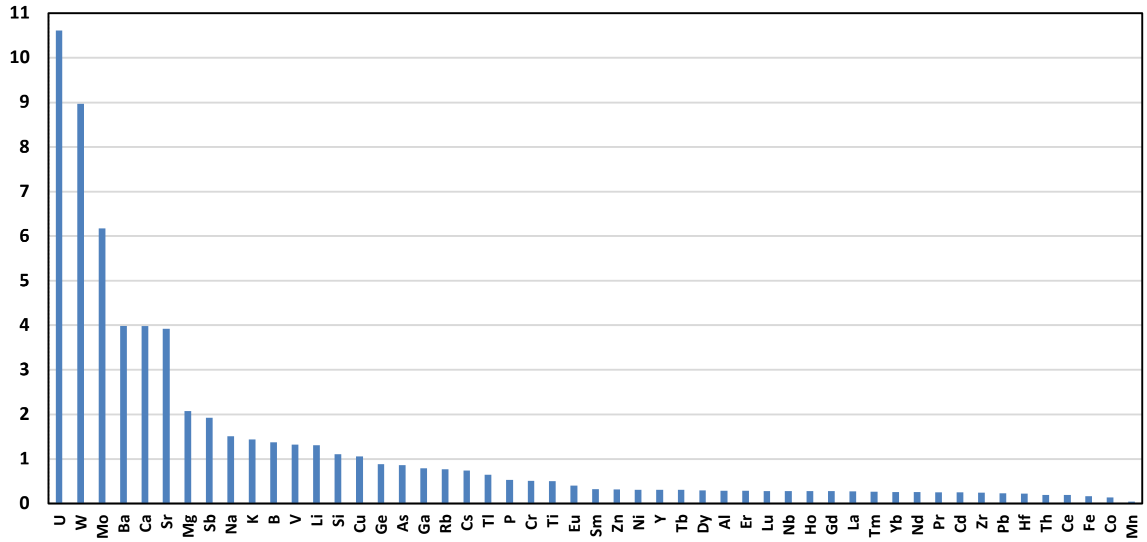

Figure S3. A histogram of elemental ratio in **2** northern tributaries (downstream of Irtysh) to **8** southern tributaries (upstream of Irtysh) of the Ob River.

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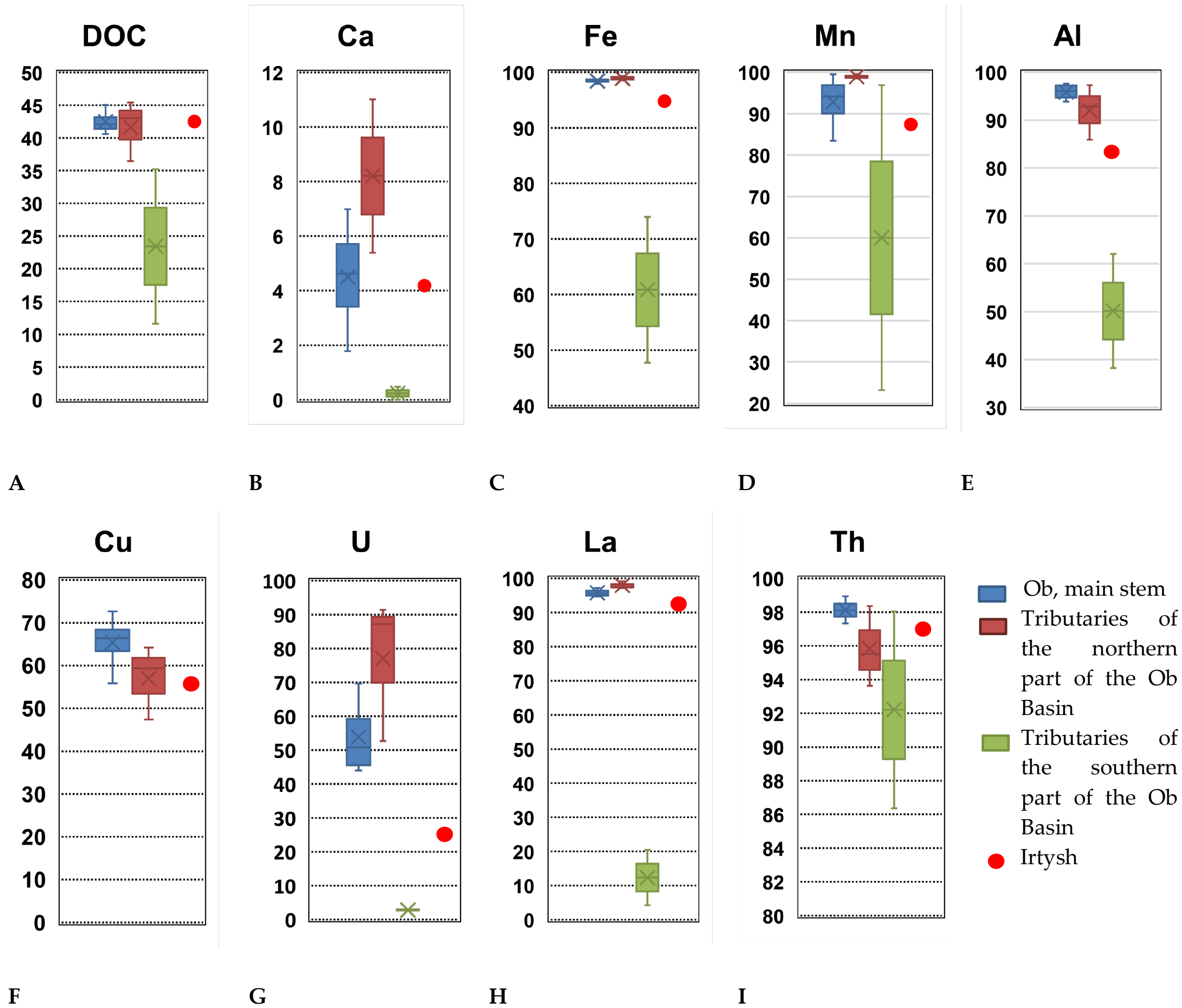
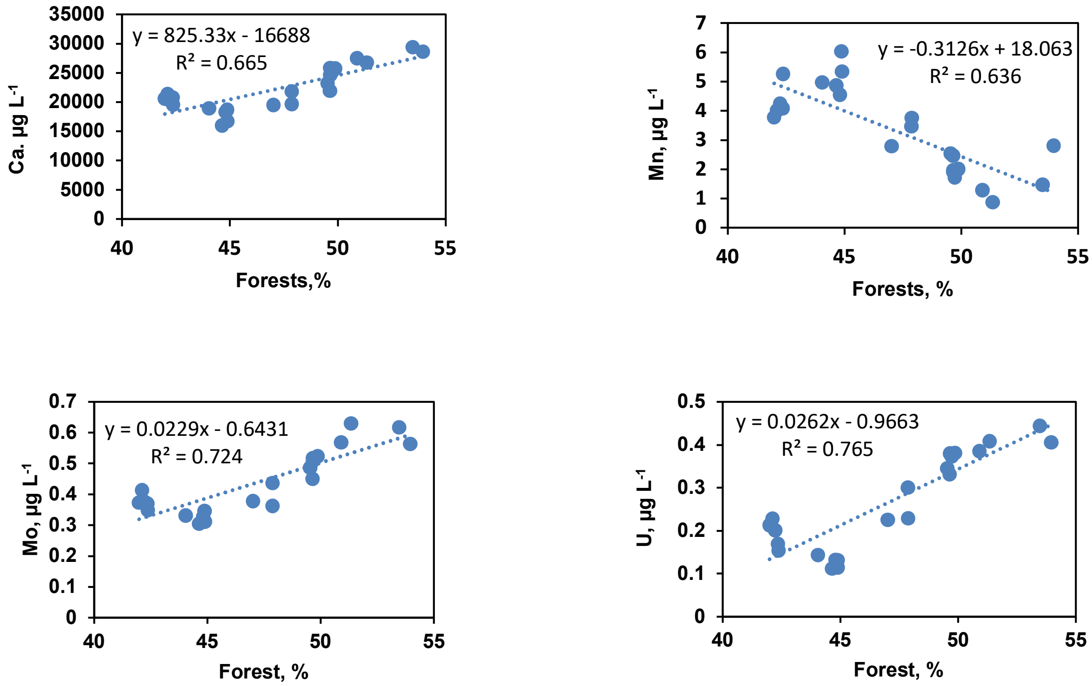
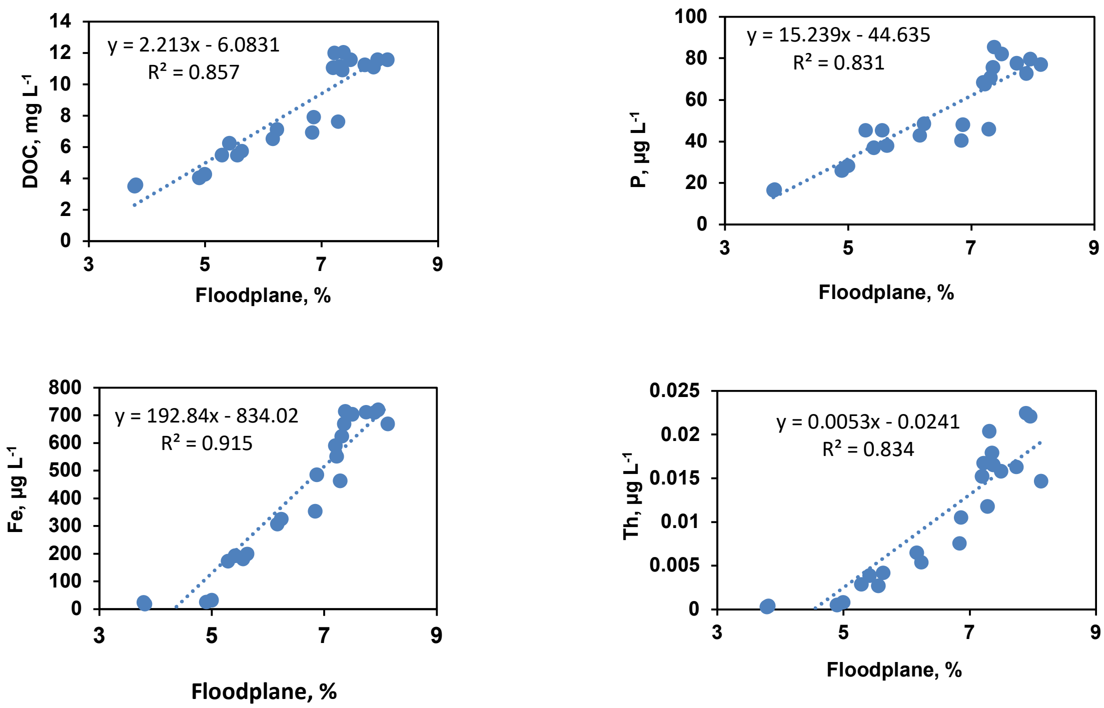


Figure S4. Proportion of colloidal (1 kDa - 0.45 µm) fraction of DOC (A), Ca (B), Fe (C), Mn (D), Al (E), Cu (F), U (G), La (H) and Th (I) in the main stem (blue box plot column) and tributaries of the southern (blue circles) and northern (red circles) part of the Ob Basin.



A



B

Fig. S5. Examples of major and trace element concentration with landscape parameters of the Ob River main stem: **A:** Impact of the forest coverage on Ca, Mn, Mo and U; **B:** Impact of the floodplain coverage on DOC, P, Fe and Th concentrations; **C:** Impact of the fluvio-glacial deposits on DOC, Al, Fe and La concentrations.

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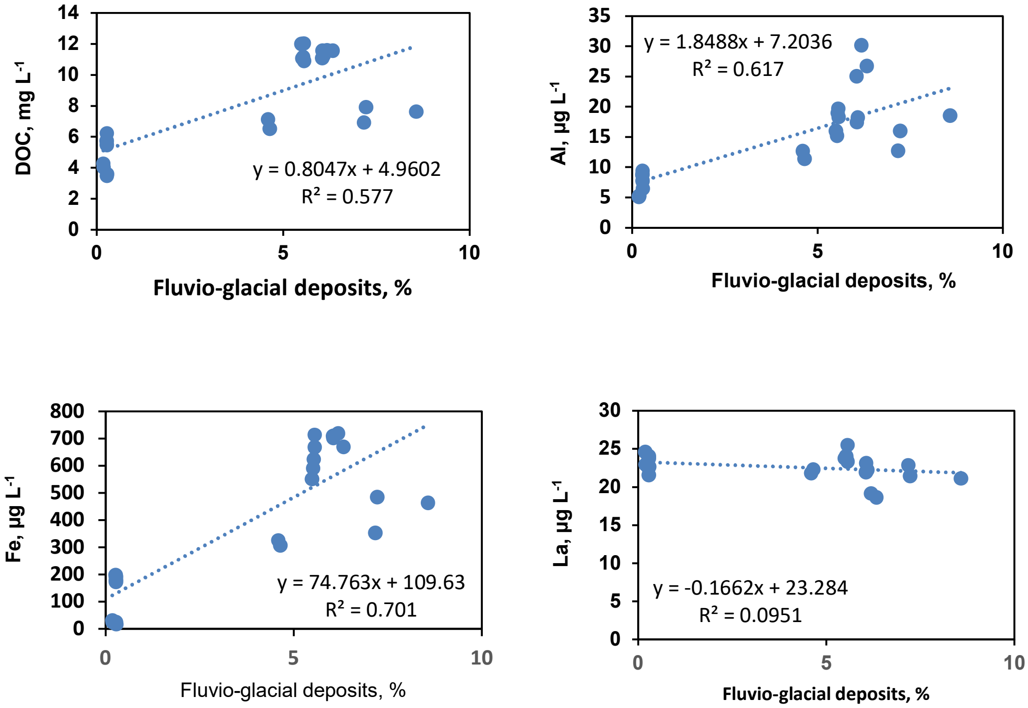

C

Fig. S5, continued. Examples of major and trace element concentration with landscape parameters of the Ob River main stem: **C:** Impact of the fluvio-glacial deposits on DOC, Al, Fe and La concentrations.

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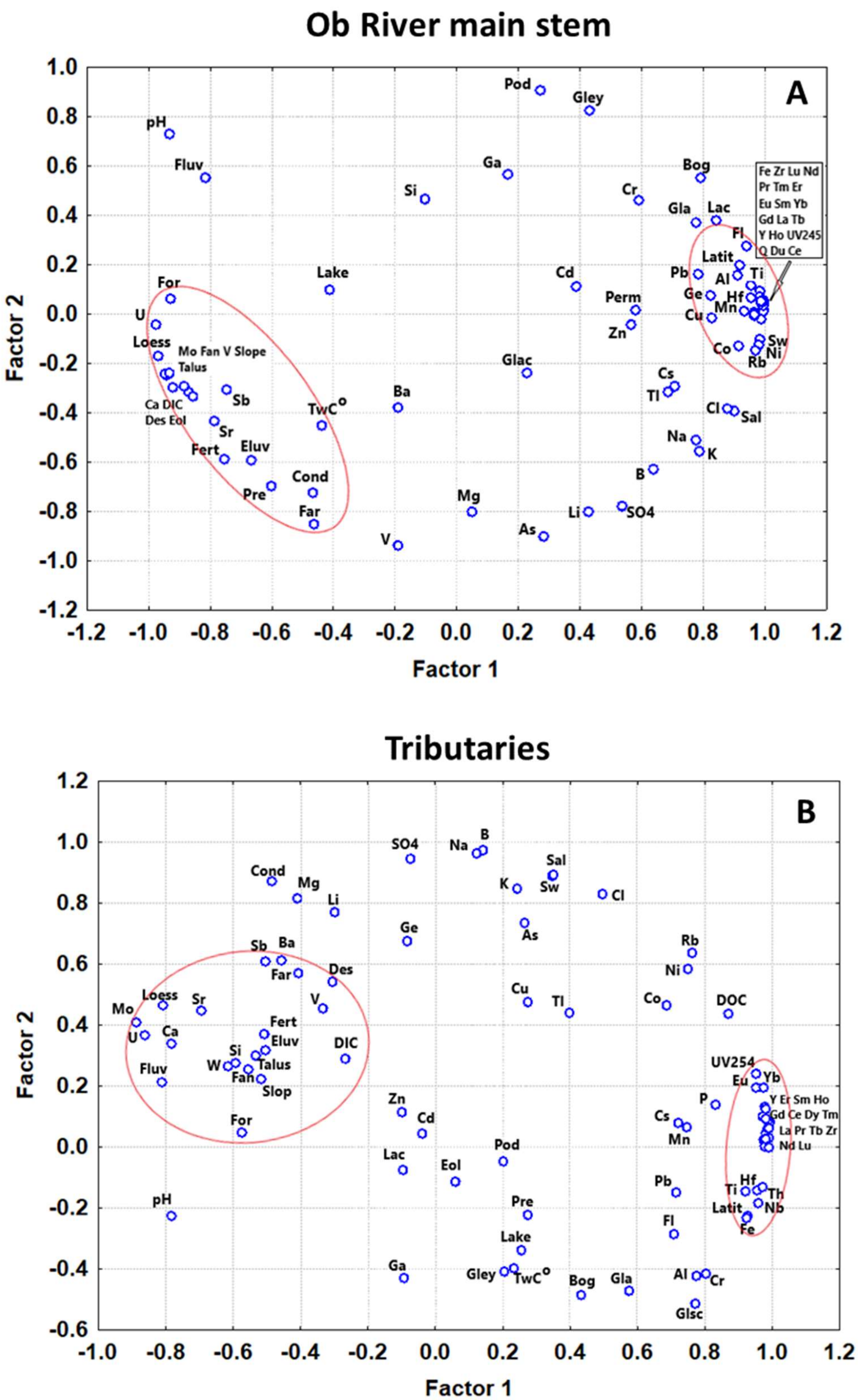

Fig. S6. Results of PCA treatment of the solute data and watershed characteristics for the Ob River main stem (a) and tributaries (b). See Fig. 7 for abbreviation of landscape parameters.

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Table S1. List of sampled sites at the main stem of the Ob River and its tributaries.

Name	Number on the map	Descriptions, km	N	E	date
Ob's g1	1	0	66.788694	69.143972	08.07.16, 21:37
Ob's g2	2	74.241	66.554056	67.839944	09.07.16, 11:50
Ob's 1	3	120.2	66.561925	67.032767	10.07.2016
Ob's g3	4	153.8	66.544861	66.511833	13.07.16, 11:30
Ob's 2	5	209.7	66.158222	65.763806	13.07.16, 19:00
Ob's 3	6	299.7	65.4005	65.641667	14.07.16, 0:28
Ob's g4	7	344.9	65.034722	65.240833	14.07.16, 07:48
Ob's g5	8	408.6	64.664889	65.613806	14.07.16, 19:06
Ob's 4	9	452.4	64.378278	65.904444	15.07.16, 00:10
Ob's p4	10	527.3	63.852222	65.368917	15.07.16, 07:30
Ob's 5	11	610.2	63.269361	64.991528	15.07.16, 15:10
Ob's 6	12	648.4	62.968083	65.069278	15.07.16, 20:26
Ob's p5	13	687.5	62.7064	65.508883	16.07.16, 0:45
Ob's 7	14	744.4	62.411503	66.333239	16.07.16, 19:15
Ob's g6	15	821.2	62.022069	67.369903	17.07.16, 1:22
Ob's 8	16	909.0	61.461306	68.224083	17.07.16, 8:00
Ob's 9	17	1149.2	61.302833	71.286006	18.07.16, 8:03
Ob's 10	18	1253.9	61.243361	72.849944	18.07.16, 19:27
Ob's 11	19	1291.9	61.232167	73.538694	19.07.16, 0:25
Ob's 12	20	1419.9	61.09325	75.326528	19.07.16, 8:00
Ob's 13	21	1469.9	61.004361	76.085556	19.07.16, 13:15
Ob's 14	22	1550.6	60.720639	77.114083	19.07.16, 23:13
Ob's 15	23	1624.8	60.420694	77.978444	20.07.16, 5:25
Ob's 16	24	1702.0	60.124361	78.915167	20.07.16, 12:06
Ob's 17	25	1752.4	59.811222	79.116	20.07.16, 17:51
Ob's 18	26	2185.3	58.159194	82.941139	22.07.16, 15:15
Ob's 19	27	2230.4	57.883861	83.345361	22.07.16, 19:15
Ob's 20	28	2361.7	57.245944	84.348944	23.07.16, 07:23
Ob's 21	29	2410.2	56.906833	84.448892	23.07.16, 12:51
Ob's p1	p_1 (Pasaydeyakha)	0	67.709	72.904389	06.07.2016
Ob's p2	p_2 (Poluy)	291.0	66.522917	66.599611	10.07.16, 20:53
Ob's p3	p_3 (Pitljar)	397.0	65.771694	65.510778	13.07.16, 22:20
Ob's p6	p_4 (Irtysh)	975.7	61.067947	68.929806	17.07.16, 17:22
Ob's p7	p_5 (Nazym)	1172.0	61.193594	68.922253	17.07.16, 19:21
Ob's p8	p_6 (Ljamin)	1172.0	61.280639	71.792972	18.07.16, 11:39
Ob's p11	p_7 (Tym)	1623.0	59.431417	80.02025	21.07.16, 03:09
Ob's p10	p_8 (Vasjugan)	1257.0	59.121222	80.744258	21.07.16, 11:31
Ob's p12	p_9 (Kopilovskaja Ket')	1369.0	58.911256	81.559931	21.07.16, 20:00
Ob's g7	p_10 (Parabel')	2189.0	58.727083	81.573639	21.07.16, 23:33
Ob's p13	p_11 (Chulym)	2542.0	57.729083	83.822494	23.07.16, 01:00
Ob's p14	p_12 (Tom')	2677.0	56.795861	84.530528	23.07.16, 12:00

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Table S2. A: Main landscape parameters (% of the watershed coverage) of the tributaries and several key points at the Ob River main stem.

Sampling point	latitude	catchment area. km²	Q. m³/s	Forests	Bogs	Lakes	Floodplain	Farmland	Permafrost	Podzol	Gley soils	Saline soils	Fertile soil
g1	66.7887	2479211	31683	45	20	0.02	8.13	19	1.99	10.97	5.77	1.73	18.92
1	66.5619	2450170	30835	45	20	0.00	7.96	19	1.25	11.05	5.83	1.76	19.14
2	66.1582	2416527	30050	45	19	0.01	7.89	19	0.75	11.04	5.50	1.78	19.41
3	65.4005	2399324	29616	45	19	0.00	7.74	19	0.62	11.00	5.34	1.79	19.55
4	64.3783	2360079	28625	44	19	0.00	7.50	20	0.43	10.66	4.96	1.82	19.87
5	63.2694	2222409	25151	42	19	0.00	7.38	21	0.03	11.32	5.14	1.93	21.10
6	62.9806	2220366	25099	42	19	0.00	7.35	21	0.03	11.32	5.11	1.94	21.12
7	62.4115	2210933	24861	42	19	0.00	7.31	21	0.06	11.37	5.13	1.94	21.21
g6	62.0221	2203777	24680	42	19	0.00	7.20	21	0.03	11.40	4.93	1.95	21.28
8	61.4613	2196865	24506	42	19	0.02	7.22	21	0.00	11.43	4.86	1.96	21.35
9	61.3028	1100150	18220	47	21	0.27	7.28	15	0.00	16.59	5.69	0.07	19.20
10	61.2434	1059511	16353	48	20	0.01	6.86	15	0.00	17.22	5.91	0.07	19.94
11	61.2322	1058205	16295	48	20	0.37	6.84	15	0.00	17.22	5.88	0.07	19.97
12	61.0933	963155	12046	50	17	0.00	6.24	17	0.00	15.36	6.03	0.08	21.94
13	61.0044	951873	11542	50	17	0.50	6.16	17	0.00	14.88	6.03	0.08	22.20
14	60.7206	873109	8021	50	15	0.01	5.63	19	0.00	12.40	5.95	0.09	24.20
15	60.4207	865715	7730	50	14	0.03	5.55	19	0.00	12.09	5.91	0.09	24.40
16	60.1244	853949	7607	50	14	0.05	5.42	19	0.00	11.48	5.98	0.09	24.74
17	59.8112	846169	7538	50	14	0.22	5.29	19	0.00	11.25	6.04	0.09	24.97
18	58.1592	547112	5763	54	3	0.01	5.00	30	0.00	5.13	3.77	0.14	38.62
19	57.8839	518674	5464	53	2	0.05	4.90	31	0.00	5.30	1.71	0.15	40.73
20	57.2459	360677	3799	51	1	0.00	3.79	37	0.00	5.78	1.99	0.20	44.33
21	56.9068	344424	3628	51	0	0.77	3.81	39	0.00	6.05	1.53	0.21	45.62
Tom'	56.7958	74581	702	61	0	0.04	2	38	0.00	0.74	0.00	0.11	84.36
Thculym	57.7291	133600	628	66	2	0.17	8	23	0.00	2.83	0.00	0.04	37.28
Parabel'	58.7271	24600	90	58	38	0.37	4	0	0.00	2.69	37.56	0.00	0.00
Vasjugan	59.1212	66404	214	60	40	0.20	1	0	0.00	10.70	30.62	0.00	0.00
Tym	59.4314	37366		53	40	0.39	8	0	0.00	59.45	0.00	0.00	0.00
L'yamin	61.2806	27400		15	67	10.95	11	0	0.00	22.70	0.82	0.00	0.00
Nazym	61.1936	11395		57	30	1.70	13	0	0.00	26.48	17.39	0.00	0.00
Irtysh	61.0679	2183722	5980	41	18	0.53	6.85	21	0.00	10.95	4.51	1.97	21.48
Poluj	66.5229	20000	138	45	41	0.50	11	0	10.00	12.46	33.19	0.00	0.00

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Table S2. B: Main genetic types of Quaternary deposits, % of the watershed. Slopewash, Desertium, and Fanalluvial deposits represent less than 2% and not listed here.

	Pre-Quaternary	Loess	Glacial	Eluvial	Lacustrine	Fluvial	Talus	Fluvio-glacial	Eolian
The Ob River main stem									
g1	2.03	17.98	5.12	2.84	16.18	22.40	1.04	6.33	0.54
1	2.04	18.20	4.97	2.87	16.16	22.41	1.05	6.18	0.55
2	2.07	18.45	4.69	2.89	15.99	22.41	1.06	6.05	0.56
3	2.08	18.58	4.52	2.90	15.94	22.36	1.07	6.09	0.56
4	2.11	18.89	4.02	2.94	15.74	22.33	1.09	6.06	0.57
5	2.16	20.06	2.13	2.98	15.73	22.30	1.16	5.56	0.60
6	2.16	20.08	2.09	2.98	15.73	22.29	1.16	5.56	0.60
7	2.16	20.16	1.96	3.00	15.69	22.27	1.16	5.54	0.61
g6	2.13	20.23	1.88	3.01	15.69	22.24	1.17	5.52	0.61
8	2.11	20.29	1.88	3.01	15.64	22.19	1.17	5.49	0.61
9	1.83	21.20	3.06	2.59	12.88	28.71	2.15	8.57	0.71
10	1.86	21.71	2.83	2.69	12.97	28.82	2.24	7.22	0.67
11	1.86	21.73	2.83	2.69	12.98	28.80	2.24	7.17	0.67
12	2.04	22.27	2.23	2.96	12.66	29.31	2.46	4.59	0.64
13	2.07	22.00	2.23	2.99	12.48	29.30	2.49	4.65	0.64
14	2.18	23.43	1.85	3.26	12.91	29.68	2.72	0.27	0.66
15	2.20	23.19	1.85	3.29	12.90	29.66	2.74	0.27	0.66
16	2.23	22.89	1.88	3.34	13.03	29.34	2.78	0.28	0.67
17	2.25	22.75	1.89	3.37	13.15	29.05	2.80	0.28	0.68
18	3.06	25.77	2.93	5.21	5.39	27.23	4.33	0.18	0.89
19	3.22	25.39	3.09	5.49	3.45	27.27	4.57	0.19	0.94
20	4.15	25.97	4.40	4.02	2.83	26.20	4.94	0.27	0.83
21	4.35	24.58	4.60	4.21	1.95	26.35	5.18	0.29	0.86
Tributaries									
Tom'	1.57	27.62	0.67	6.07	0.52	39.46	3.03	0.00	0.02
Tchulym	1.37	33.46	0.13	10.46	4.13	31.92	4.40	0.00	1.41
Parabel'	0.61	20.33	0.00	0.00	44.15	34.89	0.00	0.00	0.00
Vasjugan	1.13	24.56	0.00	0.00	47.69	27.03	0.00	0.00	0.00
Tym	0.98	18.47	0.00	0.00	33.39	39.05	0.00	3.69	0.83
L'yamin	1.46	0.00	13.26	0.00	3.42	18.52	0.00	59.74	2.75
Nazym	21.36	0.00	25.87	0.00	0.96	16.29	0.00	33.12	0.31
Irtysh	2.02	20.15	1.70	3.03	15.05	21.65	1.18	5.12	0.61
Poluj	0.00	0.00	27.28	0.00	28.05	21.07	0.00	23.67	0.00

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Table S3 A and B. A correlation matrix of element concentration in the Ob main stem tributaries and landscape coverage (%) of the watersheds. Significant ($p < 0.05$, n = 23 for Ob and n = 9 for tributaries)) correlations are given in red.

A: Landscape and soil characteristics

	Latitude	Forest	Bogs	Lakes	Floodplain	Farmland	Permafrost	Podzol	Gley soil	Saline soil	Fertile soil
S.C.	-0.69	0.34	-0.53	0.01	-0.69	0.57	-0.43	-0.62	-0.44	0.29	0.54
pH	-0.85	0.74	-0.04	0.56	-0.56	-0.56	-0.81	0.33	0.37	-0.74	0.14
UV_{254}	0.93	-0.80	0.51	-0.28	0.75	-0.19	0.73	0.31	0.18	0.39	-0.56
Cl	0.66	-0.75	0.23	-0.40	0.42	0.29	0.61	-0.05	-0.25	0.69	-0.24
SO_4	0.10	-0.30	-0.35	-0.52	-0.09	0.73	0.34	-0.61	-0.52	0.83	0.27
DOC	0.88	-0.80	0.48	-0.28	0.73	-0.12	0.67	0.29	0.11	0.43	-0.51
DIC	-0.76	0.48	-0.41	0.17	-0.80	0.21	-0.61	-0.28	-0.13	-0.06	0.46
Li	-0.01	-0.18	-0.30	-0.43	-0.06	0.62	0.25	-0.48	-0.57	0.69	0.22
B	0.18	-0.28	-0.07	-0.32	0.22	0.49	0.42	-0.44	-0.49	0.61	-0.01
Na	0.24	-0.33	-0.10	-0.42	0.17	0.57	0.49	-0.48	-0.47	0.71	0.04
Mg	-0.39	0.11	-0.42	-0.16	-0.29	0.63	-0.14	-0.55	-0.60	0.44	0.40
Al	0.90	-0.64	0.54	-0.27	0.76	-0.29	0.74	0.43	0.21	0.21	-0.59
Si	-0.29	0.31	-0.18	0.07	-0.18	-0.07	-0.20	-0.12	0.03	-0.19	0.14
P	0.86	-0.69	0.42	-0.35	0.78	-0.13	0.76	0.24	0.11	0.39	-0.50
K	0.39	-0.42	-0.01	-0.44	0.25	0.40	0.57	-0.41	-0.26	0.71	-0.12
Ca	-0.92	0.68	-0.48	0.31	-0.69	0.25	-0.74	-0.36	-0.29	-0.25	0.53
Ti	0.93	-0.70	0.54	-0.31	0.82	-0.19	0.77	0.33	0.13	0.33	-0.59
V	-0.31	0.11	-0.21	-0.12	-0.42	0.38	-0.08	-0.50	-0.29	0.39	0.16
Cr	0.68	-0.49	0.70	0.05	0.62	-0.53	0.38	0.63	0.35	-0.08	-0.66
Mn	0.80	-0.63	0.53	-0.26	0.53	-0.17	0.69	0.14	0.20	0.35	-0.56
Fe	0.97	-0.74	0.56	-0.39	0.80	-0.26	0.81	0.37	0.22	0.32	-0.61
Co	0.66	-0.50	0.38	-0.27	0.63	0.02	0.70	-0.09	-0.10	0.35	-0.46
Ni	0.85	-0.70	0.49	-0.24	0.82	-0.13	0.74	0.19	0.04	0.37	-0.57
Cu	0.71	-0.59	0.52	-0.26	0.58	-0.24	0.62	0.15	0.10	0.36	-0.62
Zn	0.29	0.02	0.14	-0.08	0.32	0.03	0.53	-0.44	0.11	0.14	-0.32
Ga	0.07	0.09	0.38	0.23	0.19	-0.45	-0.08	0.47	0.15	-0.43	-0.30
Ge	0.45	-0.44	0.36	-0.10	0.34	-0.07	0.45	-0.04	-0.09	0.29	-0.41
As	0.11	-0.26	-0.10	-0.27	0.02	0.48	0.36	-0.36	-0.38	0.64	0.03
Rb	0.86	-0.81	0.46	-0.30	0.70	-0.11	0.67	0.29	0.04	0.50	-0.51
Sr	-0.85	0.53	-0.51	0.25	-0.63	0.39	-0.64	-0.45	-0.40	-0.04	0.55
Y	0.96	-0.79	0.50	-0.42	0.75	-0.17	0.78	0.29	0.13	0.45	-0.57

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	Latitude	Forest	Bogs	Lakes	Floodplain	Farmland	Permafrost	Podzol	Gley soil	Saline soil	Fertile soil
Zr	0.96	-0.78	0.54	-0.35	0.76	-0.21	0.81	0.33	0.19	0.38	-0.59
Nb	0.93	-0.76	0.56	-0.38	0.71	-0.20	0.76	0.35	0.16	0.40	-0.60
Mo	-0.94	0.67	-0.48	0.35	-0.70	0.26	-0.77	-0.36	-0.34	-0.26	0.53
Cd	0.40	-0.20	0.08	-0.23	0.33	-0.02	0.56	-0.26	0.22	0.21	-0.24
Sb	-0.62	0.44	-0.43	0.10	-0.66	0.25	-0.39	-0.44	-0.16	0.06	0.39
Cs	0.61	-0.52	0.30	-0.22	0.50	0.11	0.68	-0.02	-0.11	0.46	-0.37
Ba	-0.37	0.08	-0.28	-0.01	-0.27	0.35	-0.26	-0.30	-0.35	0.26	0.29
La	0.97	-0.77	0.50	-0.43	0.76	-0.18	0.78	0.30	0.16	0.44	-0.58
Ce	0.98	-0.78	0.52	-0.39	0.77	-0.21	0.81	0.30	0.19	0.40	-0.60
Pr	0.97	-0.77	0.50	-0.43	0.75	-0.18	0.79	0.31	0.16	0.44	-0.57
Nd	0.97	-0.78	0.52	-0.42	0.77	-0.19	0.80	0.32	0.16	0.43	-0.59
Sm	0.95	-0.79	0.49	-0.43	0.74	-0.16	0.77	0.30	0.14	0.46	-0.56
Eu	0.94	-0.80	0.48	-0.41	0.72	-0.15	0.77	0.31	0.12	0.47	-0.55
Gd	0.97	-0.78	0.52	-0.42	0.76	-0.19	0.80	0.30	0.16	0.42	-0.58
Tb	0.96	-0.79	0.51	-0.42	0.76	-0.18	0.80	0.30	0.14	0.43	-0.58
Dy	0.96	-0.79	0.50	-0.45	0.74	-0.17	0.79	0.30	0.14	0.44	-0.57
Ho	0.96	-0.79	0.50	-0.43	0.72	-0.17	0.79	0.31	0.15	0.46	-0.57
Er	0.95	-0.79	0.49	-0.39	0.74	-0.17	0.78	0.30	0.14	0.45	-0.56
Tm	0.97	-0.79	0.50	-0.37	0.75	-0.19	0.80	0.28	0.19	0.42	-0.57
Yb	0.95	-0.80	0.49	-0.38	0.72	-0.17	0.77	0.29	0.14	0.45	-0.57
Lu	0.96	-0.77	0.54	-0.36	0.76	-0.21	0.79	0.34	0.19	0.39	-0.59
Hf	0.93	-0.73	0.46	-0.31	0.75	-0.13	0.74	0.33	0.12	0.37	-0.50
W	-0.81	0.58	-0.34	0.32	-0.78	0.07	-0.73	-0.16	-0.11	-0.27	0.40
Tl	0.43	-0.51	0.32	-0.14	0.41	0.15	0.43	0.01	-0.36	0.35	-0.30
Pb	0.81	-0.60	0.54	-0.26	0.58	-0.35	0.67	0.19	0.41	0.20	-0.61
Th	0.95	-0.78	0.53	-0.35	0.73	-0.20	0.76	0.37	0.21	0.39	-0.56
U	-0.96	0.73	-0.45	0.38	-0.68	0.18	-0.81	-0.31	-0.28	-0.36	0.51

B: Main genetic types of Quaternary deposits

	Loess	Glacial	Eluvial	Lacustrine	Fluvial	Fluvio-Glacial	Eolian
S.C.	0.60	-0.64	0.60	-0.10	0.25	-0.82	0.19
pH	0.62	-0.32	-0.05	-0.56	0.57	-0.39	0.28
UV_{254}	-0.83	0.38	-0.47	0.54	-0.67	0.76	-0.48
Cl	-0.57	0.31	-0.15	0.29	-0.75	0.54	-0.42
SO4	0.02	-0.23	0.42	0.19	-0.26	-0.25	-0.27
DOC	-0.77	0.29	-0.41	0.50	-0.68	0.70	-0.46
DIC	0.61	-0.53	0.39	-0.20	0.44	-0.75	0.26
Na	-0.21	-0.16	0.21	0.37	-0.36	-0.07	-0.39
Mg	0.34	-0.63	0.52	0.06	0.10	-0.62	0.10
Al	-0.82	0.56	-0.55	0.35	-0.58	0.85	-0.52
Si	0.26	-0.56	0.16	0.15	0.59	-0.42	0.04
P	-0.78	0.26	-0.38	0.59	-0.56	0.62	-0.44
K	-0.35	-0.03	0.07	0.57	-0.41	0.01	-0.27
Ca	0.76	-0.66	0.49	-0.31	0.62	-0.87	0.49
Ti	-0.82	0.37	-0.48	0.53	-0.62	0.75	-0.49
Cr	-0.69	0.28	-0.66	0.38	-0.43	0.71	-0.37
Mn	-0.67	0.28	-0.47	0.62	-0.65	0.58	-0.57
Fe	-0.85	0.45	-0.53	0.55	-0.64	0.80	-0.50
Co	-0.65	0.15	-0.33	0.50	-0.49	0.45	-0.64
Ni	-0.81	0.27	-0.43	0.57	-0.58	0.65	-0.46
Cu	-0.78	0.08	-0.49	0.74	-0.37	0.45	-0.43
Zn	-0.37	0.21	-0.16	0.51	-0.18	0.08	-0.37
Ge	-0.52	-0.20	-0.30	0.60	-0.17	0.22	-0.51
Rb	-0.81	0.22	-0.41	0.54	-0.62	0.68	-0.45
Sr	0.69	-0.75	0.56	-0.24	0.50	-0.85	0.40
Y	-0.83	0.37	-0.47	0.55	-0.61	0.74	-0.51
Zr	-0.84	0.41	-0.49	0.56	-0.65	0.76	-0.51
Nb	-0.82	0.35	-0.51	0.56	-0.65	0.73	-0.54
Mo	0.77	-0.65	0.49	-0.38	0.64	-0.84	0.46
Cd	-0.39	0.11	-0.16	0.40	-0.19	0.08	-0.61
Sb	0.46	-0.48	0.37	-0.01	0.49	-0.74	0.10
Cs	-0.66	0.42	-0.29	0.39	-0.60	0.51	-0.52
Ba	0.29	-0.73	0.38	0.13	0.18	-0.54	0.22
La	-0.83	0.40	-0.48	0.54	-0.62	0.75	-0.49
Ce	-0.85	0.44	-0.50	0.57	-0.63	0.77	-0.50
Pr	-0.83	0.39	-0.47	0.54	-0.61	0.75	-0.50
Nd	-0.84	0.41	-0.49	0.56	-0.63	0.76	-0.48
Sm	-0.82	0.35	-0.46	0.55	-0.62	0.74	-0.49
Eu	-0.82	0.34	-0.45	0.54	-0.62	0.72	-0.49
Gd	-0.84	0.39	-0.48	0.56	-0.61	0.76	-0.52
Tb	-0.84	0.39	-0.48	0.55	-0.62	0.75	-0.50
Dy	-0.82	0.38	-0.47	0.54	-0.63	0.75	-0.50
Ho	-0.83	0.39	-0.47	0.55	-0.62	0.75	-0.52
Er	-0.83	0.35	-0.46	0.55	-0.62	0.74	-0.50
Tm	-0.84	0.41	-0.47	0.55	-0.63	0.76	-0.52
Yb	-0.83	0.39	-0.48	0.55	-0.62	0.75	-0.51
Lu	-0.84	0.38	-0.49	0.56	-0.63	0.76	-0.52
Hf	-0.80	0.50	-0.42	0.42	-0.67	0.78	-0.48
W	0.61	-0.44	0.28	-0.33	0.56	-0.66	0.33
Pb	-0.75	0.29	-0.56	0.60	-0.44	0.57	-0.63
Th	-0.82	0.38	-0.47	0.53	-0.67	0.75	-0.52
U	0.78	-0.63	0.45	-0.40	0.68	-0.83	0.51

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Table S4. Proportion of colloidal fraction of elements in the Ob River downstream of Irtysh (4 points) and 7 tributaries. Elements not listed in this table (Li, B, Si, Cl, DIC…) exhibited less than 5-10 % of colloids in all studied samples.

	p-1 (Pasay-deyakha)	2 (Ob′)	p-2 (Poluy)	p-3 (Pitljar)	8 (Ob′)	10 (Ob′)	11 (Ob′)	p-4 (Irtysh)	p-9 (Ket')	p-12 (Tom')
DOC	43	43	45	36	41	45	42	42	12	35
Al	86	95	93	97	98	97	94	79	62	38
P	98	98	87	94	84	85	93	74	41	78
Ca	nd	7	11	5	5	2	4	4	0	0
Ti	71	73	79	78	74	79	78	60	34	9
V	76	74	68	58	65	61	64	36	11	11
Cr	60	54	61	59	46	55	64	15	27	nd
Mn	99	92	99	100	100	83	96	99	23	97
Fe	99	99	99	99	99	98	98	96	74	48
Co	92	68	84	69	65	67	64	85	15	47
Ni	41	51	76	54	49	51	47	46	31	14
Cu	47	66	59	64	56	73	67	55	nd	nd
Zn	92	nd	96	67	83	77	47	92	nd	nd
Ga	86	nd	nd	87	8	68	84	85	47	12
Ge	14	0	11	13	0	12	1	16	nd	0
As	56	43	37	39	47	44	36	22	9	0
Y	97	96	96	95	95	95	94	89	76	73
Zr	99	96	99	98	98	98	98	95	93	89
Nb	nd	51	84	96	96	94	89	60	23	nd
Mo	8	1	0	4	10	1	6	4	0	0
Cd	78	nd	85	38	43	nd	25	29	nd	nd
Sb	0	0	21	26	19	16	12	17	0	2
Cs	nd	<10	36	43	54	46	33	13	nd	nd
Ba	0	17	35	12	14	13	12	13	0	1
La	99	95	97	97	97	96	95	91	4	20
Ce	99	97	98	99	99	99	98	97	71	63
Pr	99	98	98	98	98	98	97	95	86	88
Nd	99	98	98	98	97	98	97	94	78	84
Sm	98	97	96	97	98	96	95	96	80	92
Eu	97	91	97	85	85	88	88	82	52	23
Gd	99	97	97	97	97	97	96	94	68	80
Tb	99	97	97	97	95	97	97	94	76	77
Dy	98	97	97	97	96	96	96	95	78	86
Ho	98	98	97	97	97	95	96	90	85	89
Er	97	97	95	96	95	96	96	91	69	61
Tm	96	94	92	95	97	99	92	86	78	51
Yb	96	96	95	96	94	95	91	89	54	74
Lu	93	98	94	92	93	92	90	84	69	45
Hf	98	97	99	92	88	94	96	88	68	75
W	nd	0	43	36	48	37	17	19	4	3
Tl	44	0	67	30	26	11	0	nd	5	13
Pb	99	98	99	100	100	99	96	95	25	nd
Th	94	99	96	98	97	98	98	97	86	98
U	87	70	91	53	56	46	44	25	3	3