Supplemental text

The conceptual model presented in Figure 5 considers a suite of drivers of change that are prevalent throughout the pan-Arctic watershed, and current literature evidence for their effect on each of our focal constituents. Though we focus on large-scale drivers with known impact, we acknowledge that this conceptual exercise is not exhaustive (see also *Additional drivers*, below). We also acknowledge that evidence for some of the relationships we explore is relatively poorly developed. Below, we review the literature underpinnings of this conceptual exercise, highlighting uncertainty where appropriate.

Active layer deepening: Here, we consider deepening of the seasonally thawed active layer into the top of the permafrost table, and the lateral expansion of thaw in regions of discontinuous permafrost. Deepening and expansion of thaw enables flow through deeper flow paths within soils, and increased transport of super- and sub-permafrost groundwater to riverine networks[74]. As a result, the effect of active layer deepening on riverine chemistry will depend on soil composition, and particularly whether changes in flow path enable increased water-soil interaction with mineral-rich soils at depth[26,27]. Because of variation in soil profiles throughout the pan-Arctic, the net result of active layer deepening on *dissolved organic carbon (DOC)* within aquatic networks is expected to be location-specific and mixed[35], with documented negative relationships between active layer depth and stream DOC concentrations in regions with mineral soils[75]. In contrast, active layer deepening is expected to increase weathering and/or *alkalinity* transport to aquatic networks via a variety of processes. These include: increasing contributions from ion-rich groundwater, possible increases in soil-water interaction via increasing within-catchment residence time, and flow through deeper soils in regions where mineral soils predominate[76-79]. Deeper active layers associated with soil warming have been associated with increased dissolved inorganic nitrogen in soils[80], while active layer depth has been shown to correlate with increased stream water *nitrate* concentrations[75]. Increasing groundwater contributions have also been hypothesized to increase nitrate transport to surface waters[77]. Thus, we assess that active layer deepening in absence of other factors is likely to increase nitrate within aquatic networks.

Thermokarst: At sites or in regions where ground ice (i.e., water frozen in permafrost in excess of the saturated water content in the thawed state) is prevalent, permafrost thaw causes thermokarst, or landscape collapse, to occur[33]. Because ground ice is patchily distributed throughout the Arctic (for example, see ref. [81] for a recent assessment for Canada), thermokarst is a regionally-distributed process[82]. Over flat terrain, thermokarst is most likely to enable the creation and expansion of lakes, ponds, and wetlands, with modest effects on fluvial networks[33]. Therefore, we focus our assessment on documented effects of hillslope thermokarst. Similar to active layer deepening, the effects of thermokarst on *DOC* transport to aquatic networks will depend on soil composition, with increases in DOC concentration observed in some thermokarst-affected systems[40,83], but decreases observed in others[34,84]. Particularly in regions where thermokarst exposes mineral soils, we expect *alkalinity* concentrations to increase in fluvial networks following thermokarst[85], though changes should be muted to neutral where permafrost is comprised of organic-rich or well-weathered sediments. Wide-ranging reviews[29,86] find that thermokarst generally increases delivery of inorganic nutrients (including *nitrate*) to aquatic systems at the site of thaw.

Shrubification: The northward expansion of shrubs into tundra landscapes (also referred to as greening) is a widespread response to climate change in the north[87]. The advance of northern and mountainous treelines has been documented in several studies to increase DOC concentrations in affected freshwater systems, including via long-term paleolimnological assessments[41,88]. As a result, we assess that shrubification will increase *DOC* transport to fluvial networks, following increases in organic matter biomass and litter layers[87]. The expansion of land plants, and resultant increases in root respiration and thus carbonic acid production, is widely understood to increase the rate of weathering within soils[38], thus increasing *alkalinity* production. Expansion of woody vegetation is expected to increase uptake of inorganic nitrogen, and nitrogen sequestration in tissue[44,80], with some evidence that temperature and CO_2-induced increases in growth rate may be outpacing concomitant increases in nitrogen mineralization, and thus causing increased N limitation in shrubs[89]. Thus, we expect shrubification to decrease *nitrate* availability for transport to fluvial networks.

Increasing microbial processing: Here, we consider temperature-induced changes in processing and uptake rates by autotrophic and heterotrophic microbes, in soils and soil pore waters (i.e., the hydrologic flow paths that transport waterborne constituents), and in aquatic networks. In soils, warming augments both the generation and microbial mineralization of *DOC*. However, experimental work suggests that the Q_{10} for within-soil DOC mineralization is greater than that for DOC generation[42]. Similarly, DOC mineralization is typically understood to increase with increasing temperature within aquatic networks[18,90]. Therefore, we assess that temperature-induced increases in microbial processing are likely to decrease DOC within fluvial networks. Increasing pCO_2 in soil pore waters following temperature-induced increases in soil respiration will increase *alkalinity* generation, similar to the effect of increasing pore water pCO_2 following atmospheric enrichment[39,91,92].

In soils and pore waters, increasing temperature has been shown to increase *nitrate* uptake by microbes (i.e., immobilization)[93], and its loss via denitrification[17]. However, these changes occur alongside increasing rates of transformation throughout the nitrogen cycle[44]: a recent meta-analysis suggests that co-occurring increases in ammoniafication and subsequent nitrification lead to no net change in soil nitrate concentrations with warming in northern soils[17]. Such holistic work has been more limited in freshwater systems: laboratory experiments have shown increasing nitrate uptake by microbes with warming[16,94]; *in situ* experimentation has shown increased uptake of inorganic nitrogen under warmer conditions[94,95]; and incubation[96] and meta-analytic[97] work suggests increasing rates of denitrification with increasing temperature, with an effect that may be particularly pronounced in fluvial headwaters[96]. Although the increases in organic matter respiration expected with increasing temperature may augment nitrogen mineralization (i.e., ammonification) (see, for example, ref. [98] for Arctic coastal sediments), the relatively high C:N ratios for organic matter in these large rivers (particularly in the dissolved phase)[54,64] seem likely to result in tight nitrogen cycling and overall immobilization[99,100]. Thus, although current evidence is not robust, we hypothesize that increasing microbial processing will cause a net decline in nitrate within aquatic networks, all else being equal.

Additional drivers of change: In addition to the key drivers of change considered in Figure 5 and above, we acknowledge that several additional factors may be regionally important, or engender change beyond the specific timespan of our assessment. Here, we consider river impoundment, land use and land cover change, and declining rates of acid deposition (sulfate, nitrate) as additional drivers of change.

Impoundment: Impoundments (i.e., dams) occur throughout the pan-Arctic watershed, with particular prevalence in Russia (Extended Data Table 1). Although the majority of impoundments in our study region were constructed and filled prior to the onset of our data collection[21], new structures did come online more recently in the Yenisey (2012) and Kolyma (2013) watersheds (Extended Data Table 1). Considered seasonally, one key effect of impoundment is that reservoirs collect water throughout the year, but provide discharge at discreet time points to meet agricultural or hydroelectric needs. As a result, riverine impoundment will change the seasonal concentration profile of biogeochemical constituents relative to the un-regulated state[101]. Across seasons, impoundments are known to enable the release of *DOC* from newly flooded sediments; an effect that declines with time since innundation[102]. However, meta-analytic approaches suggest that the increasing water residence time that results from riverine impoundment will decrease DOC in oligotrophic to mesotrophic systems, but increase DOC in eutrophic systems[103]. Thus, although the effect of impoundment on downstream DOC appears to depend on both reservoir age and trophic status, in general we expect impoundment to decrease DOC concentrations in most Arctic watersheds, compared to the pre-impoundment state. We do not expect impoundments to have a major effect on *alkalinity*, although some transformations within the inorganic carbon cycle are possible following CO_2 generation by organic carbon mineralization, particularly when captured in the bottom waters of stratified, hypolimnetic release, reservoirs. Meta-analyses and models[20] indicate that impoundments should act as a net sink for both *nitrate* and total nitrogen, with increasing residence time leading to increases in denitrification[104] and nitrogen burial[20].

Declining deposition of SO_4 and NO_3^-: Sulfate deposition has declined throughout Europe and North America over the past 30 years, with this declining trend ongoing throughout the ArcticGRO data collection period[24]. On a relative basis, modelled declines appear to range to as high as ~2% y^{-1} across our study catchments, with effects that are greatest in the Ob' basin, which is most influenced by the pronounced declines in SO_4 emissions throughout western Europe[24]. Inorganic N deposition has also been modelled to be declining over recent decades in the western Russian watersheds (Ob', Yenisey, modelled declines to ~1% y^{-1}; 1984-2016), but in this case models indicate little change in N deposition for the more easterly Russian catchments (Lena, Kolyma) and the Yukon watershed, and a possible increase in N deposition in the Mackenzie[105]. These changes can be expected to affect each of our focal constituents. Declining acid deposition has been broadly associated with increased *DOC* transport to inland waters[106]. In these reasonably well buffered watersheds (riverine pH of 7 or greater), declining acid deposition should also be expected to decrease *alkalinity* generation via the effects of declining acidity on weathering reactions in soils[107]; this effect contrasts with the expectation for poorly buffered regions, where declining acidity will allow soils to recover from acid-induced denudation[108], or be less subject to acid-induced reactions within the inorganic carbon system that generate CO_2 at the expense of alkalinity[107]. Where relevant, declines in inorganic N deposition could contribute to the *nitrate* trends that we observe.

Land use and land cover change: Recent Landsat-based assessments of land use and land cover change (2000-2020) indicate a decline in cropland cover through the southern reaches of the Ob' and Yenisey watersheds, and a potential decline in forest cover throughout several of the ArcticGRO catchments (defined as the Landsat-assessed loss of vegetation > 5m height)[109]. Declines in cropland extent could contribute to the *nitrate* trends that we observe. The effects of vegetation loss, which could arise following deforestation or disturbance (wildfire, insects) are more difficult to assess, because they will depend on whether vegetation has been removed or is able to decay in place, and effects on soils (e.g., the degree to which combustion of organic soil layers has occurred following wildfire). However, to some extent, this change may counteract some of the effects described above for shrubification.

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Table S1: Statistical outputs for Mann-Kendall trend analyses of changes in constituent flux and discharge over time. Unit indicates the unit of the trend.

River	Constituent	Unit	Kendall statistic	Variance	Z	P	Median annual Sen slope	5% confidence limit	95% confidence limit
Kolyma	Alkalinity	Gg $CaCO_3$ y^{-1}	30	589.3333	1.1946	0.2322	39.8713	-22.29	110.90
Kolyma	DOC	Gg y^{-1}	10	589.3333	0.3707	0.7108	3.5194	-23.44	37.91
Kolyma	NO_3	Mg N y^{-1}	-48	589.3333	-1.9361	0.0529	-164.6957	-338.83	15.19
Kolyma	NH_4	Mg N y^{-1}	72	589.3333	2.9247	0.0034	128.2589	40.36	290.04
Kolyma	TDP	Mg P y^{-1}	16	589.3333	0.6179	0.5366	35.5396	-45.41	92.12
Kolyma	SiO_2	Gg SiO_2 y^{-1}	-32	589.3333	-1.2770	0.2016	-6.8435	-19.98	3.78
Kolyma	Na	Gg y^{-1}	22	589.3333	0.8650	0.3870	1.3580	-2.85	6.16
Kolyma	K	Gg y^{-1}	15	588.3333	0.5772	0.5638	0.3000	-1.16	2.55
Kolyma	Mg	Gg y^{-1}	48	589.3333	1.9361	0.0529	6.7383	-0.48	14.93
Kolyma	Ca	Gg y^{-1}	28	589.3333	1.1122	0.2661	15.6613	-19.49	50.07
Kolyma	Ba	Mg y^{-1}	40	589.3333	1.6065	0.1082	22.8355	-8.26	52.28
Kolyma	Cl	Gg y^{-1}	-32	589.3333	-1.2770	0.2016	-0.4283	-1.22	0.32
Kolyma	SO_4	Gg S y^{-1}	92	589.3333	3.7485	0.0002	24.4801	14.50	39.06
Kolyma	Li	Mg y^{-1}	50	589.3333	2.0184	0.0435	1.0589	0.07	2.37
Kolyma	Rb	Mg y^{-1}	46	589.3333	1.8537	0.0638	0.5023	-0.08	1.54
Kolyma	Sr	Mg y^{-1}	38	589.3333	1.5241	0.1275	148.0470	-49.32	351.29
Kolyma	U	Mg y^{-1}	5	588.3333	0.1649	0.8690	0.0114	-0.08	0.15
Lena	Alkalinity	Gg $CaCO_3$ y^{-1}	54	589.3333	2.1832	0.0290	440.1695	119.39	806.44
Lena	DOC	Gg y^{-1}	-14	589.3333	-0.5355	0.5923	-27.7115	-131.38	91.24
Lena	NO_3	Mg N y^{-1}	-98	589.3333	-3.9957	0.0001	-882.9091	-1119.87	-560.03
Lena	NH_4	Mg N y^{-1}	-42	589.3333	-1.6889	0.0912	-171.8580	-381.33	72.16
Lena	TDP	Mg P y^{-1}	-16	589.3333	-0.6179	0.5366	-102.1435	-277.62	232.80
Lena	SiO_2	Gg SiO_2 y^{-1}	-20	589.3333	-0.7827	0.4338	-25.1583	-57.22	32.75
Lena	Na	Gg y^{-1}	80	589.3333	3.2542	0.0011	143.9130	48.51	225.09
Lena	K	Gg y^{-1}	40	589.3333	1.6065	0.1082	3.8205	-1.72	9.16
Lena	Mg	Gg y^{-1}	30	589.3333	1.1946	0.2322	19.8720	-15.88	61.99
Lena	Ca	Gg y^{-1}	32	589.3333	1.2770	0.2016	74.6456	-37.04	235.01
Lena	Ba	Mg y^{-1}	30	589.3333	1.1946	0.2322	55.5017	-59.06	195.03
Lena	Cl	Gg y^{-1}	-20	589.3333	-0.7827	0.4338	-41.6492	-162.98	99.17
Lena	SO_4	Gg S y^{-1}	26	589.3333	1.0298	0.3031	18.4017	-12.84	55.46
Lena	Li	Mg y^{-1}	80	589.3333	3.2542	0.0011	28.0152	13.86	40.43
Lena	Rb	Mg y^{-1}	58	589.3333	2.3480	0.0189	4.0725	0.49	8.55
Lena	Sr	Mg y^{-1}	60	589.3333	2.4304	0.0151	1030.2855	226.49	1911.49
Lena	U	Mg y^{-1}	48	589.3333	1.9361	0.0529	1.6049	-0.06	3.93
Mackenzie	Alkalinity	Gg $CaCO_3$ y^{-1}	-16	589.3333	-0.6179	0.5366	-50.5888	-374.32	178.71

Mackenzie	DOC	Gg y^{-1}	-30	589.3333	-1.1946	0.2322	-12.1595	-32.80	10.05
Mackenzie	NO_3	Mg N y^{-1}	-98	589.3333	-3.9957	0.0001	-661.4835	-803.69	-446.45
Mackenzie	NH_4	Mg N y^{-1}	-52	589.3333	-2.1008	0.0357	-62.3808	-105.76	-2.81
Mackenzie	SiO_2	Gg SiO_2 y^{-1}	-94	589.3333	-3.8309	0.0001	-24.2130	-33.66	-16.75
Mackenzie	Na	Gg y^{-1}	-30	589.3333	-1.1946	0.2322	-8.6012	-26.71	9.02
Mackenzie	K	Gg y^{-1}	-52	589.3333	-2.1008	0.0357	-3.6990	-5.93	-0.60
Mackenzie	Mg	Gg y^{-1}	-32	589.3333	-1.2770	0.2016	-15.8908	-36.43	13.95
Mackenzie	Ca	Gg y^{-1}	-14	589.3333	-0.5355	0.5923	-31.9075	-148.62	87.52
Mackenzie	Ba	Mg y^{-1}	-38	589.3333	-1.5241	0.1275	-105.1217	-301.74	27.40
Mackenzie	Cl	Gg y^{-1}	-102	589.3333	-4.1605	0.0000	-72.2270	-92.31	-51.43
Mackenzie	SO_4	Gg S y^{-1}	-70	589.3333	-2.8423	0.0045	-65.1810	-106.11	-18.62
Mackenzie	Li	Mg y^{-1}	10	589.3333	0.3707	0.7108	5.5788	-12.59	25.11
Mackenzie	Rb	Mg y^{-1}	-26	589.3333	-1.0298	0.3031	-0.9014	-2.95	1.12
Mackenzie	Sr	Mg y^{-1}	34	589.3333	1.3594	0.1740	382.6188	-237.00	1262.48
Mackenzie	U	Mg y^{-1}	44	589.3333	1.7713	0.0765	1.4746	-0.31	3.81
Ob’	Alkalinity	Gg $CaCO_3$ y^{-1}	88	589.3333	3.5838	0.0003	592.8575	353.25	804.30
Ob’	DOC	Gg y^{-1}	60	589.3333	2.4304	0.0151	81.1150	25.39	150.20
Ob’	NO_3	Mg N y^{-1}	10	589.3333	0.3707	0.7108	431.5385	-1273.26	1712.66
Ob’	NH_4	Mg N y^{-1}	-52	589.3333	-2.1008	0.0357	-1108.2804	-2105.09	-152.14
Ob’	TDP	Mg P y^{-1}	64	589.3333	2.5951	0.0095	611.8149	236.09	1112.15
Ob’	SiO_2	Gg SiO_2 y^{-1}	34	589.3333	1.3594	0.1740	41.0642	-13.25	65.36
Ob’	Na	Gg y^{-1}	70	589.3333	2.8423	0.0045	62.6068	18.41	98.73
Ob’	K	Gg y^{-1}	32	589.3333	1.2770	0.2016	4.3713	-4.01	9.60
Ob’	Mg	Gg y^{-1}	58	589.3333	2.3480	0.0189	18.5398	3.17	42.72
Ob’	Ca	Gg y^{-1}	74	589.3333	3.0071	0.0026	142.8552	58.45	219.21
Ob’	Ba	Mg y^{-1}	24	589.3333	0.9474	0.3434	47.8111	-73.20	130.88
Ob’	Cl	Gg y^{-1}	14	589.3333	0.5355	0.5923	14.3796	-37.62	50.04
Ob’	SO_4	Gg S y^{-1}	40	589.3333	1.6065	0.1082	7.7553	-3.00	19.88
Ob’	Li	Mg y^{-1}	68	589.3333	2.7599	0.0058	14.0346	6.36	25.21
Ob’	Rb	Mg y^{-1}	44	589.3333	1.7713	0.0765	7.5795	-0.87	13.03
Ob’	Sr	Mg y^{-1}	74	589.3333	3.0071	0.0026	839.7940	301.12	1252.90
Ob’	U	Mg y^{-1}	110	589.3333	4.4900	0.0000	2.7831	2.19	3.83
Yenisey	Alkalinity	Gg $CaCO_3$ y^{-1}	20	589.3333	0.7827	0.4338	93.7826	-176.92	451.71
Yenisey	DOC	Gg y^{-1}	-58	589.3333	-2.3480	0.0189	-74.2046	-147.78	-20.09
Yenisey	NO_3	Mg N y^{-1}	-80	589.3333	-3.2542	0.0011	-2148.0166	-3190.24	-1152.28
Yenisey	NH_4	Mg N y^{-1}	48	589.3333	1.9361	0.0529	341.2729	-15.85	666.41
Yenisey	TDP	Mg P y^{-1}	-84	589.3333	-3.4190	0.0006	-342.8054	-454.94	-246.73
Yenisey	SiO_2	Gg SiO_2 y^{-1}	-62	589.3333	-2.5128	0.0120	-49.1961	-83.32	-16.69
Yenisey	Na	Gg y^{-1}	-70	589.3333	-2.8423	0.0045	-50.2415	-78.14	-25.30
Yenisey	K	Gg y^{-1}	-84	589.3333	-3.4190	0.0006	-6.1506	-9.63	-4.53
Yenisey	Mg	Gg y^{-1}	-52	589.3333	-2.1008	0.0357	-21.3139	-43.29	-2.45
Yenisey	Ca	Gg y^{-1}	-66	589.3333	-2.6775	0.0074	-132.5925	-215.03	-36.94
Yenisey	Ba	Mg y^{-1}	-48	589.3333	-1.9361	0.0529	-55.6277	-117.51	0.88
Yenisey	Cl	Gg y^{-1}	-100	589.3333	-4.0781	0.0000	-135.0972	-175.87	-97.12

Yenisey	SO_4	Gg S y^{-1}	-74	589.3333	-3.0071	0.0026	-32.0800	-44.98	-18.40
Yenisey	Li	Mg y^{-1}	-64	589.3333	-2.5951	0.0095	-11.3557	-16.20	-4.22
Yenisey	Rb	Mg y^{-1}	-70	589.3333	-2.8423	0.0045	-2.7934	-4.75	-1.25
Yenisey	Sr	Mg y^{-1}	-36	589.3333	-1.4417	0.1494	-546.9151	-1138.92	211.64
Yenisey	U	Mg y^{-1}	0	589.3333	0.0000	1.0000	0.0426	-1.49	1.44
Yukon	Alkalinity	Gg $CaCO_3$ y^{-1}	46	589.3333	1.8537	0.0638	124.7965	-10.12	268.00
Yukon	DOC	Gg y^{-1}	-6	589.3333	-0.2060	0.8368	-4.4282	-36.00	23.51
Yukon	NO_3	Mg N y^{-1}	38	589.3333	1.5241	0.1275	192.1561	-46.48	334.41
Yukon	NH_4	Mg N y^{-1}	-20	589.3333	-0.7827	0.4338	-43.3041	-138.11	47.85
Yukon	TDP	Mg P y^{-1}	44	589.3333	1.7713	0.0765	37.4515	-4.95	77.01
Yukon	SiO_2	Gg SiO_2 y^{-1}	-20	589.3333	-0.7827	0.4338	-4.6467	-20.32	13.87
Yukon	Na	Gg y^{-1}	78	589.3333	3.1718	0.0015	7.3132	3.75	11.39
Yukon	K	Gg y^{-1}	-34	589.3333	-1.3594	0.1740	-1.3854	-3.68	0.58
Yukon	Mg	Gg y^{-1}	88	589.3333	3.5838	0.0003	31.0735	16.59	45.85
Yukon	Ca	Gg y^{-1}	34	589.3333	1.3594	0.1740	48.3538	-15.66	100.65
Yukon	Ba	Mg y^{-1}	32	589.3333	1.2770	0.2016	67.6906	-38.81	159.96
Yukon	Cl	Gg y^{-1}	-106	589.3333	-4.3252	0.0000	-4.4980	-6.53	-3.09
Yukon	SO_4	Gg S y^{-1}	92	589.3333	3.7485	0.0002	70.3203	39.88	100.10
Yukon	Li	Mg y^{-1}	46	589.3333	1.8537	0.0638	5.7331	-0.23	9.97
Yukon	Sr	Mg y^{-1}	68	589.3333	2.7599	0.0058	394.0563	176.73	674.40
Yukon	U	Mg y^{-1}	72	589.3333	2.9247	0.0034	2.1793	0.79	3.54
Sum	Alkalinity	Gg $CaCO_3$ y^{-1}	68	589.3333	2.7599	0.0058	1013.9760	291.91	1967.15
Sum	DOC	Gg y^{-1}	2	589.3333	0.0412	0.9671	8.2236	-239.79	191.17
Sum	NO_3	Mg N y^{-1}	-74	589.3333	-3.0071	0.0026	-3535.6744	-5715.49	-1724.49
Sum	NH_4	Mg N y^{-1}	-64	589.3333	-2.5951	0.0095	-1086.0411	-1968.22	-385.05
Sum	SiO_2	Gg SiO_2 y^{-1}	-34	589.3333	-1.3594	0.1740	-72.3286	-150.40	35.70
Sum	Na	Gg y^{-1}	64	589.3333	2.5951	0.0095	171.0866	39.22	263.13
Sum	K	Gg y^{-1}	-10	589.3333	-0.3707	0.7108	-1.4618	-15.97	10.09
Sum	Mg	Gg y^{-1}	22	589.3333	0.8650	0.3870	35.6633	-33.99	127.13
Sum	Ca	Gg y^{-1}	14	589.3333	0.5355	0.5923	56.1090	-246.43	400.18
Sum	Ba	Mg y^{-1}	-4	589.3333	-0.1236	0.9016	-24.2500	-280.70	350.78
Sum	Cl	Gg y^{-1}	-54	589.3333	-2.1832	0.0290	-222.8720	-422.44	-34.16
Sum	SO_4	Gg S y^{-1}	16	589.3333	0.6179	0.5366	26.2374	-52.70	105.15
Sum	Li	Mg y^{-1}	58	589.3333	2.3480	0.0189	39.4350	7.67	68.34
Sum	Rb	Mg y^{-1}	46	589.3333	1.8537	0.0638	10.9305	-2.40	20.56
Sum	Sr	Mg y^{-1}	64	589.3333	2.5951	0.0095	2229.1793	552.02	3952.05
Sum	U	Mg y^{-1}	76	589.3333	3.0894	0.0020	9.3455	4.39	13.60
Ob’	Discharge	km^3 y^{-1}	48	589.3333	1.9361	0.0529	172.7885	-0.33	293.00
Yenisey	Discharge	km^3 y^{-1}	-42	589.3333	-1.6889	0.0912	-148.3167	-366.11	65.42
Lena	Discharge	km^3 y^{-1}	-4	589.3333	-0.1236	0.9016	-28.6771	-276.00	275.17
Kolyma	Discharge	km^3 y^{-1}	8	589.3333	0.2883	0.7731	8.0938	-70.45	103.90
Yukon	Discharge	km^3 y^{-1}	34	589.3333	1.3594	0.1740	48.3500	-24.17	108.89
Mackenzie	Discharge	km^3 y^{-1}	-56	589.3333	-2.2656	0.0235	-135.2500	-189.00	-33.00

Table S2: Statistical outputs for seasonal Mann-Kendall trend analyses of changes in concentration over time. Units indicate the input unit, and correspond to the median value. Sen's slope is provided as units y^{-1}.

River	Constituent	Unit	N	Median value	Kendall statistic	Variance	Z	P	Median annual Sen slope	5% confidence limit	95% confidence limit	R^2 (Q adjusted)	Method (Q adjusted)
Kolyma	Ca	mg L^{-1}	74	13.99	192	5625.3	2.5466	0.0109	0.1375	0.0553	0.2493	0.68	log-log
Kolyma	Mg	mg L^{-1}	74	3.30	354	5625.3	4.7065	0.0000	0.0847	0.0527	0.1095	0.57	log-log
Kolyma	Alkalinity	mg $CaCO_3$ L^{-1}	67	28.42	169	4155.7	2.6061	0.0092	0.3921	0.1243	0.6652	0.62	log-log
Kolyma	Li	µg L^{-1}	65	0.49	195	3684.3	3.1961	0.0014	0.0091	0.0052	0.0149	0.43	log-log
Kolyma	U	µg L^{-1}	58	0.04	-35	2597.0	-0.6672	0.5047	-0.0002	-0.001	0.0002	0.02	log-log
Kolyma	Ba	µg L^{-1}	65	11.79	262	3683.3	4.3005	0.0000	0.2703	0.1909	0.3495	0.72	log-log
Kolyma	Sr	µg L^{-1}	65	87.50	225	3684.3	3.6904	0.0002	1.601	0.8978	2.2679	0.67	log-log
Kolyma	SO_4	mg S L^{-1}	66	7.31	332	4048.0	5.2024	0.0000	0.3849	0.2625	0.4943	0.39	log-log
Kolyma	Rb	µg L^{-1}	65	0.24	149	3684.3	2.4383	0.0148	0.0034	0.0006	0.0071	0.29	log-log
Kolyma	K	mg L^{-1}	73	0.55	24	5417.3	0.3125	0.7547	0.0009	-0.002	0.0036	0.01	log-log
Kolyma	NH_4	µg N L^{-1}	74	13.80	88	5564.0	1.1663	0.2435	0.3198	-0.1219	0.6899	0.03	log-log
Kolyma	SiO_2	mg SiO_2 L^{-1}	75	5.97	-220	5772.0	-2.8826	0.0039	-0.0742	-0.1098	-0.031	0.51	log-log
Kolyma	NO_3	µg N L^{-1}	73	40.08	-88	5284.0	-1.1968	0.2314	-0.7226	-1.8171	0.2698	0.61	gam
Kolyma	Na	mg L^{-1}	74	2.03	224	5625.3	2.9732	0.0029	0.0242	0.0107	0.0341	0.63	log-log
Kolyma	Cl	mg L^{-1}	65	0.28	-188	3905.3	-2.9924	0.0028	-0.0077	-0.0139	-0.0037	0.38	gam
Kolyma	TDP	µg L^{-1}	73	6.50	-151	5416.3	-2.0382	0.0415	-0.1963	-0.348	-0.0386	0.07	log-log
Kolyma	DOC	mg L^{-1}	74	4.28	10	7740.0	0.1023	0.9185	0.0083	-0.0809	0.0681	0.45	log-log
Lena	Ca	mg L^{-1}	74	18.65	166	5456.0	2.2338	0.0255	0.1935	0.044	0.379	0.79	log-log
Lena	Mg	mg L^{-1}	74	5.40	234	5456.0	3.1544	0.0016	0.0925	0.0364	0.1372	0.76	log-log
Lena	Alkalinity	mg $CaCO_3$ L^{-1}	75	50.59	317	5617.0	4.2163	0.0000	0.9058	0.5254	1.3885	0.78	log-log
Lena	Li	µg L^{-1}	65	1.94	149	4075.0	2.3185	0.0204	0.0377	0.0077	0.0683	0.43	log-log
Lena	U	µg L^{-1}	72	0.28	56	5066.7	0.7727	0.4397	0.002	-0.0023	0.0067	0.64	log-log
Lena	Ba	µg L^{-1}	72	16.06	166	5066.7	2.318	0.0204	0.1964	0.0629	0.3213	0.77	log-log
Lena	Sr	µg L^{-1}	72	140.66	202	5066.7	2.8238	0.0047	2.6145	0.9507	4.1155	0.79	log-log
Lena	SO_4	mg S L^{-1}	73	4.87	86	5149.3	1.1845	0.2362	0.0396	-0.0191	0.1048	0.74	log-log
Lena	Rb	µg L^{-1}	65	0.69	211	4075.0	3.2897	0.001	0.0098	0.0047	0.0143	0.19	log-log
Lena	K	mg L^{-1}	73	0.69	182	5213.3	2.5068	0.0122	0.0074	0.0029	0.0111	0.03	log-log
Lena	NH_4	µg N L^{-1}	75	15.90	-99	5617.0	-1.3076	0.191	-0.411	-0.8344	0.0709	0.05	log-log
Lena	SiO_2	mg SiO_2 L^{-1}	75	5.82	-23	5617.0	-0.2935	0.7691	-0.0079	-0.0533	0.0335	0.67	log-log
Lena	NO_3	µg N L^{-1}	75	30.58	-53	5617.0	-0.6938	0.4878	-0.5265	-1.548	0.7478	0.67	gam
Lena	Na	mg L^{-1}	73	11.86	216	5213.3	2.9777	0.0029	0.4161	0.1604	0.6265	0.77	log-log
Lena	Cl	mg L^{-1}	73	18.13	72	5149.3	0.9894	0.3225	0.2707	-0.1541	0.676	0.75	log-log
Lena	TDP	µg L^{-1}	75	6.99	-107	5617.0	-1.4143	0.1573	-0.1709	-0.3718	0.023	0.03	log-log
Lena	DOC	mg L^{-1}	75	7.84	-5	5617.0	-0.0534	0.9574	-0.0058	-0.0936	0.0831	0.3	log-log

Mackenzie	Ca	mg L^{-1}	76	38.75	167	6221.0	2.1046	0.0353	0.261	0.0583	0.3949	0.31	log-log
Mackenzie	Mg	mg L^{-1}	76	10.08	277	6221.0	3.4993	0.0005	0.0688	0.0357	0.0958	0.31	log-log
Mackenzie	Alkalinity	mg $CaCO_3$ L^{-1}	72	95.73	203	5212.3	2.7979	0.0051	0.474	0.2365	0.7903	0.58	log-log
Mackenzie	Li	µg L^{-1}	64	5.20	13	3967.0	0.1905	0.8489	0.0035	-0.0319	0.0253	0.52	log-log
Mackenzie	U	µg L^{-1}	73	0.72	147	5436.3	1.9802	0.0477	0.0063	0.0012	0.0111	0.05	log-log
Mackenzie	Ba	µg L^{-1}	73	54.62	-51	5436.3	-0.6781	0.4977	-0.0518	-0.2046	0.0692	0.03	log-log
Mackenzie	Sr	µg L^{-1}	73	244.13	238	5437.3	3.2141	0.0013	2.5252	1.414	3.4139	0.65	log-log
Mackenzie	SO_4	mg S L^{-1}	75	14.45	-13	5941.0	-0.1557	0.8763	-0.0128	-0.102	0.0889	0.1	log-log
Mackenzie	Rb	µg L^{-1}	63	0.64	5	3892.3	0.0641	0.9489	0	-0.0029	0.0029	0.01	log-log
Mackenzie	K	mg L^{-1}	76	0.93	42	6220.0	0.5199	0.6032	0.001	-0.0018	0.003	0.69	log-log
Mackenzie	NH_4	µg N L^{-1}	76	9.23	57	6221.0	0.71	0.4777	0.0472	-0.1616	0.3332	0.16	log-log
Mackenzie	SiO_2	mg SiO_2 L^{-1}	76	3.77	-341	6221.0	-4.3107	0	-0.0518	-0.0679	-0.0371	0.18	log-log
Mackenzie	NO_3	µg N L^{-1}	74	62.67	-347	5821.0	-4.535	0	-1.7728	-2.5634	-1.1382	0.63	log-log
Mackenzie	Na	mg L^{-1}	76	8.26	65	6221.0	0.8114	0.4171	0.0126	-0.0239	0.0394	0.88	log-log
Mackenzie	Cl	mg L^{-1}	75	8.91	-291	5941.0	-3.7624	0.0002	-0.2131	-0.2798	-0.119	0.73	log-log
Mackenzie	DOC	mg L^{-1}	75	4.57	109	5941.0	1.4012	0.1612	0.0301	-0.0073	0.0598	0.29	log-log
Ob'	Ca	mg L^{-1}	73	18.32	112	5230.0	1.5349	0.1248	0.1265	-0.0032	0.256	0.6	log-log
Ob'	Mg	mg L^{-1}	75	4.55	70	5616.0	0.9207	0.3572	0.0132	-0.0119	0.0347	0.69	log-log
Ob'	Alkalinity	mg $CaCO_3$ L^{-1}	73	59.97	222	5213.3	3.0608	0.0022	1.012	0.4994	1.4705	0.65	log-log
Ob'	Li	µg L^{-1}	64	2.65	75	4000.3	1.17	0.242	0.0092	-0.004	0.025	0.73	log-log
Ob'	U	µg L^{-1}	73	0.26	119	5212.3	1.6344	0.1022	0.0036	0	0.0081	0.57	gam
Ob'	Ba	µg L^{-1}	73	16.12	-2	5213.3	-0.0138	0.9889	-0.0021	-0.1823	0.1435	0.11	log-log
Ob'	Sr	µg L^{-1}	72	110.91	151	5065.7	2.1075	0.0351	1.044	0.2843	1.6905	0.66	log-log
Ob'	SO_4	mg S L^{-1}	71	2.35	-85	4762.3	-1.2172	0.2235	-0.0139	-0.0325	0.0083	0.3	log-log
Ob'	Rb	µg L^{-1}	66	0.74	129	4159.0	1.9848	0.0472	0.0102	0.0019	0.0216	0.05	log-log
Ob'	K	mg L^{-1}	76	1.07	-5	5793.0	-0.0526	0.9581	-0.0005	-0.0109	0.0092	0.03	log-log
Ob'	NH_4	µg N L^{-1}	75	28.78	135	5617.0	1.7879	0.0738	1.1809	0.0685	3.027	0.5	log-log
Ob'	SiO_2	mg SiO_2 L^{-1}	75	6.58	30	5616.0	0.387	0.6988	0.0201	-0.0607	0.1068	0.68	log-log
Ob'	NO_3	µg N L^{-1}	75	126.12	77	5617.0	1.0141	0.3106	1.6745	-1.0651	4.8461	0.37	gam
Ob'	Na	mg L^{-1}	74	7.25	242	5456.0	3.2627	0.0011	0.0899	0.0449	0.1469	0.76	log-log
Ob'	Cl	mg L^{-1}	72	5.32	27	4924.3	0.3705	0.711	0.0232	-0.0617	0.1069	0.61	log-log
Ob'	TDP	µg L^{-1}	75	39.79	20	5566.0	0.2547	0.799	0.1488	-0.781	1.0267	0.04	log-log
Ob'	DOC	mg L^{-1}	75	9.98	27	5615.0	0.347	0.7286	0.0174	-0.0665	0.1096	0.34	log-log
Yenisey	Ca	mg L^{-1}	72	20.65	-177	4924.3	-2.5081	0.0121	-0.1584	-0.2809	-0.0591	0.7	log-log
Yenisey	Mg	mg L^{-1}	72	4.67	-127	4892.3	-1.8014	0.0716	-0.0314	-0.0476	-0.004	0.78	log-log
Yenisey	Alkalinity	mg $CaCO_3$ L^{-1}	72	59.05	106	4941.3	1.4937	0.1353	0.2611	-0.0438	0.6603	0.74	log-log
Yenisey	Li	µg L^{-1}	61	1.76	-69	3275.7	-1.1881	0.2348	-0.0099	-0.0235	0.0032	0.59	log-log
Yenisey	U	µg L^{-1}	69	0.25	36	4347.3	0.5308	0.5955	0.001	-0.0022	0.004	0.47	log-log
Yenisey	Ba	µg L^{-1}	69	12.35	-99	4346.3	-1.4865	0.1371	-0.1091	-0.2131	0.0068	0.45	log-log
Yenisey	Sr	µg L^{-1}	69	169.77	-12	4347.3	-0.1668	0.8675	-0.0795	-1.1851	0.6425	0.73	log-log
Yenisey	SO_4	mg S L^{-1}	74	3.40	-196	5325.3	-2.6722	0.0075	-0.0404	-0.0649	-0.0178	0.68	log-log
Yenisey	Rb	µg L^{-1}	60	0.47	-31	3099.7	-0.5388	0.59	-0.0011	-0.0046	0.0027	0.28	log-log
Yenisey	K	mg L^{-1}	72	0.62	-169	4892.3	-2.4019	0.0163	-0.0074	-0.012	-0.003	0.4	log-log

Yenisey	NH_4	µg N L^{-1}	76	11.37	73	5793.0	0.946	0.3442	0.3292	-0.1757	0.6253	0.15	log-log
Yenisey	SiO_2	mg SiO_2 L^{-1}	76	5.99	-79	5793.0	-1.0248	0.3055	-0.0221	-0.0634	0.0156	0.42	log-log
Yenisey	NO_3	µg N L^{-1}	76	19.74	-41	5793.0	-0.5255	0.5992	-0.3638	-1.4121	1.0418	0.46	gam
Yenisey	Na	mg L^{-1}	73	7.72	-165	5100.3	-2.2964	0.0217	-0.0772	-0.1182	-0.0259	0.67	log-log
Yenisey	Cl	mg L^{-1}	74	9.67	-224	5325.3	-3.0558	0.0022	-0.1829	-0.2901	-0.0943	0.57	log-log
Yenisey	TDP	µg L^{-1}	75	11.48	-244	7688.0	-2.7714	0.0056	-0.3468	-0.5154	-0.1318	0.09	log-log
Yenisey	DOC	mg L^{-1}	74	5.36	-10	5392.0	-0.1226	0.9025	-0.0048	-0.0624	0.0522	0.79	log-log
Yukon	Ca	mg L^{-1}	72	33.10	-15	4992.3	-0.1981	0.8429	-0.0222	-0.1586	0.1085	0.62	log-log
Yukon	Mg	mg L^{-1}	73	9.15	322	5253.3	4.4288	0	0.1171	0.0794	0.1513	0.57	log-log
Yukon	Alkalinity	mg $CaCO_3$ L^{-1}	71	82.03	66	4831.3	0.9351	0.3497	0.1435	-0.1213	0.4504	0.8	log-log
Yukon	Li	µg L^{-1}	62	2.81	54	3423.3	0.9058	0.365	0.0115	-0.0092	0.0331	0.09	log-log
Yukon	U	µg L^{-1}	72	0.84	63	5092.3	0.8688	0.3849	0.0028	-0.0022	0.0093	0.39	log-log
Yukon	Ba	µg L^{-1}	73	49.83	-64	5253.3	-0.8692	0.3847	-0.1034	-0.4797	0.1353	0.3	log-log
Yukon	Sr	µg L^{-1}	73	145.95	284	5253.3	3.9045	0.0001	1.2018	0.7544	1.6802	0.56	log-log
Yukon	SO_4	mg S L^{-1}	72	12.24	353	4992.3	4.9819	0	0.2986	0.1995	0.3912	0.02	log-log
Yukon	Rb	µg L^{-1}	49	1.23	5	1799.0	0.0943	0.9249	0.001	-0.0213	0.0284	0.08	log-log
Yukon	K	mg L^{-1}	75	1.29	-202	5709.3	-2.6601	0.0078	-0.0178	-0.0291	-0.0059	0.01	log-log
Yukon	NH_4	µg N L^{-1}	75	6.67	-62	5709.3	-0.8073	0.4195	-0.1575	-0.5453	0.275	0.64	gam
Yukon	SiO_2	mg SiO_2 L^{-1}	75	6.67	-56	5709.3	-0.7279	0.4667	-0.027	-0.07	0.0288	0.43	log-log
Yukon	NO_3	µg N L^{-1}	74	105.43	100	5533.3	1.3309	0.1832	0.6771	-0.2928	1.5748	0.81	log-log
Yukon	Na	mg L^{-1}	73	3.16	174	5253.3	2.3869	0.017	0.0184	0.0053	0.0294	0.54	log-log
Yukon	Cl	mg L^{-1}	70	0.87	-286	4655.3	-4.177	0	-0.0234	-0.0314	-0.0133	0.39	log-log
Yukon	TDP	µg L^{-1}	75	7.25	-24	5709.3	-0.3044	0.7608	-0.0268	-0.1937	0.1549	0.02	log-log
Yukon	DOC	mg L^{-1}	75	5.17	4	5709.3	0.0397	0.9683	0.006	-0.0473	0.0652	0.54	log-log