



Patterns in riverine carbon, nutrient and suspended solids export to the Eastern James Bay: links to climate, hydrology and landscape

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Abstract The Eastern James Bay has been undergoing major shifts in its physical, chemical, and ecological functioning over the past decades, driven by a combination of climate and landscape changes, and human intervention that includes the damming of major regional rivers. Rivers play a role in the functioning of the Bay, delivering not only freshwater and key materials but also transmitting environmental and climatic signals. Here we present a study of the spatial variability of riverine export fluxes and yields of freshwater, dissolved organic carbon (DOC), total suspended solids, total nitrogen, and total phosphorus of 18 major rivers flowing into Eastern James Bay, in the boreal region of Québec (CA). We characterized discharge patterns and identified and modeled the main landscape drivers of riverine material

concentrations in these watersheds. We then combined these to model the contribution of unsampled watersheds to generate a regional budget of riverine freshwater and material export to the Bay. The peak of water export occurs in spring for free-flowing rivers, whereas for the dammed river La Grande, which accounts for half of the freshwater inputs, the peak discharge was shifted to winter due to high energy demands. The large gradient of watershed areas, discharge and environmental conditions resulted in the wide range of material concentrations across these boreal rivers, and we show that overall, the James Bay is a hot spot of DOC loading to the entire Hudson Bay System. We further reconstructed past (pre-damming) riverine export and compared this with current, and potential future scenarios, and we demonstrate that damming and climate change are impacting the patterns of water and material export of these boreal rivers to the Eastern James Bay.

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Introduction

Streams and rivers are the main vectors of organic and inorganic materials from land to the oceans, and sites of active biogeochemical transformation and emissions within the aquatic continuum (Battin et al. 2008; Cole et al. 2007). Despite comprising a

small fraction of the Earth's surface, fluvial networks play a key role integrating terrestrial, aquatic, and atmospheric processes, contributing significantly to regional and global biogeochemical cycles and material budgets (Cole et al. 2007; Raymond et al. 2013) as well as sustaining high rates of biological production in coastal systems (McClelland et al. 2014). High-latitude regions are particularly relevant to global biogeochemical cycles as they hold a high density of freshwater systems and drain vast areas of peatland and organic rich soils, which account for as much as 50% of global soil carbon stock (Dixon et al. 1994; Finlay 2003; Freeman et al. 2001). These watersheds export a disproportionate amount of terrestrial carbon (C) to the ocean as compared to other major biomes worldwide (Benner et al. 2004; Raymond et al. 2007). This is the case for a large portion of Canada that drains into the Hudson Bay system in the arctic-subarctic oceans, composed of the James Bay, the Hudson Bay and Ungava Bay basins that collectively drain approximately one-third of Canada's territory and supply the equivalent of ~18% of the total riverine discharge reaching the Arctic Ocean (Déry et al. 2005; Déry and Wood, 2005; Mundy et al. 2010; Shiklomanov et al. 2000).

There have been several reports of riverine dissolved organic and inorganic carbon (DOC, DIC) export to the Hudson Bay system (mostly from large rivers, see Mundy et al. 2010 for a compilation; Clair et al. 2013; Godin et al. 2017; Lung et al. 2018), and it has been estimated that a total of roughly 5.5 Teragrams of DOC per year are exported to the Bay by rivers, which is equivalent to ~23% of total riverine inputs into the central Arctic Ocean (Mundy et al. 2010; Raymond et al. 2007). The amount of C exported by the rivers draining into the Hudson Bay varies greatly between watersheds, and is not a simple function of absolute discharge, since the export yields (g C exported per m² of watershed per year) can vary by over an order of magnitude (Lung et al. 2018; Mundy et al. 2010). In contrast, there are far fewer studies considering nutrient export by these northern watersheds (Kuzyk et al. 2010; Zhang et al. 2021) but it has been estimated that Northern Canadian rivers export proportionally less nitrogen to the ocean compared to other watersheds across Europe and the United States due to significantly less agricultural activity, urban development, and atmospheric deposition (Clair et al. 2013).

The variability in C export yields between rivers is linked in part to regional runoff (Lung et al. 2018; Mundy et al. 2010; Raymond et al. 2007), and the concentration of materials in river water. Some studies have reported that the concentration of C and other elements tend to peak at highest discharge, i.e., snowmelt and rainfall events (Holmes et al. 2012; Mattsson et al. 2005; McClelland et al. 2016), yet others have reported little or no link between discharge and material concentrations in rivers (Lung et al. 2018). While runoff is driven mostly by regional climate, concentration is driven by watershed features such as geology, soil composition, land cover and human activities (e.g., agriculture) across watersheds, which are also important regulators of material leaching from land into aquatic systems (Ågren et al. 2007; Boardman et al. 2019; Maavara et al. 2017). For example, total and dissolved C concentrations in northern streams and rivers have been linked to the proportion of wetlands and peat cover within the catchment (Aitkenhead and McDowell 2000; Creed et al. 2008; Huotari et al. 2013), whereas total nitrogen showed a relationship with mean annual temperature, forest cover, specific discharge, and agricultural activity in northern rivers of Sweden (Sponseller et al. 2014). Phosphorus and suspended solids, on the other hand, were found to be related to watershed area and land-cover (wetlands and pasture) variables in an agricultural watershed in Canada (Rutledge and Chow-Fraser 2019), and have also been related to lithology and soil characteristics within watersheds elsewhere (Bywater-Reyes et al. 2017; Mueller and Pitlick 2014; Porder and Ramachandran 2013). The configuration of the aquatic network further influences the mobilization, processing, retention, and transport along the aquatic continuum (e.g., catchment slope, Zarnetske et al. 2018), and therefore determines the nature and amounts of materials that reach the oceans. For example, the concentration of DOC in boreal and temperate streams and rivers has been reported to be negatively correlated with the proportion of upstream lakes in the catchment, which act as a sink of C (Larson et al. 2007; Mattsson et al. 2015).

There is evidence to suggest that watersheds in the southeastern Hudson Bay, including the James Bay basin of the Hudson Bay system may be characterized by consistently higher DOC export yields (Mundy et al. 2010) relative to watersheds across the boreal and subarctic biomes (Lung et al. 2018; Raymond

et al. 2007); this region might therefore represent a hot spot of riverine export of organic matter to the Arctic Ocean. This existing evidence is limited to a handful of watersheds (Mundy et al. 2010), however. In addition, the underlying climatic and environmental drivers for this elevated C export are not well constrained, and it is unclear if other materials, such as nutrients and suspended solids, also follow a similar pattern.

The drivers of riverine discharge and material concentration are extremely dynamic, and in northern landscapes in particular, the hydrologic cycle and land cover that influence riverine export have been dramatically changing (Déry et al. 2009; Wang et al. 2020). In the case of the Eastern James Bay, the development of major hydropower projects have led to significant shifts in both discharge patterns (Déry et al. 2016; Prinsenber 1980; Roy and Messier 1989) and landscape features (such as flooded area Roy and Messier 1989), and therefore likely on the export of water and materials to coastal ecosystems (Déry et al. 2016). Riverine export and ice cap melting have profound implications on the mix of fresh and salt waters in river plumes (Taha et al. 2019) and in consequence can affect the temperature, salinity, density, and turbidity of coastal waters, which in turn have major implications on the functioning and productivity of the marine ecosystem. These effects are especially notable in the mouth of La Grande River, the second largest river in Québec, which has had its natural streamflow pattern shifted due to extensive damming to fulfill hydropower demands (La Grande hydroelectric complex project led by Hydro-Québec), in a project that altered the regional hydrology due to the diversion of other rivers (Roy and Messier 1989; Déry et al. 2016). In addition, studies in the region have confirmed climate change projections for subarctic and arctic regions (IPCC 2007), with significant trends towards earlier ice break-up events, longer ice-free seasons, and increased precipitation (Gagnon and Gough 2005; Gough et al. 2004; Guay et al. 2015; Taha et al. 2019).

Quantifying and understanding the patterns and shifts in riverine material export implies addressing the hydrological component (discharge), as well as quantifying material concentrations across a wide range of watershed types and constraining the drivers of the variability in these concentrations, which ultimately requires assessing climatic, watershed and

network features. The aim of this study was to examine the spatial variability of DOC, TSS, and nutrients (TN and TP) concentrations and export across 18 free-flow and regulated rivers flowing along the entire Eastern James Bay in the boreal region of Québec with watersheds spanning a wide range in size, land cover, underlying geology, and climate. In addition, we aimed to identify the main watershed-scale drivers of the variability in the riverine concentrations of materials and create models to estimate materials concentration in unsampled rivers across the region. Combining empirical and modeled data we aim to generate a regional estimate of total water and material export from the ensemble of rivers to the Eastern James Bay. We further aimed to reconstruct water and material exports by regional rivers in pre-damming conditions and assess how it may differ from our current estimates. We finally aim to generate a projection of DOC export by rivers into the James Bay coast by the year of 2050 based on hydrological projections for Québec in Canada and long-term trends in DOC concentrations measured across North America. We hypothesized that total material exports are mainly driven by the magnitude of river discharge, however landscape-scale properties will modulate materials concentration across these northern watersheds.

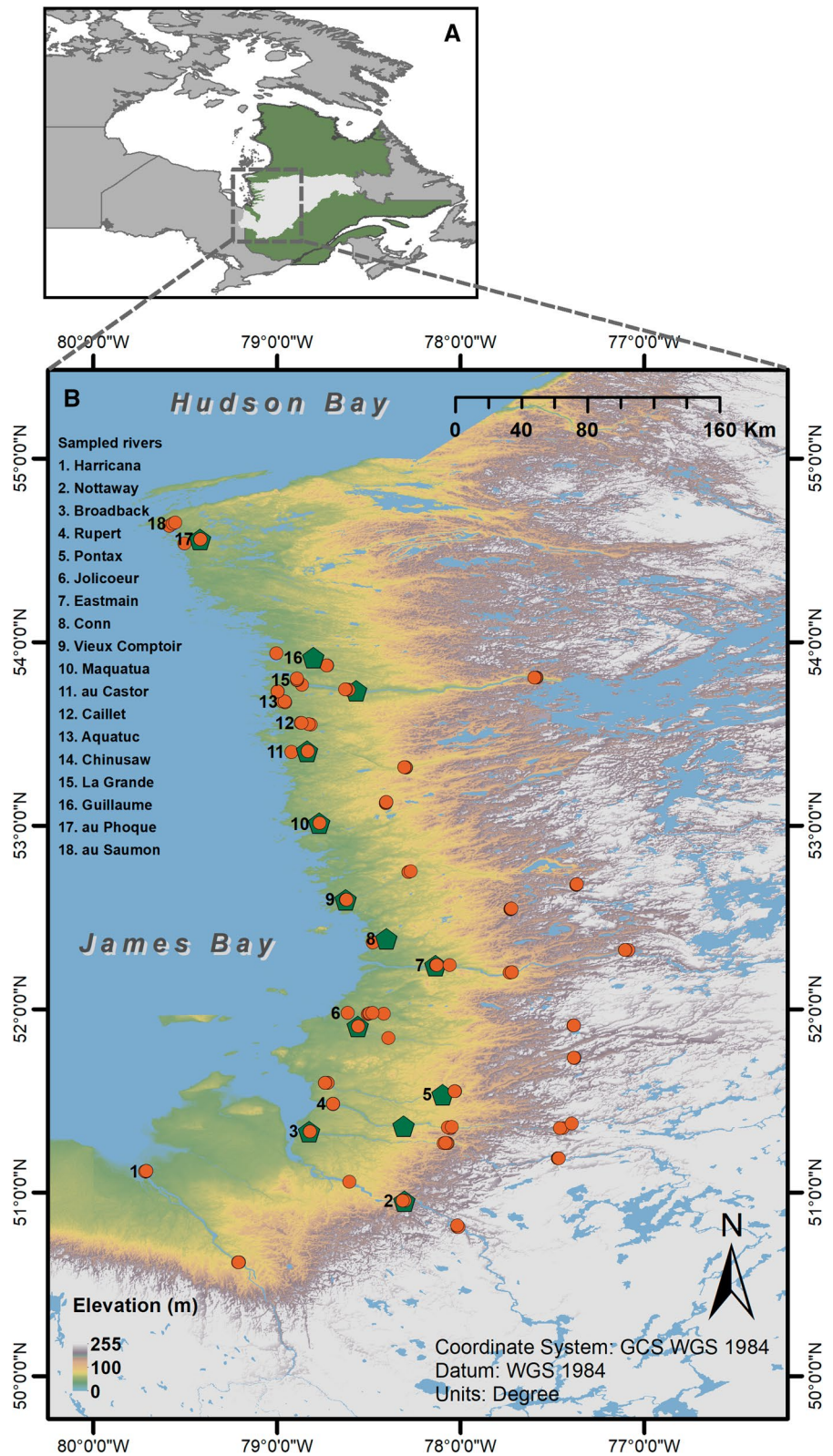
Methods

Study area and sampling

The research presented here was carried out as part of the broader Coastal Habitat Comprehensive Research Project Eeyou Istchee, whose main objective was to understand the reasons underlying the generalized decline of eelgrass (*Zostera marina* Linnaeus, 1753) along the eastern James Bay coast and possible links with waterfowl migration. For this, it is needed to quantify riverine inputs, i.e., freshwater, carbon, nutrients, and turbidity, as they affect the coastal environmental conditions. Information on this project can be found online: <https://www.eeyoucoastalhabitat.ca>.

The study area is in the Eastern James Bay region of northern Québec (Fig. 1), which comprises the traditional territory of indigenous Cree people (*Eeyou Istchee*, which means the land of the Eeyou/Eenou people). It occupies the southern end of the Hudson Bay in the subarctic sea and connects to

Fig. 1 Map of the **A** study area Eastern James Bay in the boreal region of Québec showing **B** all sampling sites over the 5 campaigns in the 18 sampled rivers. Orange dots represent sampling stations, and green pentagons represent the location of hydrometric stations. The hydrometric station in Harricana River is in a sampling site more upstream (coordinates 48.5702° N, -78.1215° W), and does not appear on the map



the Arctic Ocean via the Foxe Channel and extends roughly from 49 to 55 degrees north latitude covering more than 350,000 km² (Roy and Messier 1989). The Eastern James Bay drainage basin lies predominantly within the Canadian Shield and Hudson Lowlands, and the landscape is mostly composed of a coastal plain area with scattered peat bogs and rich clay deposits, and a plateau riddled by numerous lakes (Roy and Messier 1989). The coast is featured by sparse and continuous eelgrass meadows (*Zostera marina*). The climate is classified as moist continental mid-latitude (Köppen Climate Classification), which is characterized by a long and cold winter with temperatures reaching –30 °C, ice break-up happening around mid-April to May, maximum annual temperatures of 20 °C (in July), remaining positive until mid-October and mean annual temperatures of –3 °C (Canadian Climate Normals 1981–2010, measured at the nearby meteorological station, La Grande Rivière A, Fig. S1). The precipitation is higher during late summer and fall, with total monthly precipitation highest during September (110 mm) and lowest during February (22 mm). The largest part of the Eastern James Bay territory is covered by isolated patches of permafrost (0 to 10% coverage) (Obu et al. 2019).

Field operations were carried out with full consent from local land users, in collaboration with local communities and with the engagement of the Niskamoon Corporation of the Cree Grand Council. Water sampling was conducted starting from Harricana River (48° 34' 12.6" N, 78° 07' 17.3" W) in the south to au Saumon River in the north (54° 33' 35.0" N, 79° 25' 04.5" W) and included 18 rivers in total (Fig. 1), with watersheds covering more than 90% of the total area of the Eastern James Bay region. We performed water sampling in the river's subsurface in different sections, river mouth, half-way, upstream, once during five consecutive seasons, from summer 2018 to summer 2019 (Fig. S2). However, due to logistical challenges, this sampling strategy with three stations per river cannot be always applied (climatic conditions, or lack of permission to operate in restricted indigenous lands), but each of the studied rivers had at least one sampling station close to the river mouth. For sampling, we adopted a combined strategy to sample remote areas by using helicopters (hovering or on land), boats and cars, accessed by roads and bridges. Nonetheless, some small rivers could not be sampled during the winter because they

were completely frozen (rivers Aquatic, au Phoque, au Saumon, Caillet, Chinusaw, Guillaume).

Riverine discharge data

Our methodological approach is summarized in the Fig. S3 and described in detail below. Historical discharge data from rivers flowing into the eastern James Bay were recovered from the Hydrometric Database (HYDAT, Water Survey of Canada, 2020, https://wateroffice.ec.gc.ca/mainmenu/historical_data_index_e.html). These include hydrometric stations (HS) that recorded data for periods between 10 and 85 years (Table S1). These historical data were used as input for developing a regional empirical model of annual river discharge based on watershed area (see details below). For each of these instrumented rivers we used an average of the years for which a full record was available (details in Table S1).

In addition to the historical data, we also used contemporary measurements to characterise the current patterns of discharge of selected James Bay rivers. We have used discharge data from HS currently operated by Hydro-Québec, which include: (1) Four stations in large rivers in operation since 2016 (Broadback, Nottaway, Pontax, Rupert): We calculated average daily discharges for these rivers based on 4 years of available data record, from October 1, 2016 to September 30, 2020 (Water Year definition, U.S. Geological Survey). However, the HS Pontax was closed in August 2019, and we only have 3 years of discharge record available for this site. (2) Eight new HS installed in 2018 in the context of this research project (Coastal Habitat Comprehensive Research Project—Eeyou Istchee) in the following rivers: au Castor, au Phoque, Conn, Eastmain, Guillaume, Jolicoeur, Maquatua, and Vieux Comptoir, for which we recovered data for a period of 2 years, from October 1, 2018 to September 30, 2020. All these HS record water temperature and hourly water level measurements that are transmitted via satellite to a server and are then converted to discharge estimates using empirical calibration equations developed for each site. Calibrations of the new HS were performed by Hydro-Québec Hydrology team using a Doppler device integrated into a remote-controlled boat, which was used to map the bathymetry of the cross section of the river channel at the site of the pressure

sensor, as well as to determine a continuous profile of water velocity and direction. These variables are then integrated to obtain an average water velocity and volume of the river channel at the site, and this procedure was repeated at different flow regimes on at least 4 different occasions, in order to develop a calibration function to transpose continuous water level measurements to river discharge. The stations were visited on other occasions for maintenance and to ensure that the sensors were not displaced in the period between calibrations and therefore that the baseline had not shifted.

We gathered data corresponding to this same 4-year interval for La Grande River, derived from the La Grande 1 generating station that is less than 30 km from the mouth (data provided by Hydro-Québec) and for Harricana River from a site ~480 km upstream of the mouth (source: <https://www.cehq.gouv.qc.ca/>). For the HS Harricana, due to the large distance from the mouth, we used the annual hydrography (daily contributions in % to the total annual discharge) to assess seasonal patterns in this watershed but we modeled the mean annual discharge at the mouth using the watershed area as described below. Complete hydrological data and gaps in our dataset are presented in the Supplementary Information Section (“Hydrographs at hydro-metric stations of rivers in the Eastern James Bay”, Figs. S8 to S20).

The empirical relationship that we developed using measured annual river discharge and watershed area, based on the historical data described above (Fig. S4a), was then used to estimate discharge of non-instrumented rivers for which these data were not available. The model was validated by comparing modeled and measured annual discharge in instrumented rivers, the latter derived from current HS (R -squared=0.97, Fig. S4b). In addition, we derived an average annual hydrograph based on all 13 annual hydrographs of instrumented rivers (except for La Grande River, which has a regulated flow), which we applied for estimating daily discharge contribution of non-instrumented rivers (these hydrographs are presented in the results section). This was done by multiplying the predicted annual discharge ($\text{m}^3 \text{s}^{-1}$) generated by the empirical model using watershed area by the daily contributions (%) by this “average annual hydrograph”. We assume that this average annual hydrograph represents the regional trend in discharge

over the year for unregulated rivers draining into the Eastern James Bay.

Environmental variables

Physico-chemical variables such as dissolved oxygen (mg L^{-1}), water temperature (degrees Celsius), electrical conductivity (microsiemens cm^{-1}) and pH were measured in situ at the subsurface (0.5 m) using a YSI probe (600XLV2-M, Yellow Springs Instruments, Yellow Springs, OH, USA) that was calibrated daily. Additionally, for each site, water was sampled in 20 L acid washed polycarbonate containers for further subsampling in the laboratory. In the laboratory, samples for dissolved organic C (DOC) were filtered in duplicates through $0.45 \mu\text{m}$ polyethersulfone filters and measured on a TIC-TOC Analyzer using wet persulfate oxidation (OI Analytical, TIC TOC 1010 College Station, TX, USA). The absorption coefficient at 440 nm (a proxy for colored DOC) was measured with an Ultrospec3100 spectrophotometer (Thermo Fisher Scientific Inc., Waltham, MA, USA). For total suspended solids (TSS), we averaged the dry weight of material after filtration in duplicates of pre-combusted and pre-weighed GF-F filters.

Total nitrogen (TN) concentration was determined in duplicates using alkaline persulfate digestion with an Alpkem Flow-Solution IV autoanalyzer (O I Analytical, College Station, TX, USA). Total phosphorus (TP) was measured in duplicates after persulfate digestion as orthophosphate with the spectrophotometric molybdenum blue technique (890 nm, Ultrospec 2100 pro, Biochrom Ltd., Cambridge, UK).

Export of materials

Export of DOC, nutrients and TSS was estimated as the product of the constituent concentration (mg or $\mu\text{g L}^{-1}$) in the water and the river discharge (Q in $\text{m}^3 \text{s}^{-1}$) either measured or modeled at the river mouth, as follows:

$$Export_{DOC,TSS,TP,TN} = [DOC, TSS, TP, TN] \times Q.$$

We used a mean material concentration value of all measured values (up to 5 campaigns, river mouth) to calculate an annual mean concentration of DOC, TSS, TN, and TP, which were then used to calculate the annual export. To calculate the uncertainties of

our export estimations, we used a Monte Carlo propagation approach, where the annual mean and standard deviation material concentration (separately for DOC, TN, TP and TSS) of each river were used to generate 1000 random values following a normal distribution using the functions *rnorm* in R platform. These values were used to feed a Monte Carlo propagation (size = 1000), which multiplied materials' concentration by the fixed annual discharge of each river ("Riverine discharge data" section). We present the 25% and 75% quartiles for each sampled river generated by the Monte Carlo propagation. Uncertainty in river discharge could not be directly quantified in this study, but the fits of the empirical calibration equations used to estimate discharge from water level data were in general very strong.

For the unsampled watersheds (Fig. S6), we developed empirical models to predict river material concentrations (annual average) based on watershed properties (described below in the "Empirical models for material concentration and other statistical analyses" section), and we used these together with modeled average annual discharge to derive average annual export of each constituent.

Watershed properties

Watershed area, when not provided with the discharge data from gauged stations, was determined using the Canadian Elevation Digital Model (http://ftp.geogratis.gc.ca/pub/nrcan_rncan/elevation/cdem_mnec/), HydroSHEDS (<https://www.hydrosheds.org>) in ArcGIS 10, and the Hydrology Toolset of ArcGIS (tools Flow direction, Flow accumulation, Snap Pour Point and Watershed), which were used to delineate the watersheds above the sampling sites. For each watershed polygon, the percentage of the different land covers (i.e., coverage by forest, roads, wetlands, water) was determined using the Land Use 2010 dataset with a spatial resolution of 30 m and an overall accuracy of 92.7% (Agriculture and Agri-Food Canada 2010). For elevation and slopes, we interpolated the watershed shapefiles into the elevation raster (from Canadian Digital Elevation Model) using the ArcGIS tool *interpolate shape* and then we calculated the average watershed slope of each delimited watershed using the tool *Add z information*. Clay within watersheds were extracted from a global compilation (www.soilgrids.org) in VRT format and visualized in

ArcMap 10.7.1. After clipping our delimited watersheds, we calculated a weighted arithmetic mean of clay content (surface 0–5 cm).

For soil classification we used Soil Landscapes of Canada version 3.2 (Soil Landscapes of Canada Working Group 2010), whereas for the classification of geology units, we used the Geologic Map of North America (Garrity and Soller 2009).

Empirical models for material concentration and other statistical analyses

We used Elastic Net regressions to select the most robust subset of watershed properties driving DOC, TSS, TN, and TP annual average concentrations in rivers draining the Eastern James Bay. This approach performs a selection step that helps mitigate multicollinearity issues (very common in environmental data), reduces overfitting on sample data, and produces a more accurate model of complex data. The explanatory variables used as inputs for models included: clay content (surface 0–5 cm), average watershed slope, watershed area, land covered by water, forest, grasslands, forested wetlands (individual classes or as the sum of treed wetlands and forested wetlands) and wetlands (individual classes or as the sum of open wetlands, wetland with shrub cover, wetlands with herbaceous cover) and percentage cover by Organic soils. For TSS and TP (to improve poor prediction models using the same variables as used for DOC and TN), we have performed an explanatory analysis including the percentage cover by different rock types (metamorphic, volcanic, plutonic, sedimentary rocks) and soil orders (brunisollic, podzolic, regosolic). We ran Elastic Net using a machine learning approach by first randomly splitting the data into a training set (70% of data to build a predictive model = 34 samples) and a test set (30% of the data to evaluate the model performance = 12 samples). Using the Caret package (Kuhn 2008) function *train()* in R, we set the function arguments to: (a) center and scale the data a priori; (b) specify the type of resampling using method "repeatedcv" to run a tenfold cross-validation on the training data using 10 re-samplings. We used bestTune to automatically select the best tuning parameters (alpha and lambda) and compute the final model with the best set of predictors and coefficients, i.e., those that minimize the root mean squared error (RMSE) and maximize the coefficient of

determination (R-squared). We also tested the power of each model in making predictions on the test data and computed the R squared. We further used these regression models to estimate the annual average concentration of DOC, TN, TPP, and TSS in rivers that were not sampled due to their remote location, logistic difficulties, climatic conditions, or lack of permission to operate in restricted indigenous lands.

All statistical analyses used in this study were performed in R version 4.0.2, the packages *vegan* and *plyr* (Wickham 2011) were used for data manipulation. Figures were created using R (*ggplot2*, Wickham 2016) and ArcMap (version 10.7.1).

Reconstructing pre-dam exports

To reconstruct the pre-dam export of C and nutrients to the James Bay, we used data from the environmental monitoring network that was set by Hydro-Québec to assess the hydrologic, chemical, and biological conditions of impacted watersheds before, during and after the development of the La Grande hydroelectric complex (Schetagne et al. 2005). We used these data to build the water balance and riverine export of C and nutrients into the Bay for La Grande River before damming, and for Eastmain-Opinaca and Rupert rivers (the rivers for which historical data are available) before diversion. More details about our methods are described in the Supplementary Information Section “Reconstructing riverine exports in the past”. Because this dataset does not include individual measurements taken during the monitoring program or data standard deviation, we have assumed that uncertainties in past exports correspond to the mean of first and third quartiles (Monte Carlo propagation) calculated from the 18 sampled rivers in this study.

Results

General features of the studied watersheds, carbon, suspended solids, and nutrients concentrations

The Eastern James Bay receives riverine flows from thirty-eight watersheds, which encompass a total area of about 386,880 km² and span a large gradient of surface area, annual discharges, and environmental conditions, which ultimately resulted in the large variability of C (DOC), suspended solids (TSS), and

nutrient (TN, TP) concentrations in the river waters (Table 1, Table S2). In this study we sampled 18 of these 38 major watersheds (which encompass more than 90% of the Eastern James Bay region) spanning a large range of watershed areas, ranging from 44 km² (Chinusaw River), to more than 62,000 km² and an annual discharge of 1166 m³ s⁻¹ (Nottaway), and La Grande River has by far the largest watershed area with approximately 210,000 km², and an annual discharge of 3782 m³ s⁻¹ (Table S2), due to the various river diversions (see Supplementary Information for a regional description).

DOC mean concentrations in the river mouth ranged between 4.0 and 25.4 mg L⁻¹, with the lowest values consistently found in La Grande River. TSS and TP were the most variable constituents, ranging from 1.0 to 233.2 mg L⁻¹, and from 6.5 to 247.4 µg L⁻¹, respectively, both values peaking in the campaigns performed in spring and summer. TN ranged from 0.15 to 0.71 mg L⁻¹. The lowest concentrations of TSS and nutrients were also consistently observed in La Grande (Table 1).

The concentration of constituents measured across rivers and within different river sections and seasons were correlated overall, albeit the degree of coupling varied between constituent pairs (Fig. 2). DOC was strongly related to TN ($R^2=0.50$, $p<0.001$, Fig. 2a), and weakly related with TSS ($R^2=0.18$, $p<0.001$, Fig. 2b). Nutrients were well correlated between them ($R^2=0.43$, $p<0.001$, Fig. 2d), and the strongest relationship among all elements was found between TSS and TP ($R^2=0.67$, $p<0.001$, Fig. 2e).

Watershed-scale drivers of constituent concentrations

We used the annual average of each constituent concentration calculated for each river section (i.e., upstream, half-way, and mouth) for all further statistical analyses in this section. We developed Elastic Net regression models to overcome multicollinearity issues and to identify the most robust combination of drivers of material concentrations (Table 2). Our models based on watershed properties explained on average over 70% of the variability in annual mean riverine material concentrations (Table 2) and were particularly effective for DOC (83%). When we used the models to make predictions on the test data, we found that they performed satisfactorily well for DOC, TN, and TP (R^2 of 0.73, 0.46 and 0.54, respectively),

Table 1 Mean and standard deviation of physico-chemical variables measured at river mouth in each one of the 18 rivers sampled during this study

River	pH	Electrical conductivity ($\mu\text{S cm}^{-1}$)	Color (Abs 440 nm)	DOC (mg L^{-1})	TSS (mg L^{-1})	TN ($\mu\text{g L}^{-1}$)	TP ($\mu\text{g L}^{-1}$)
Aquatuc	6.4 (1.2)	39.7 (22.5)	14.5 (3.1)	19.3 (5.1)	6.2 (1.2)	388.8 (115.9)	19.1 (4.0)
Au Castor	6.8 (0.8)	26.3 (8.4)	7.8 (1.3)	11.8 (1.7)	4.3 (2.0)	264.7 (33.9)	14.7 (5.2)
Au Phoque	7.0 (0.6)	60.3 (17.7)	3.2 (0.9)	7.6 (0.8)	3.4 (1.0)	300.7 (15.3)	23.8 (4.4)
Au Saumon	7.4 (1.0)	790.8 (680.7)	2.1 (0.9)	6.2 (0.6)	12.8 (15.4)	404.4 (89.8)	24.2 (10.1)
Broadback	6.9 (0.7)	23.0 (6.0)	6.7 (1.4)	10.4 (1.5)	54.8 (100.0)	296.6 (113.9)	68.0 (100.4)
Caillet	6.7 (0.8)	42.8 (19.7)	13.2 (1.8)	17.4 (3.3)	12.0 (5.2)	356.0 (88.1)	30.5 (10.3)
Chinusaw	6.5 (0.7)	112.5 (73.7)	18.1 (3.3)	21.8 (4.4)	60.6 (94.4)	537.1 (72.0)	103.4 (50.3)
Conn	6.8 (0.9)	40.2 (21.0)	12.4 (2.0)	15.4 (3.9)	7.7 (5.4)	429.4 (176.8)	29.3 (10.2)
Eastmain	6.2 (0.6)	23.6 (8.6)	10.3 (1.4)	13.8 (1.5)	18.3 (11.7)	296.4 (39.3)	50.9 (24.9)
Guillaume	6.6 (0.7)	36.8 (20.7)	13.9 (3.4)	18.9 (5.0)	8.7 (2.4)	378.4 (97.0)	29.0 (7.3)
Harricana	7.7 (0.8)	89.8 (51.9)	8.1 (1.5)	14.4 (0.7)	20.9 (18.5)	437.7 (109.5)	38.5 (18.3)
Jolicoeur sud	6.4 (0.5)	71.0 (51.6)	14.5 (3.8)	16.4 (4.1)	7.9 (1.4)	374.9 (98.8)	33.5 (11.1)
La Grande	6.3 (0.5)	11.2 (2.2)	1.8 (0.3)	4.3 (0.3)	2.9 (1.8)	178.1 (37.5)	9.5 (3.6)
Maquatua	7.5 (2.1)	25.0 (7.2)	8.1 (1.3)	12.5 (1.6)	4.8 (3.3)	269.1 (33.2)	13.9 (6.5)
Nottaway	7.2 (0.6)	22.5 (8.3)	7.5 (1.3)	12.4 (1.2)	9.2 (4.9)	341.9 (24.9)	26.1 (11.7)
Pontax	6.7 (0.8)	35.8 (16.3)	10.1 (5.6)	13.5 (4.3)	33.6 (44.4)	306.5 (73.3)	37.7 (26.2)
Rupert	6.6 (1.2)	16.3 (8.2)	3.6 (0.9)	6.0 (0.8)	7.9 (7.5)	211.9 (26.9)	10.4 (2.0)
Vieux Comptoir	6.5 (0.5)	28.6 (6.5)	8.6 (2.5)	12.0 (3.3)	3.9 (2.6)	283.6 (45.1)	15.4 (1.8)

Saltwater intrusion is a common phenomenon in small rivers such as Au Saumon River ($790.8 \mu\text{S cm}^{-1}$)

but was less satisfactory for TSS ($R^2=0.12$), which is probably due to the small sample number, limited temporal cover, and large within river variability that is difficult to account for this constituent. DOC and TN were strongly negatively related to the proportion of water coverage within the watersheds (Fig. 3a, Table 2). All constituents increased as a function of the coverage of various types of wetland habitats (Fig. 3b–e, Table 2). Models for DOC and TN concentrations also included positive relationships with the clay content in surface soil of their watersheds and percentage land cover by organic soils. The TP model included a negative relationship with the percentage watershed cover by brunisolic soils, and positive relations with forests and wetlands within the watersheds (Table 2). The model for TSS variation included % watershed cover by wetlands (rich in herbaceous plants), volcanic rocks, brunisolic soils and grasslands. The overall negative relation between TN with watershed slope (Fig. 3f) may be linked to the variability in watershed surface area across the region, since small watersheds are located on the flat coast landscape, which in turn may be linked also to

increased coverage of wetlands and increased organic C contents in soils.

Hydrologic patterns

Data from the 14 existing hydrometric stations show that the riverine discharge varied along the year and among rivers, but highest daily contributions consistently peaked around the 140th day of the year, which corresponds to the peak of snowmelt in spring (Fig. 4a, Fig. S7). Smaller rivers tended to have a more accentuated discharge peak (e.g., Guillaume, Vieux Comptoir, Conn, etc.), whereas large rivers had a longer but less sharp peak (e.g. Broadback, Nottaway, etc.). The northernmost instrumented river, au Phoque, had a somewhat delayed peak (approximately 10 days later, Fig. 4a) probably reflecting latitudinal climate effects. La Grande River did not have a clear spring discharge peak as observed for free-flowing rivers because the spring runoff is used to fill up reservoirs but tended to have overall higher daily contributions between December and March, which correspond to the peak of hydroelectric energy generation

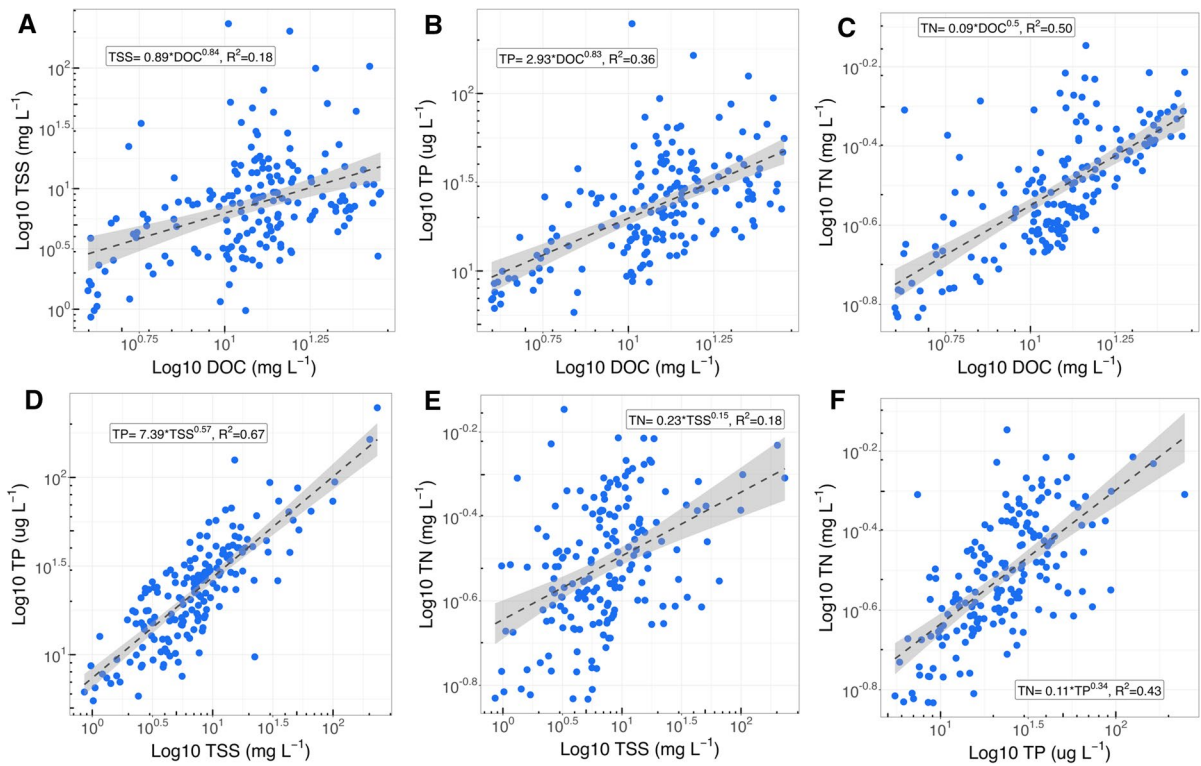


Fig. 2 Relationship between dissolved organic carbon (DOC), total suspended solids (TSS), total phosphorus (TP), and total nitrogen (TN) concentrations measured in 18 rivers located in

the Eastern James Bay. Samples from all seasons and river sections are included. All relationships are significant ($p < 0.05$)

Table 2 Equations derived from elastic net regression based on empirical data collected in this study

Variable	Intercept	Drivers	RMSE	R2
DOC	14.84	$-2.57 \times \text{Water} - 1.37 \times \text{Slope} + 1.24 \times \text{Wetland} + 0.79 \times \text{Clay} + 0.55 \times \text{Organic} - 0.06 \times \text{Grassland}$	3.86	0.83
TN	0.34	$-0.029 \times \text{Water} + 0.025 \times \text{Clay} + 0.024 \times \text{Wetland} - 0.022 \times \text{Grassland} - 0.020 \times \text{Slope}$	0.10	0.67
TP	27.53	$-2.65 \times \text{Brunisolic} + 1.15 \times \text{Forest} + 0.68 \times \text{Wetland}$	10.68	0.64
Log(TSS)	2.23	$0.18 \times \text{Wetland_Herb} + 0.10 \times \text{Volcanic} - 0.005 \times \text{Brunisolic} - 0.0003 \times \text{Grassland}$	0.59	0.65

The landscape properties were scaled and centered a priori. The presence of a variable in the model equation does not guarantee that it alone has a clear linear relationship with the response variable

demands. In general, river discharge was higher in spring, which accounted for 34% (Broadback) to 57% (Jolicoeur and Conn) (Table S3) of the annual discharge into the Bay. The average discharge contribution of summer (months of July, August, and September) and fall (October, November, and December) were similar, and varied between 18 to 28 and 13 to 25%, respectively (Table S3). The lowest contribution occurred during the winter (months of January, February, and March), ranging between 2 and 18% (rivers

Guillaume and Broadback, respectively) of the total annual discharge. In contrast, in the regulated river La Grande winter discharge contributed on average 31% of the total annual discharge, followed by fall (26%), and the lowest contributions occurred in summer (22%) and spring (21%). On average, 47% of the total freshwater discharge reaches the eastern coast during spring (but excluding La Grande), followed by similar contributions in fall and summer, 23% and 20% in respectively, and 10% in winter.

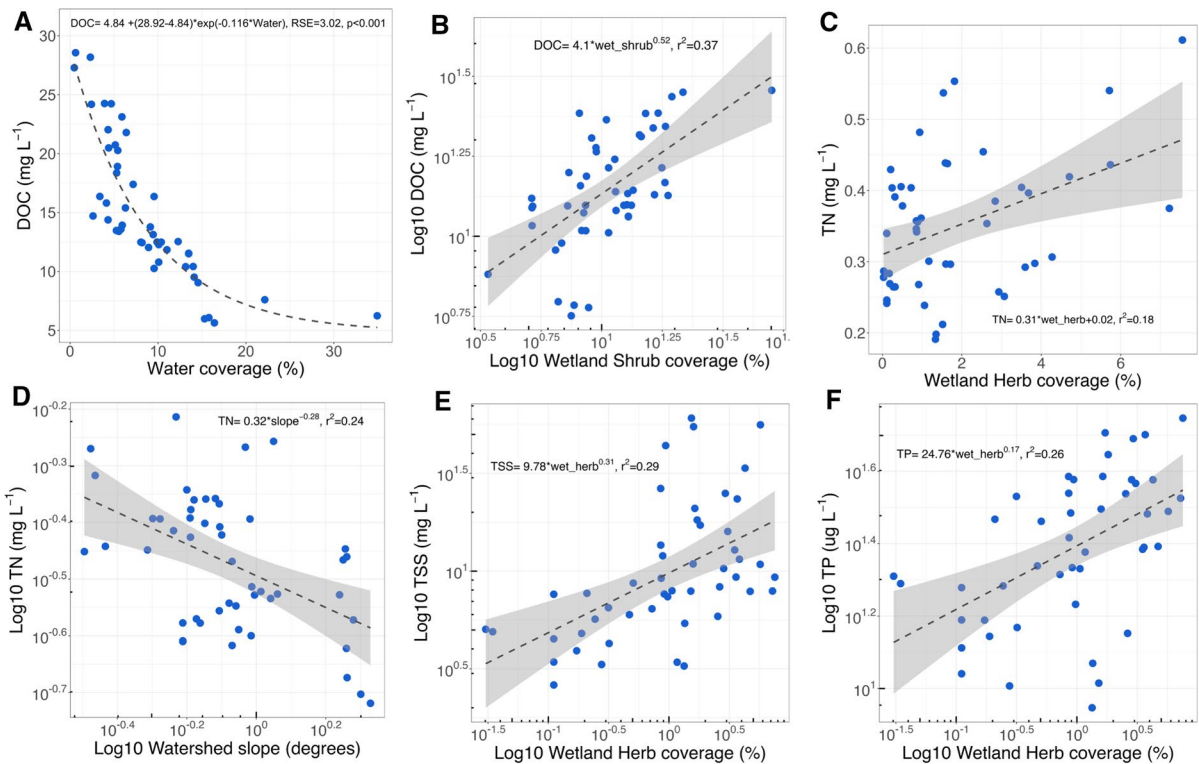


Fig. 3 Significant relationships between material concentrations and watershed properties. La Grande River not included

The total annual freshwater inputs to the Eastern coast of James Bay are in the order of 227 km³ per year, of which c.a. 213 km³ year⁻¹ (or 94%) are delivered by the 18 rivers sampled in this study. The remaining water input (14 km³ year⁻¹ is associated with 20 small and medium unsampled watersheds, Fig. S6) whose discharge (surface or groundwater inputs to the coast) was estimated using our empirical model based on watershed area (Fig. S4). On average, 47% of the total freshwater discharge from unregulated rivers reaches the eastern coast during spring, followed by 23% in fall, 20% in summer, and 10% in winter. The inclusion of La Grande, which contributes on average to 53% of the total annual discharge but which has a very different annual hydrograph, significantly alters the pattern of freshwater delivery to the Bay, with a smaller contribution of discharge in spring (31%), and a much higher contribution in winter (21%), with summer and fall remaining similar (21% and 25%, respectively).

Spatial patterns of yield and export of carbon, suspended solids and nutrients into the Bay

The export yield (or simply yield) was calculated as the ratio between annual riverine export at the mouth of each river and its corresponding watershed area and represents the riverine export per unit of watershed area (Fig. 5, Table S4). The highest annual DOC yields (> 8.8 g C m⁻² year⁻¹) were found in rivers at both ends of the watershed size and discharge spectrum (Guillaume, Jolicoeur and Harricana). Likewise, the lowest yields (< 2.5 g C m⁻² year⁻¹) occurred in opposite ends of the watershed range: in La Grande River (Fig. 5a, Table S4), which had the lowest DOC concentrations and highest total watershed area, and in au Saumon River, which has among the smallest watersheds (68 km²). TN yields followed a similar spatial pattern as DOC (Fig. 5e, f), but with a narrower variation of 2.5-fold range of values, with a maximum of 0.27 g N m⁻² year⁻¹ (Harricana) and minimum of 0.10 g N m⁻² year⁻¹ in La Grande River.

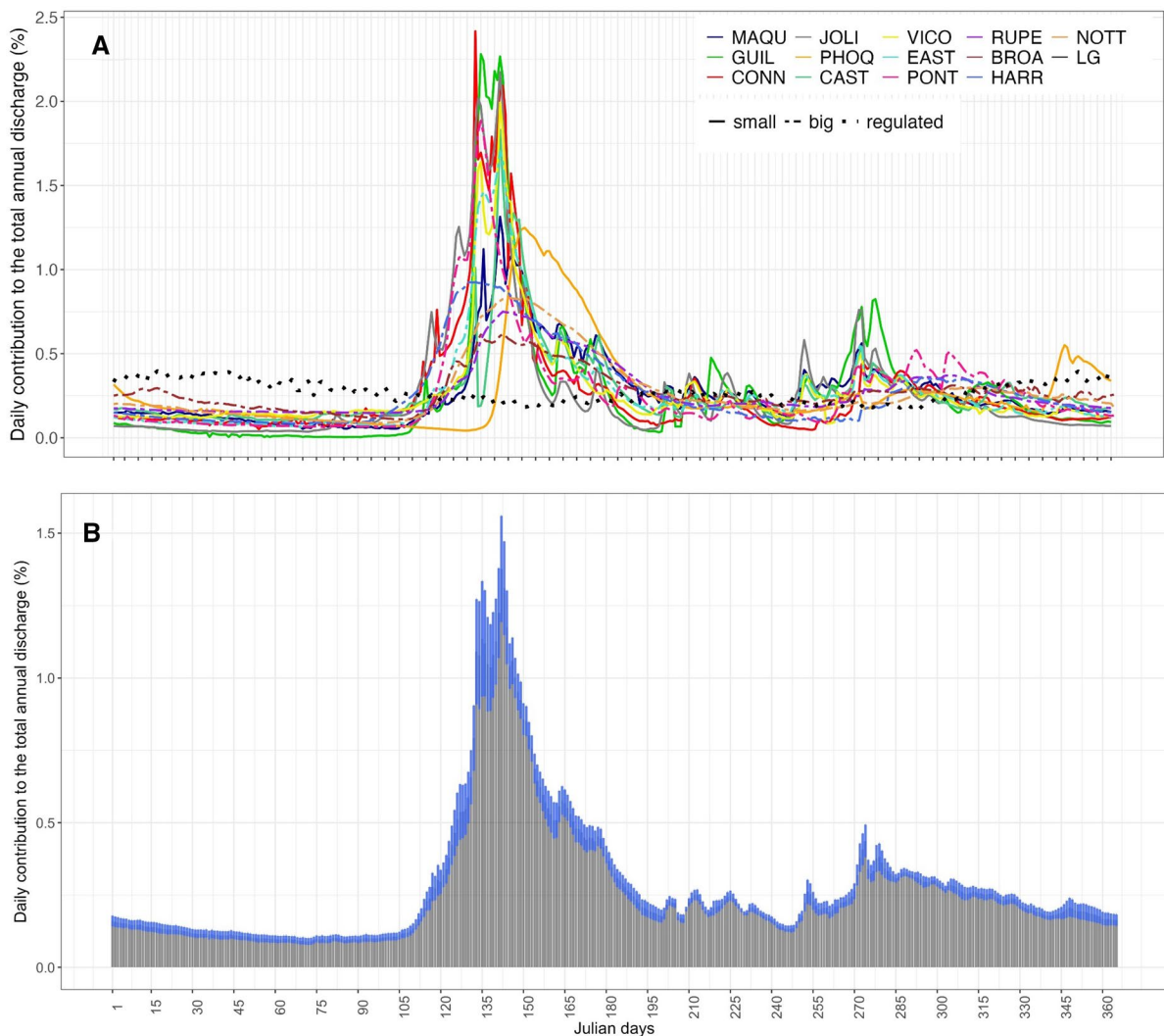


Fig. 4 **A** Mean daily discharge contribution in Julian days to the total annual discharge (sum of all daily discharges) from each of the hydrometric stations installed by our group. Abbreviations and symbols for rivers: Maquatua (MAQU), Guillaume (GUIL), Conn (CONN), Jolicoeur (JOLI), au Phoque (PHOQ), au Castor (CAST), Viex Comptoir (VICO), Eastmain (EAST), Pontax (PONT), Rupert (RUPE), Broadback (BROA),

Harricana (HARR), Nottaway (NOTT), and La Grande (LG). The continuous lines represent small rivers, those with mean annual discharge $< 50 \text{ m}^3 \text{ s}^{-1}$. Note that the dammed river La Grande (black dotted line) follows a distinct pattern from other rivers (for individual plots check Fig. S7). **B** Average discharge daily contribution (%) and standard error (in blue) based on (A) (La Grande not included)

The pattern of TSS and TP yields differed somewhat to that of DOC and TN but was consistent with each other (Fig. 5a, c, e, g), both varying at least one order of magnitude, with the highest TSS yields (up to $35 \text{ g TSS m}^{-2} \text{ year}^{-1}$) generally occurring in southern rivers and the lowest yields ($1.7 \text{ g TSS m}^{-2} \text{ year}^{-1}$) recorded in La Grande River. TP export yield also peaked in some southern rivers (up to $0.043 \text{ g TP m}^{-2} \text{ year}^{-1}$), and whereas lower in higher latitude

rivers, notably La Grande ($0.005 \text{ g TP m}^{-2} \text{ year}^{-1}$) (Fig. 5g). In contrast to yields, the absolute export of DOC and other constituents to the Bay was mostly a function of the magnitude of the river discharge (Fig. 5b, d, f, h). In fact, La Grande River was the largest exporter of DOC and nutrients to the Bay, despite having the lowest concentrations and export yields, yet it was not the largest exporter of TSS to the bay (Fig. 5, Table 1).

We estimate that the ensemble of rivers flowing into the Eastern coast of the James Bay export in the order of 2.52 (variation between first and third quartiles 1.50–4.74) and 1.77 Tg (1.61–1.88) of TSS and DOC, respectively, and 55,923 (48,924–62,812) and 4753 (3225–6867) tonnes of TN and TP, respectively (Table 3). About 92 to 95% of total riverine inputs are delivered by rivers sampled in the scope of this project, which altogether encompass about 93% of the total watershed area in the Eastern James Bay region.

Past and current scenarios

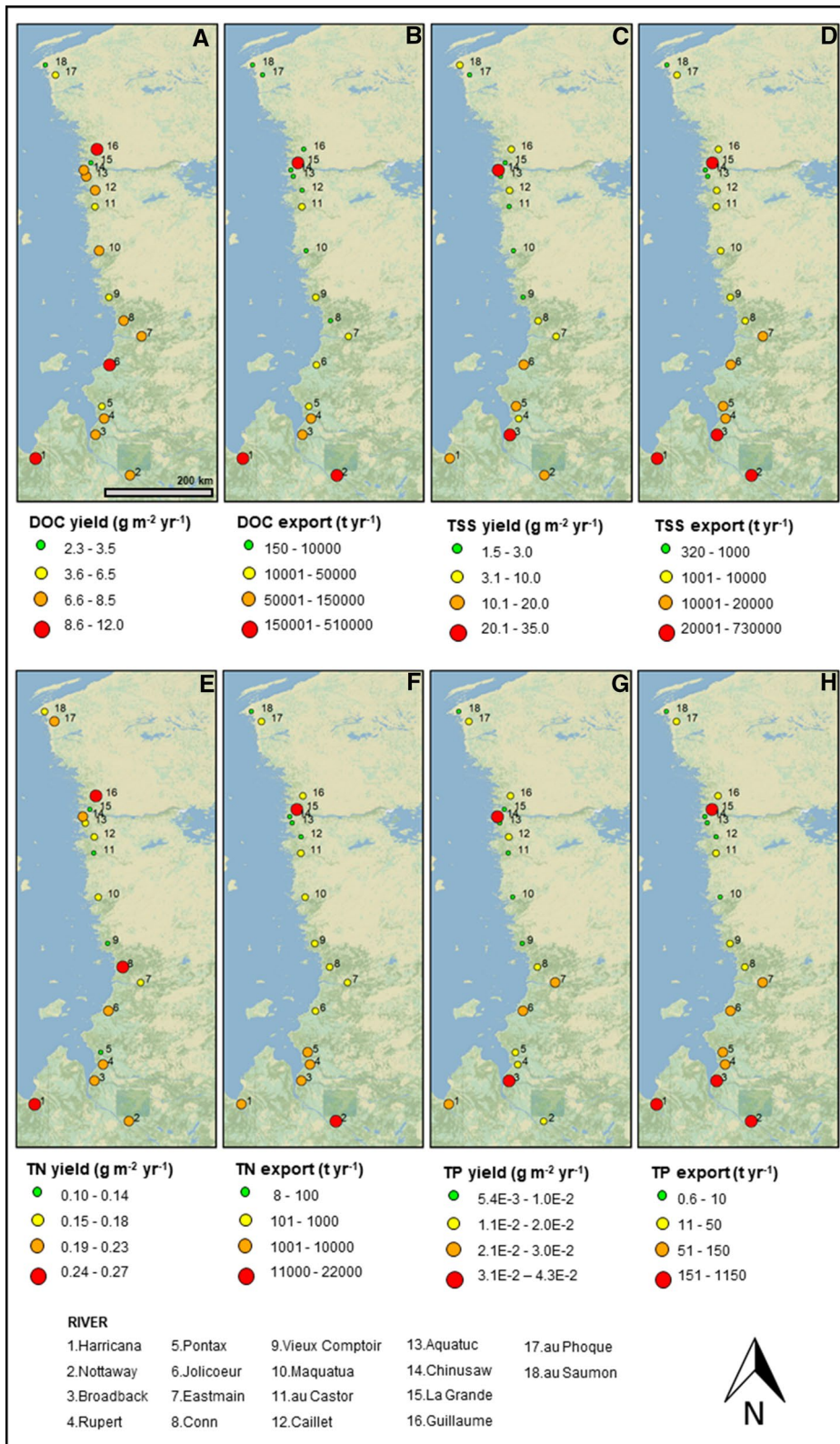
It is interesting to explore how riverine export to the James Bay may have been altered by the massive river damming and diversion that have occurred within watersheds since the 1970s, and for this it is necessary to contrast pre-damming and current river conditions. There were transient changes in physico-chemical water conditions during and immediately following flooding, including increases in C and nutrient concentrations (Tables S5–S7), often observed over the first years after reservoir flooding (Talbot et al. 2021), followed by a gradual return to pre-impoundment conditions within the first decade within La Grande River (Table S6). We estimate the overall annual pre-dam exports in 1978 (pre-impoundment) by La Grande, Eastmain, and Rupert rivers to have been in the order of 7.30×10^5 tonnes of DOC, 1.83×10^4 tonnes of TN and 1588 tonnes of TP, based on historical data. The uncertainties related with past exports could range in a similar interval estimated for current exports, mean percentage of variation between first and third quartiles (11.3% lower than mean and 12.4% higher than mean for DOC, 13.9 and 13.6% for TN and 27.1 and 28.5% for TP). Our data suggest that the current export by the same three rivers is in the order of 6.18×10^5 tonnes of DOC per year, 2.45×10^4 tonnes of TN and 1317 tonnes of TP (Table 4), which implies a decrease of 15% and 17% in DOC and TP export, respectively, and a 34% increase in TN relative to 1978 estimates. We therefore conclude that regional river exports of DOC and TP may have declined, and TN exports may have increased with the hydropower development in the region, assuming that there have been no major changes in export by the other free-flowing rivers in the region.

To further explore changes in material export to the James Bay over the past three decades, we estimated how much material the diverted waters currently load into La Grande watershed, by multiplying the volume of water diverted annually from each sub-watershed (Caniapiscau = $750 \text{ m}^3 \text{ s}^{-1}$, Eastmain-Opinaca = $830 \text{ m}^3 \text{ s}^{-1}$ and Rupert = $450 \text{ m}^3 \text{ s}^{-1}$) by their material concentrations (using historical data). We combined this with the estimated pre-dam export from La Grande watershed (first panel, Table 4), to derive a first order estimate of the total mass of material delivered by the watershed to the river and compared this with our current estimate of export by the river to the Bay. La Grande watershed currently delivers a significantly larger mass of material (8.73×10^5 tonnes of DOC, 2.25×10^4 tonnes of TN and 2067 tonnes of TP) than what La Grande river currently exports into the Bay (5.12×10^5 tonnes of DOC, 2.14×10^4 tonnes of TN and 1090 tonnes of TP) (Table 4), suggesting high DOC and P retention within the network, in the order of 41% and 47%, respectively, and a somewhat lower retention of TN (in the order of 5%).

Discussion

General trends in material concentrations

The large variability in carbon (DOC), suspended solids (TSS), and nutrient (TN, TP) concentrations encountered across our study rivers reflect differences in climate, watershed area, and topographic, geological and land cover features across watersheds, which influence the leaching patterns of materials from soils into aquatic systems. Our study area includes, in general, very pristine watersheds, with minimum human impact on land use as demonstrated by the low average percentages of watershed covered by settlements (0.24%), roads (0.07%), and croplands (0.18%), and high percentages of forest (56%), wetlands (19%), and surface water (mostly lakes, 10%). In these pristine environments, nutrient concentrations are less influenced by point-source anthropogenic sources (fertilizers used in agriculture, sewage and industrial effluents for example), previously shown to be important in watersheds worldwide (Jordan et al. 1997; Mattsson et al. 2005), and more influenced by natural sources, such as rock weathering, atmospheric deposition, and pulse events such as ice melt and rainstorms, which



◀**Fig. 5** Annual export and yield of **A, B** dissolved organic carbon (DOC); **C, D** total suspended solids (TSS); **E, F** total nitrogen (TN); **G, H** total phosphorus (TP) of the 18 sampled rivers flowing into the Eastern James Bay. Note that the position of La Grande mouth (15) has been shifted towards the bay to avoid overlap with other sites (sampling site at 53.7927° N, 78.8924° W)

promote leaching of accumulated materials from the catchment.

The concentrations of constituents measured in all rivers and within different river sections were overall co-related, but the degree of coupling varied between constituents. DOC and TN, and TP and TSS strongly co-variated with each other across the landscape, but DOC, TN and TSS were only weakly correlated. This indicates differential sources and processing within watersheds, acting either on loading from land to water or during hydrologic transport.

Material concentrations measured at the mouth of rivers represent the net balance between inputs from soil export, autochthonous production, and fixation (for DOC and TN), and removal through various biogeochemical processes including microbial and photo-degradation, sorption to mineral particles and burial in sediments. All four constituents were positively related to the various types of wetland coverage within the watershed, suggesting that this particular land feature is important in mediating the transfer (or storage) of materials from land to aquatic networks. This relationship was particularly strong for DOC, which agrees with previous studies that demonstrated that the presence of wetlands and peat cover can increase C concentrations in northern rivers and streams (Aitkenhead and McDowell 2000; Creed et al. 2008; Laudon et al. 2011). There are some reports of the influence of wetland coverage on riverine TN, TP and TSS, showing both, increasing N and P concentrations with increased wetland coverage (Pellerin et al. 2004; Rutledge and Chow-Fraser 2019), and negative relationships, identifying wetlands acting as traps of nutrients and sediments (Jordan et al. 2011; Lane et al. 2018; Prior and Johnes 2002; N retention by wetlands Saunders and Kalff 2001), the latter not being the case in these boreal landscapes.

The strong negative relationship found between both DOC and TN with the percentage of water surface within the watershed, also observed in other boreal regions for DOC and for Hg (Burns

et al. 2012; Fink-Mercier et al. 2022) reinforces the important role of lentic ecosystems as sinks of materials originating from land. It also suggests that the network-scale removal of DOC and TN is a key driver of these two constituents in these boreal rivers, certainly stronger than the source term. It is interesting to note that TP only had a very weak relationship to the proportion of water in the watershed, suggesting that its patterns of processing and removal are very different from those of DOC and TN. TP was tightly related to TSS ($R^2=0.67$), likely due the strong affinity of phosphorus to clay (House et al. 1998), which predominates in this region (the Great Clay Belt, Dresser 1913).

It is well recognized in the literature the challenges associated with the quantification of suspended solids in rivers because it is highly temporally variable and influenced by multiple factors (Mueller and Pitlick 2014; Bywater-Reyes et al. 2017). TSS concentration is generally a function of both sediment supply and transport capacity (Bywater-Reyes et al. 2017), with soils representing the main source of suspended sediment to the water, being smaller and lighter soil aggregates more easily transported over long distances in aquatic systems during intense transport events such as rainstorms and snowmelt. In this way, lithology and geomorphology would play an important role in determining TP and TSS leaching from land into aquatic systems, as we observed in our Elastic models where brunisolic soils and volcanic rocks appeared as drivers of such materials. After entering the water, these materials are transported together (the strong relationship between TP and TSS was observed across all river sections) along the river continuum and delivered to the ocean without much significant loss to burial, and thus the absence of strong relationships with the proportion of water in the watershed. The fact that we found much greater variability in their concentrations (and higher uncertainty as demonstrated by range variability in the first and third quartiles of Monte Carlo propagation) across watersheds compared to DOC and TN (Table 1) suggests that climate and hydrologic events may be associated to this large variability observed in TSS and TP concentrations (further discussion in “Climate change and riverine export” section). These include rainstorms during summer and fall and snowmelt during the spring, which enhance terrestrial erosion, leaching

Table 3 Regional estimates of total annual export of materials (plus first and third quartiles calculated by Monte Carlo propagation) to the coast of the Eastern James Bay for sampled and unsampled (based on models described above) rivers

	Values	DOC export (Teragrams year ⁻¹)	TSS export (Teragrams year ⁻¹)	TN export (Tonnes year ⁻¹)	TP export (Tonnes year ⁻¹)
Sampled rivers	Mean	1.62	2.40	52,506.55	4403.02
	1st quartile	1.48	1.43	45,981.98	2969.94
	3rd quartile	1.71	4.55	58,931.51	6417.85
Unsampled rivers	Mean	0.15	0.12	3416.00	349.60
	1st quartile	0.13	0.07	2942.20	254.72
	3rd quartile	0.17	0.19	3880.58	449.34
Regional	Mean	1.77	2.52	55,922.55	4752.62
	1st quartile	1.61	1.50	48,924.18	3224.66
	3rd quartile	1.88	4.74	62,812.09	6867.19

Table 4 Past and current estimates of riverine exports by La Grande, Eastmain-Opinaca and Rupert watersheds into James Bay, and the contribution of the main watersheds diverted into La Grande complex by this hydropower megaproject

	Watershed	Dis-charge (m ³ s ⁻¹)	Water export (km ³ year ⁻¹)	DOC export (t year ⁻¹)	TN export (t year ⁻¹)	TP export (t year ⁻¹)
Pre-damming/diversion export to James Bay	La Grande (1979)	1700	54	3.43E+05	8.04E+03	483
	Eastmain-Opinaca (1980)	910	29	2.29E+05	4.87E+03	830
	Rupert (2009)	845	27	1.58E+05	5.38E+03	275
	Total			7.30E+05	1.83E+04	1588
Input to La Grande complex	Eastmain-Opinaca	830	26	3.41E+05	7.07E+03	1204
	Caniapiscou (1982)	750	24	1.04E+05	4.48E+03	234
	Rupert	450	14	8.40E+04	2.87E+03	146
	Total			5.29E+05	1.44E+04	1584
Post-damming export to James Bay	La Grande	3780	119	5.12E+05	2.14E+04	1091
	Eastmain-Opinaca	65	2	2.68E+04	5.47E+02	97
	Rupert	395	12	7.94E+04	2.55E+03	130
	Total			6.18E+05	2.45E+04	1317

The year in parenthesis indicates the corresponding year of impoundment or diversion. Variability in these past exports could range (first and third quartiles, respectively) between 11.3% less and 12.5% more than DOC annual mean, 13.9 and 13.6% for TN, and 27.1 and 28.5% for TP, based on current patterns estimated from the 18 sampled rivers

and streamflow transport, and therefore mobilize soils and river sediments rich in both particles and TP. In contrast, these events are rare during winter, when we found overall lower TSS and TP concentrations.

Material yields and export fluxes to the James Bay

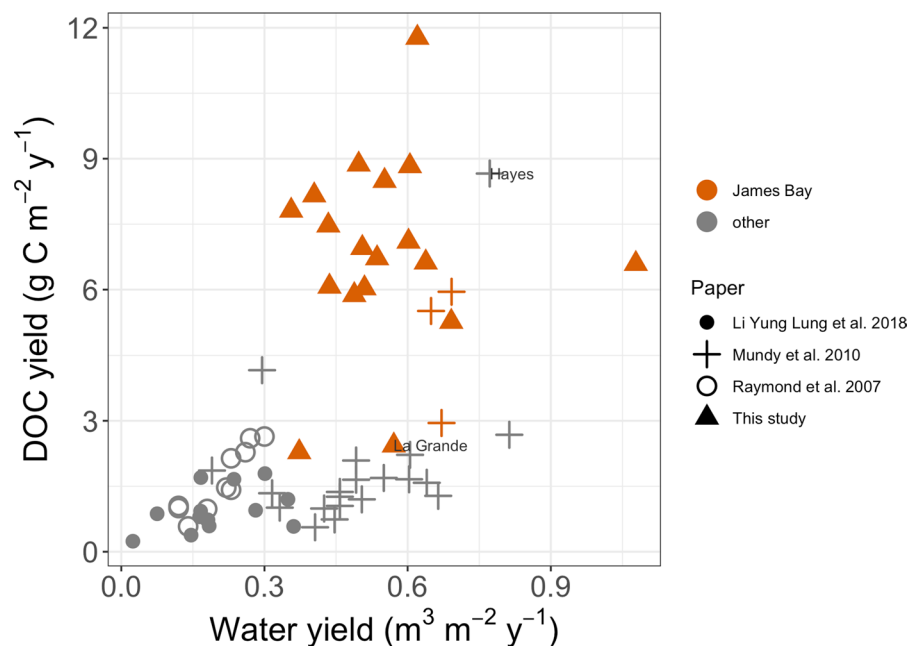
The range in material concentrations across rivers together with watershed areas resulted in a substantial range in material export yields, which represent export fluxes (g of material per year) normalized by

watershed area (m⁻²); there was no clear pattern of export yield with watershed location or size, being higher in both extremes of the spectrum of river sizes and discharges, as well as in latitude (Fig. 5). In a study of impacts of land use cover and climate on the export of organic C and nutrients from boreal Finnish catchments (covering 88% of the total area of Finland, Mattsson et al. 2005), it was found that TOC yields ranged between 1.2 and 7.1 g C m⁻² year⁻¹ (over 90% in the dissolved form, DOC), TN between 0.08 and 1.1 g m⁻² year⁻¹, and TP from 1 to

$90 \times 10^{-3} \text{ g m}^{-2} \text{ year}^{-1}$. This Finnish study is among the few to present both DOC concentration and flux data, and the watersheds involved generally resemble the ones in our study in terms of a high proportion of land covered by peatlands, coniferous forests, and lakes. Our estimates of DOC and TP yields of 2.3 to $11.8 \text{ g m}^{-2} \text{ year}^{-1}$, and 5.4 to $43 \times 10^{-3} \text{ g m}^{-2} \text{ year}^{-1}$, respectively compare well with the estimates of Mattsson et al. (2005). The range in DOC and TP concentrations was also similar, 5.2 to 23.4 mg C L^{-1} compared to 5.1 to 21.0 mg C L^{-1} (TOC, Mattsson et al. 2005), and 6.5 to $247 \text{ } \mu\text{g P L}^{-1}$ compared to 5.0 to $180 \text{ } \mu\text{g P L}^{-1}$ in Finnish watersheds, respectively. Our results for TN yields, however, were lower and less variable (0.10 to $0.27 \text{ g m}^{-2} \text{ year}^{-1}$) compared to estimates in Finland, which is probably due to the lower range in TN concentrations observed in our study (James Bay: 147 to $714 \text{ } \mu\text{g L}^{-1}$; Finland: 250 to $3500 \text{ } \mu\text{g L}^{-1}$). The presence of extensive agricultural land in the Finnish landscape may explain these differences, since there was a strong relationship between total riverine N yield and the percentage of agricultural land in the Finnish watersheds (Mattsson et al. 2005). This highlights the differential impact that human activities and changes in land use may have on riverine export of N, P and DOC to the oceans.

To place rivers of the James Bay region in a more global context, we compared the DOC yields from our studied rivers with other rivers draining into the Arctic Ocean, as a function of water yield (Fig. 6). This includes the largest rivers draining the continent as well as a range of smaller rivers. For any given water yield, the James Bay rivers have consistently higher DOC export yields (by an average of two to threefold) relative to all rivers studied along the boreal/arctic region of the America and Eurasia continents, with the exception of La Grande River, whose DOC yield falls within the range reported for the other rivers ($2.5 \text{ g C m}^{-2} \text{ year}^{-1}$). A previous study had also signaled several rivers from the southern portion of the Hudson Bay as having very high DOC yield (Mundy et al. 2010). Another large scale study, which estimated water and DOC fluxes in a wide range of watersheds draining $188,829 \text{ km}^2$ of the Western coast along southeast Alaska, northwest British Columbia and southwest Yukon Territory in Canada, also reported high DOC yields from a wide range of watersheds, from 0.5 to $60 \text{ g C m}^{-2} \text{ year}^{-1}$, with highest values corresponding to the smallest watersheds (Edwards et al. 2021); the overall mean DOC yield for this region of $6.2 \text{ g C m}^{-2} \text{ year}^{-1}$ agrees well with our own observations for the James Bay, suggesting that there are other Northern regions of comparably high organic C yields. We conclude

Fig. 6 Comparison of the relationship between annual average water and DOC yields between studies in rivers in arctic and subarctic regions, including the present study in the Eastern James Bay (in orange). The symbols indicate the study from which the data was retrieved



that these boreal watersheds draining into the Eastern James Bay, which are characterized by a high peatland coverage and organic-rich soils, represent hotspots of DOC export to the Hudson Bay system, which likely contribute significantly to the terrestrial C reservoir of the Arctic Ocean.

Material export to the Bay was essentially driven by discharge, and the large rivers were responsible for the bulk of the material export (Fig. 5, Table S4). In fact, there has been a greater focus on the role of these large rivers in regional and global budgets (e.g., Nottaway, Broadback, La Grande) (Benner et al. 2004; Mundy et al. 2010). However, our results clearly indicate that small and medium rivers may have very high yields and can generate local hotspots of DOC, TSS, or nutrients within coastal habitats, which have major implications to the functioning of these ecosystems at a more local or collectively at a regional scale. It is not surprising that material yields were higher in these small to medium watersheds, as usually material concentrations and export yields are high in streams and small rivers (Ågren et al. 2007; Mattsson et al. 2005) since they have a closer interaction with soils and have typically lower proportion of lakes and shorter residence times and therefore smaller sink terms.

Although La Grande contributes with more than half of freshwater inputs to the Eastern James Bay (53%), it contributes proportionally less to the organic and inorganic material exports (mean): about 32% of total DOC export, 15% of TSS, 40% of TN and 26% of TP. La Grande river is nevertheless the largest single contributor of total riverine export of DOC, TN and TP to the Bay, yet there are other rivers that contribute individually more to TSS export (Harricana, Broadback and Nottaway, Fig. 5). This is because La Grande River has extremely low TSS yields whereas Southern rivers in the region are under the influence of the Great Clay Belt (Dresser 1913), resulting in extremely high suspended sediment loads in these rivers. La Grande in fact had the lowest material export yields in the region, which is typical for rivers draining very large and heterogenous watersheds with complex aquatic networks and often large coverage of lakes. These systems with extended residence time offer geophysical opportunities for microbial processing of organic matter and loss of materials via sedimentation during downstream transport (Battin et al. 2008).

In the case of La Grande River, damming has profoundly altered the architecture of the fluvial network and in particular, it has added significant amounts of standing water to the network: the original La Grande watershed had about 15% total surface water coverage, which was already extremely high, and damming has further increased that to the current 24% (Fink-Mercier et al. submitted). This alteration of the natural flow of the river by damming increases standing water and overall residence time and is likely to result in net increase in nutrient and organic matter retention and favor particle settling and accumulation of sediments (Teodoru et al. 2013; van Cappellen and Maavara 2016), further lowering the overall material yields of the river to the Bay. Larger absolute amounts of C and nutrients are likely to be retained in systems that are naturally more turbid or that have higher nutrient concentrations such as eutrophic reservoirs due to higher loads (Maavara et al. 2020). However, C and nutrient transport and elimination can be affected by multiple factors. For example, remineralization and gaseous C elimination (emissions) from reservoirs in tropical regions are generally higher than emissions in boreal and temperate reservoirs, partially due to their large surface areas, high amounts of flooded biomass and warmer water temperatures which accelerates mineralization rates (Barros et al. 2011).

We estimate the total fluvial export from the ensemble of rivers draining the Eastern James Bay at around 1.77 (1.61–1.88) Tg of DOC per year, which is in broad agreement with a previous estimate for the fluvial export for the entire Hudson Bay system in the range of ~5.5 Tg year⁻¹ (Mundy et al. 2010). However, we highlight that DOC fluvial export was likely underestimated in this previous study, which included only 3 rivers located in Eastern James Bay (Rupert, Broadback, and Nottaway), totalling less than 0.6 Tg C year⁻¹. Moreover, that study did not include rivers flowing in the Western region of the James Bay, namely Moose, Albany, Attawapiskat, and Ekwan rivers. To the best of our knowledge, there is no other study that has estimated C (or nutrient) riverine exports into the James Bay as a whole, nor a regional budget encompassing all watersheds within the Hudson Bay system (Eastern and Western regions). In this regard, our study provides data from small to medium sized rivers, which are rarely accounted for in regional and global riverine budgets. Furthermore,

we were able to include unsampled rivers in this estimate and generate a regional estimate that encompass all rivers flowing into the Eastern James Bay by using empirical models of discharge and material concentration.

Past and current scenarios

The massive decrease in discharge in Eastmain and Rupert rivers caused by the diversion of their flows into La Grande reservoir complex in the last decades, and the subsequent increase in riverine exports by La Grande represents shift in the location of riverine inputs further up North in the Bay, whereas the contributions from the diverted Caniapiscau River (which naturally flows into Ungava Bay through the Koksoak River) represents a net increase of freshwater inputs in the order of $750 \text{ m}^3 \text{ s}^{-1}$ (Roy and Messier 1989) and therefore also a net increase of water and materials to La Grande watershed and to the Bay as a whole. The changes in material export to the James Bay over the past three decades occurred in the context of a combination of increases in net water discharge and material loading and selective removal or addition of materials within La Grande watershed.

From our results we can conclude that damming and diversion have led to change in the elemental stoichiometry of C:N:P, and to a generalized decrease in export of DOC and nutrients, and other materials, including mercury (Fink-Mercier et al. 2022) to the James Bay, despite the fact that there has been a net increase in the amount of water discharging into the Bay (especially higher during winter season), and of materials entering La Grande watershed. These large hydropower reservoirs now act as net sinks of C and P (Maavara et al. 2017, 2020), which are likely to be stored in sediments, decomposed, and emitted as greenhouse gases. For N, the low apparent retention could indicate uncertainties in the mass balance, or more likely the possibility that damming may enhance N mobilization or biological fixation within lentic systems.

Climate change and riverine export

Climate change is predicted to be more pronounced in boreal and arctic regions of the globe with rising concerns about the major impacts of climate and land use changes on the functioning and stability of the large

global C sinks that exist in these landscapes (Gibson et al. 2018; Zhang et al. 2008). One of the most significant effects of climate change in these regions is likely to be in the timing and magnitude of the spring peak flow associated with melting of the winter snowpack (Billett et al. 2012; Gagnon and Gough 2005), and increases in precipitation which mobilize terrestrial materials from peatlands. Short-term ice break-up events are likely to increase, which can generate a pulse of material loads to coastal systems at a much higher intensity and speed in spring, which follows winter, a period characterized by lower freshwater and in turn material loads. For the Arctic Ocean, an overall increase in river discharge in the years of 2020 and 2021 was observed relative to the reference period (1981–2010), and these high discharges seem to be related with early melt of a large snowpack in spring and unusual wet summer, which further evidence for the intensification of the arctic hydrologic cycle (Peterson et al. 2002).

In fact, several long-term studies on DOC dynamics in Canada (Couture et al. 2012; Houle et al. 2020) and elsewhere (Arvola et al. 2010; Findlay 2005; Keller et al. 2008) have reported increasing terrestrial DOC hydrologic transport, a process that has been termed “browning” (Roulet and Moore 2006), which has been shown to affect the biogeochemical and biological functioning and productivity of these systems (Kritzberg et al. 2020). Based on hydrologic projections for Québec (Guay et al. 2015) and long-term trends in DOC concentrations measured across North America (Couture et al. 2012; Houle et al. 2020; Keller et al. 2008), we generated a projection of DOC loads by rivers into the James Bay coast by the year of 2050, assuming that these trends will continue. We assumed a conservative scenario of 10% increase (lower limit from Guay et al. 2015) in annual streamflow for 2050 from a model based on gauged watersheds (monitored by hydrometric stations) concurrent with meteorological data in a time series of maximum 45 years, encompassing watershed areas varying between 10 and 69,000 km^2 (Guay et al. 2015). For DOC, we assumed a conservative annual increase of 0.02 mg per liter based on the average value in DOC increase rate for undisturbed lakes in eastern Canada over a period of minimum 3 decades (Houle et al. 2020), which is in accordance with other studies across North America (Keller et al. 2008; Couture et al. 2012). We choose a conservative scenario due

the fact that the James Bay territory spreads over a less disturbed region of Québec and that a portion of DOC is retained in the water reservoirs further lowering DOC concentrations downstream in the river. We project that for the year of 2050, rivers flowing into the James Bay will transport a total amount of 250 km³ of water per year and 2.12 Tg DOC year⁻¹ (1.95 Tg by sampled rivers), which represents an increase of 20% in DOC export (~0.35 Tg, uncertainty of around ± 10%).

We observed a much greater variability for TSS and TP concentrations within and across watersheds and seasons than for DOC and TN. The challenge of predicting riverine TSS and TP concentrations suggests that pulse events like snowmelt in spring and rain events could underlie a significant portion of variability in TP and TSS concentrations, however capturing these temporal events requires high frequency temporal studies, which are hampered in such remote watersheds. In fact, rainstorms, and wildfires, which were not captured in our study, have been shown to influence soil and water properties (Lawler et al. 2006; Lewis et al. 2019; Murphy et al. 2012) and despite their short duration relative to geological time scales, they can substantially increase runoff, rock weathering and in turn increase mobilization of sediment and organic C into surface waters (Shakesby and Doerr 2006). The consequences of these drastic events can persist for many years after the event depending on their areal extent, topological and geological properties within watersheds, and post-event climate conditions (Loiselle et al. 2020). Wildfires are key features of the boreal biome of Canada (Hutchins et al. 2020; Wang et al. 2014) and their incidence is expected to increase in the coming decades (Coogan et al. 2019). In turn, precipitation, and frequency of storms in these regions are predicted to increase under climate change scenarios as well (Guay et al. 2015) and contribute significantly to the loading of particles and other materials to aquatic systems. A more systematic sampling approach could unveil the effect of these point events on material fluxes, although the remoteness of these northern watersheds imposes challenges to field sampling. Future studies should adopt a multiple strategy using high frequency probes, remote sensing, and hydrological models to address the importance of these high-intensity events to the flux of materials from land to ocean.

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Author contributions PAdG developed the project and acquired the funds. PAdG, MLG and CFM conceptualized and carried out sampling. MLM, MLG and CFM worked on data acquisition and ArcGIS analyses. MLM carried out statistics, plots, and maps. MLM and PAdG discussed the results and wrote the manuscript. All authors reviewed, edited, and approved the final version of the manuscript.

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Data availability Environmental data is available online (<https://borealisdata.ca/dataset.xhtml?persistentId=doi:10.5683/SP3/7EQN2S>).

Declarations

Conflict of interest The authors declare no conflict of interest.

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