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# Dissolved organic matter quantity and quality from freshwater and saltwater lakes in east-central Alberta

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**Abstract.** Concentrations of dissolved organic matter (DOM) in surface waters of sub-humid to semi-arid lakes in east-central Alberta increase with increasing salinity and water residence time from about 20 to 330 mg $L^{-1}$ as dissolved organic carbon (DOC). This pattern is opposite to that observed among freshwater lakes spanning a gradient in water residence times, and is probably caused by evaporative concentration of refractory DOM. The proportion of total DOC, operationally defined as humic substances using XAD-8 resin, was high, though similar to surface waters typically referred to as "humic", and independent of salinity. Very long water residence times (hundreds of years) in saline lakes favors evapoconcentration of low-color, low molecular weight DOM, with N-content characteristic of allochthonous DOM.

**Key words:** dissolved organic carbon, dissolved organic nitrogen, humic substances, saline

## Introduction

Dissolved organic matter (DOM – typically measured as dissolved organic cabon; DOC) affects the transparency and pH of water, the fate and bioavailability of trace nutrients and contaminants, and the supply of energy to pelagic microbial foodwebs (Schindler et al. 1992, and others from this volume). DOC is composed of organic material leached and excreted from terrestrial and aquatic organic matter. Components of the DOC pool are largely unknown but many operational separations have been made using reversible sorption onto a variety of resins. Humic and fulvic acids are typically the most abundant of these forms comprising on average 50% of total DOC (Malcolm 1985).

Aquatic systems vary in the relative contribution of DOC from the catchment (allochthonous) and of DOC produced within the system (autochthonous). Allochthonous DOC is typically characterized as enriched in humic substances and highly colored with decreasing in color (absorptivity) from wetlands to streams and rivers, to groundwater (Thurman 1985). Highly stained, DOC-rich lakes are often referred to as "humic lakes" (eg. Gjessing 1992). In contrast, autochthonous DOC is thought to be lightly or uncolored (Tipping et al. 1988), and enriched in nitrogen (McKnight et al. 1994). Whole-lake radiocarbon experiments in Precambrian Shield drainage lakes reveal that the DOC pool does not equilibrate with other carbon pools, indicating that most of the DOC pool can be allochthonous (Hesslein et al. 1980; Schindler et al. 1992), and that most of the autochthonous DOM is labile (Hesslein et al. 1980). In seawater, DOC is thought to be mainly autochthonous (Hedges et al. 1992), but the DOC pool in estuarine and nearshore waters can be dominated by riverine inputs (Yan et al. 1991).

Estimates of DOM mass balance for lakes indicates significant loss of DOC within lakes (Schindler et al. 1992). Unfortunately, loss processes have not been well quantified. DOC can be removed by flocculation (Kepkay & Johnson 1989; Urban et al. 1990), microbial degradation (Tranvik 1989; Hessen 1992), and photolysis (Vallentine & Zepp 1993; Kieber et al. 1990). Individual processes probably remove different portions of the total DOC pool. The relative importance of each process is currently not known for any system and probably varies among systems because DOC source materials and physicochemical properties of the systems differ.

In humid regions, empirical studies and mass balance analyses show that concentrations of DOC in lakes tend to decrease with increasing water residence time. For example, loss of DOC was greatest during prolonged dry periods with longer water residence times (Schindler et al. 1992). Similarly, color (often used as a proxy for organic carbon) is inversely related to water residence time for headwater lakes (Meili 1992), and directly related to the relative drainage area among lakes (Engstrom 1987; Rasmussen et al. 1989).

Inland saline lakes in semi-arid to arid climates (athalassic saline lakes) violate the pattern of decreasing DOM concentration with increasing water residence time. In contrast to freshwater lakes in humid regions, the concentration of DOM in surface waters of semi-arid regions increases with proxy estimates (salinity) of water residence time (Curtis & Prepas 1993). These lakes contain some of the highest reported concentrations of dissolved organic carbon. High pH in these waters enhances the solubility of humic and fulvic acids (DeHann 1992; Aiken & Malcolm 1987), but high ionic strength, shallow depth, high bacterial biomass and dissolved nutrient concentrations in combination with long lake residence times (Campbell & Prepas 1986) favor the efficient removal of DOC.

The purpose of this study was to compare quantities and qualities of DOM among lakes with different salinities and water residence times, and attempt to identify processes that were consistent with observed patterns. First, concentrations of DOC were compared among surface waters and groundwaters from semi-arid east-central Alberta. Concentrations of dissolved organic nitrogen (DON) were compared among a smaller set of surface waters. Second, qualitative properties of DOM were characterized using XAD-8 resins, ultrafiltration, and spectrophotometric analysis. Because of the previously observed trend of increasing DOC concentration with increasing salinity (Curtis & Prepas 1993), we hypothesized that qualitative properties of the DOM would be linked systematically to salinity among lakes. Third, quantities and qualities of DOM from our region were compared with values reported in the literature for DOM from humid regions, and from seawater.

## Site description

The 23 lakes sampled were located along a north-south transect in the region of transition between aspen parkland and grassland prairie, from sub-humid to semi-arid climatic zones, and from external to internal drainage (Fig. 1). Most of the land in this region is mixed agricultural. The major crops are cereal grains, oil seeds, and forages. A smaller proportion of the land is used for grazing by cattle.

All lakes were shallow, generally having maximum depths between 0.3 and 3.0 m. The range in lake area was from a few hectares to tens of square kilometers. Dominant ions in the lakes of this region are Na-$SO_4$-$CO_3$, and most of them are alkaline. We define alkaline arbitrarily as waters having pH greater than pure water in equilibrium with atmospheric carbon dioxide and with calcite solubility (pH 8.4 at 1 atm and 25 °C with $pCO_2 = 10^{-3.5}$; Stumm & Morgan 1981). The range in pH among lakes was 8.1–9.9. Lake area was apparently unrelated to either salinity or pH.

Groundwaters were sampled around the margins of lakes, at sites of conspicuous seepage. The ecoregions, surficial geology, and basic properties of lakes of Alberta are described elsewhere (Mitchell & Prepas 1990). Factors correlating strongly with salinity are relative elevation of the lake within the catchment, and excess precipitation (both negative; Last 1992).

Lakes were operationally defined as saline when concentrations of total dissolved solids (TDS) exceed 3 g $L^{-1}$ (cf. Williams 1964). Saline lakes tend to be seepage lakes, and have very long water residence times (often >100 y). They occur in regions with negative excess precipitation ($P < E$), and where hydrologic inputs are balanced mainly by evaporation and to a lesser extent out-seepage. Where flushing rates are very low, soluble salts evapoconcentrate and the water becomes saline.

## Methods

Surface water samples were collected in polyethylene bottles from 23 lakes during the summer months of 1992 and 1993. Inverted bottles were immersed

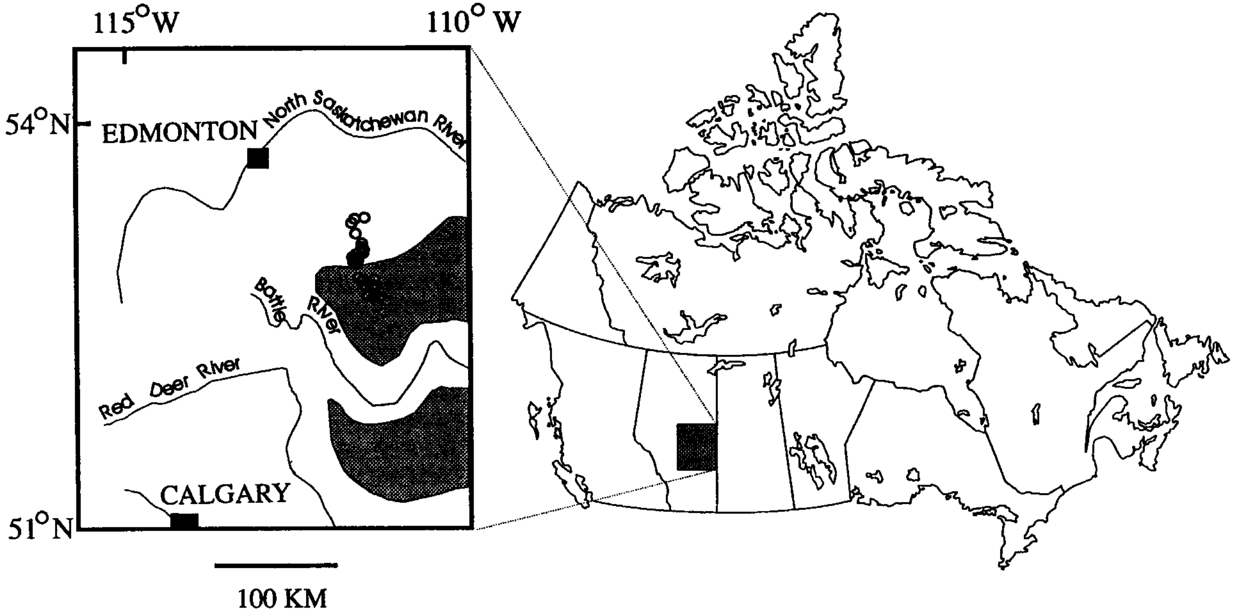

*Fig. 1.* Map of the study region (inset Canada) in east-central Alberta showing major rivers, areas of internal drainage (grey) and study sites (open circles). Adapted from Last (1992).

by hand and filled at a depth of about 0.2 m depth. Groundwater was sampled by hand from pits excavated using a shovel or by vacuum pump from piezometers. All samples were transported to the lab in darkness at approximately ambient temperature. Unfiltered subsamples were analyzed for pH and specific conductance using a Beckman Φ12 pH/ISE meter and Radiometer CDM 2e′ conductivity meter, respectively. Lakes were classified as being fresh (TDS $< 3\text{g L}^{-1}$) or saline (TDS $> 3\text{g L}^{-1}$) from specific conductance using a regression of specific conductance and total dissolved solids (TDS) for 75 lakes in east-central Alberta (calculated from data in Mitchell & Prepas 1990; $R^2 = 0.99$, $P < 0.01$). The range in salinity among samples was from 1–84 $\text{g L}^{-1}$ TDS or from 3–240% the salinity of seawater (TDS 35 $\text{g L}^{-1}$). Hereafter the relative content of salts in water will be expressed in units of specific conductance ($\text{mS cm}^{-1}$).

### *Sample preparation*

Aliquots of samples were filtered through precombusted GF/F filters in the laboratory, usually within 24 hours. Filtrates were stored in a dark refrigerator until further characterizations of the organic matter were made. Thus, all materials discussed in this paper are operationally defined as dissolved.

Aliquots of filtrate were ultrafiltered using polysulfone tangential flow cartridges (Filtron) with a molecular weight cutoff of 1,000 Daltons. The volumes of filtrate and retentate were measured and subsampled for DOC analysis.

Aliquots of GF/F filtered water were acidified to pH 2, passed through a column filled with XAD-8 exchange resin, and then eluted with 0.1N NaOH. The eluted organic materials were operationally defined as fulvic and humic acids (Thurman & Malcolm 1981).

Organic carbon content was measured in subsamples and fractions using a high temperature catalytic oxidation (HTCO) organic carbon analyzer (Ionics, 1555). The furnace temperature was 850 °C, the carrier gas was ultra high purity oxygen (Linde), and the catalyst was platinum (Ionics). Samples were acidified to pH $<2$ and sparged for three minutes with oxygen. Subsamples (60 $\mu$L) were injected into the furnace by an autosampler. Freshwater samples were analyzed directly, whereas saline samples were diluted with deionized water before analysis because undiluted samples foamed excessively during acidification and sparging.

Dissolved organic nitrogen (DON) concentrations in GF/F filtered subsamples were calculated by difference from total dissolved nitrogen (TDN) minus the sum of the inorganic forms ($NO_3^-$, $NO_2^-$ and $NH_4^+$) similar to Schindler et al. (1992). TDN, $NO_3^-$ and $NO_2^-$ were analyzed by techniques described

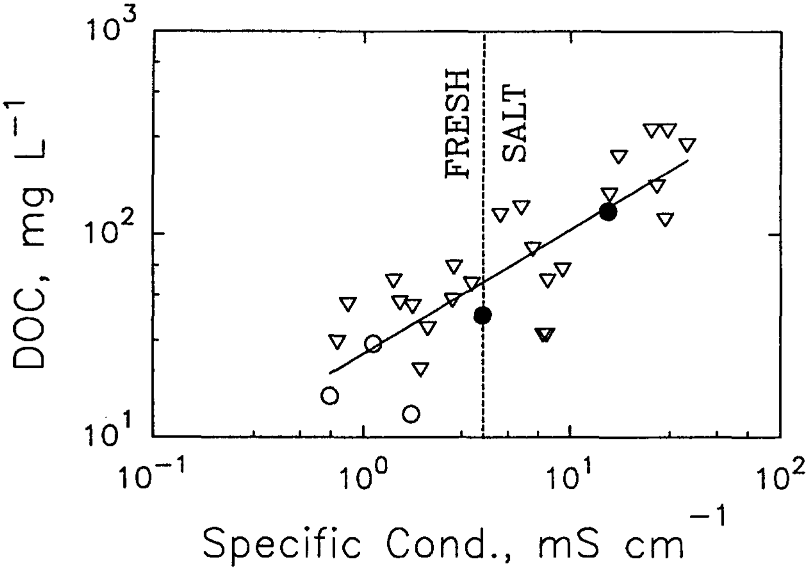

*Fig. 2.* DOC concentration at measured specific conductance in surface waters (open triangles) and near-surface groundwaters (open circles) collected at the study sites. Closed circles indicate groundwater samples of seepage between nearby lakes. The solid line is the linear regression line ($R^2 = 0.70$, $P < 0.01$). To the left of the dashed line waters are fresh, to the right waters are saline (after Williams 1964).

by Stainton et al. (1977). $NH_4^+$ was analyzed by techniques described by Solorzano (1969).

The relative color of organic matter was calculated as a specific absorption coefficient (SAC) by equation 1.

$$SAC = \frac{(2.303 \times Abs_{340})/(pathlength)}{[DOC]} \tag{1}$$

with units of cm$^2$ mg$^{-1}$, and mg cm$^{-3}$ for SAC and the concentration of DOC, respectively. Absorbance of filtered water was measured spectrophotometrically at 340 nm using a Milton Roy 501 spectrophotometer and a 10 cm path length.

## Results

The concentration of DOC in surface waters increased directly with increasing specific conductance (Fig. 2; $R^2 = 0.70$, $P < 0.01$) from 22–330 mg L$^{-1}$. Concentrations of DOC and specific conductances in samples of groundwater

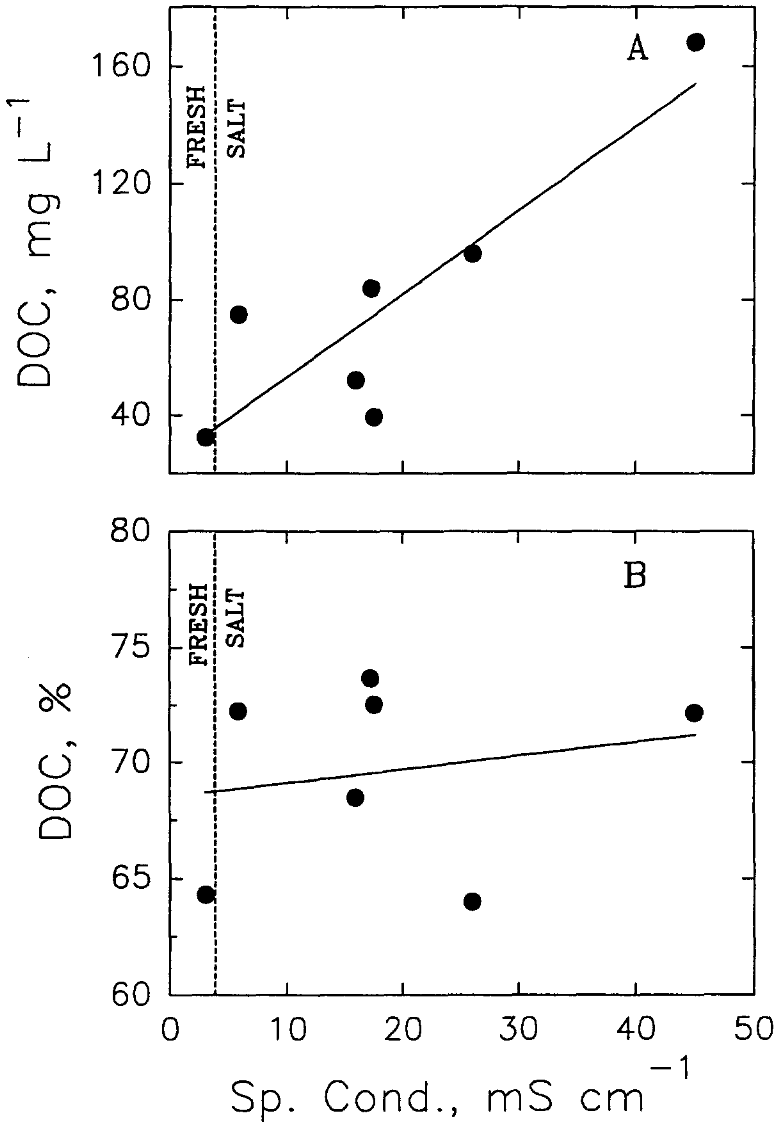

*Fig. 3.* a. The concentration of humic DOC at the specific conductance of the water sample ($R^2 = 0.76$, $P < 0.01$). b. The proportion of humic DOC ($R^2 = 0.004$). Solid lines are linear regressions and fresh and saltwater are separated by the dashed line as in Fig. 2.

were lower than in most of the surface waters (Fig. 2). Two of the groundwater samples were collected from sites between saline lakes that were separated by only a few tens of meters. Consequently, the source of DOC and ions for these samples was probably a neighbouring lake.

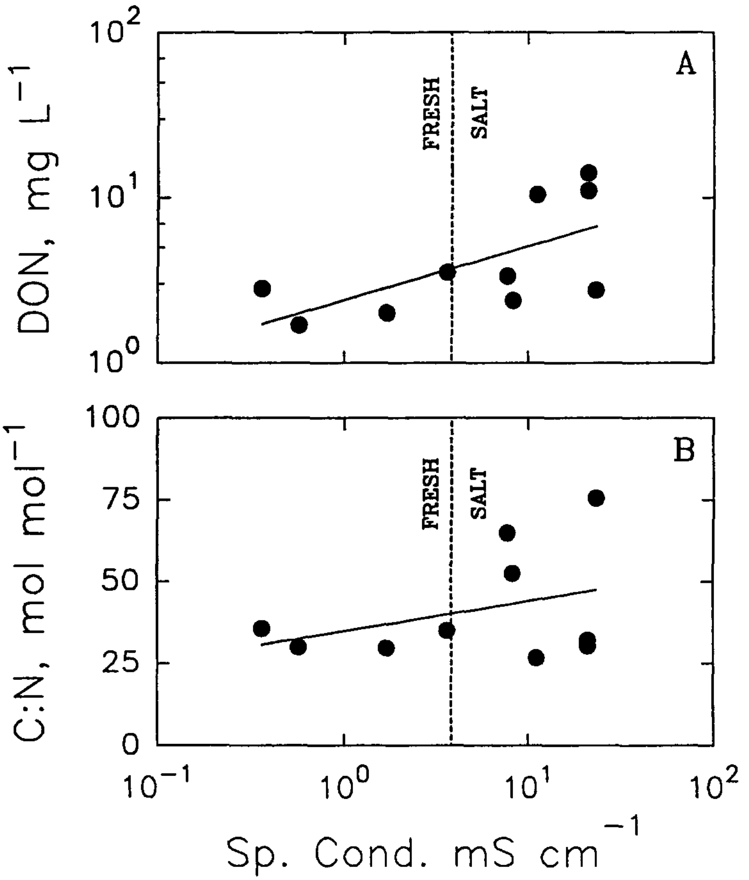

*Fig. 4.* a. The concentration of dissolved organic-N (DON) at measured specific conductance values ($R^2$ = 0.41, $P < 0.05$). b. The molar ratio of C:N in DOM ($R^2$ = 0.10). Solid lines are linear regressions and fresh and saltwater are separated by the dashed line as in Fig. 2.

The concentration of humic substances, defined operationally by reversible sorption on XAD-8 macroporous resin, increased linearly with increasing specific conductance (Fig. 3a; $R^2$ = 0.76, $P < 0.01$). The proportion of total DOC as humic substances was independent of specific conductance (Fig. 3b; $R^2$ = 0.004). Approximately 70% of DOC from all lakes was reversibly sorbed on XAD-8 resins.

Concentrations of dissolved organic nitrogen increased significantly from 2 to 14 mg L$^{-1}$ with increasing specific conductance, (Fig. 4a; $R^2$ = 0.41, $P < 0.05$), similar to the pattern for DOC (Fig. 2). Proportions of dissolved inorganic nitrogen were between 5 and 40% of TDN and were independent of specific conductance ($R^2$ = 0.003). The atomic ratio of C:N of dissolved

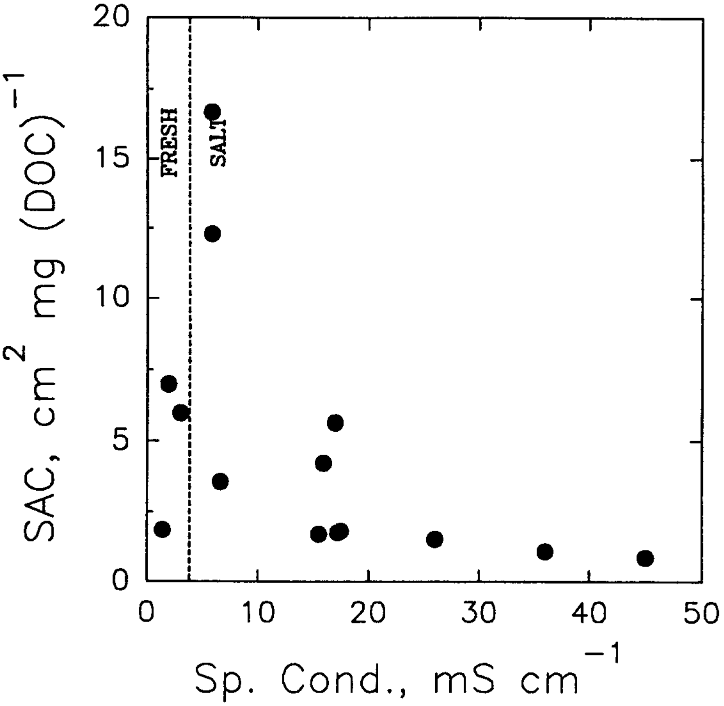

*Fig. 5.* Specific absorption coefficient (SAC, measured at 340 nm and normalized to path length and DOC concentration) of filtered water samples at measured specific conductance values. Fresh and saltwater are separated by the dashed line as in Fig. 2.

organic matter was independent of specific conductance and ranged from about 30 to 80 (Fig. 4b, $R^2 = 0.10$).

The range of relative color of organic matter, expressed as the specific absorption coefficient (SAC) decreased with increasing specific conductance from about 7 to 1 (Fig. 5). Two of the water samples had values in excess of 7, however these samples were visibly very turbid with colloidal material and their absorbance probably did not represent dissolved substances alone.

Concentrations and proportions of DOC smaller than 1 kiloDalton (kD) increased linearly with increasing specific conductance (Fig. 6a, $R^2 = 0.94$, $P < 0.01$; 6b, $R^2 = 0.83$, $P < 0.01$). Concentrations increased from 20 to 200 mg $L^{-1}$, and the proportions increased from about 35 to 70%. Concentrations of DOC larger than 1 kD were unrelated to salinity (Fig. 7a, $R^2 = 0.097$). The proportion of DOC >1 kD decreased with increasing salinity (Fig. 7b; $R^2 = 0.83$, $P < 0.01$) because the trend of increasing DOC <1 kD was so strong.

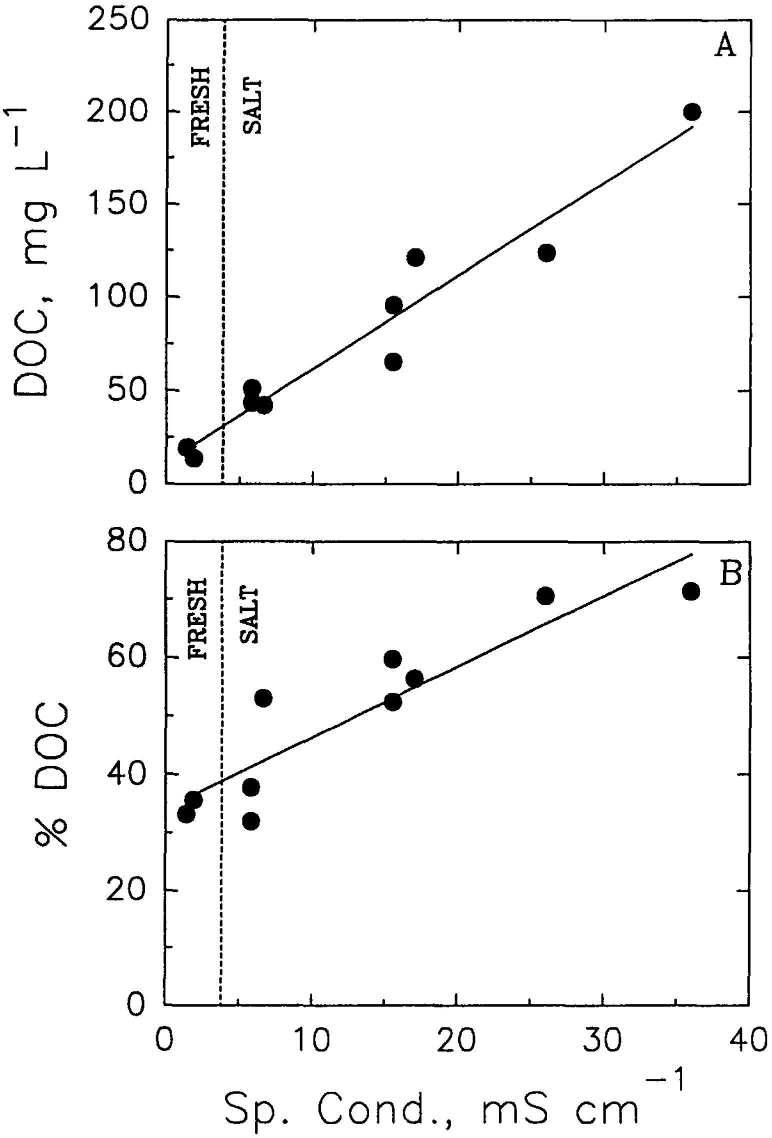

*Fig. 6.* a. Concentrations of DOC in ultrafiltered water (1000 Daltons) at measured specific conductance values ($R^2 = 0.94, P < 0.01$). b. The proportion of total DOC passing through the ultrafilter ($R^2 = 0.83, P < 0.01$). Solid lines are linear regressions and fresh and saltwater are separated by the dashed line as in Fig. 2.

## Discussion

### *DOM quantity*

The concentrations of dissolved organic carbon (DOC) measured in the Alberta freshwater lakes (22–70 mg L$^{-1}$, Fig. 2) were much higher than

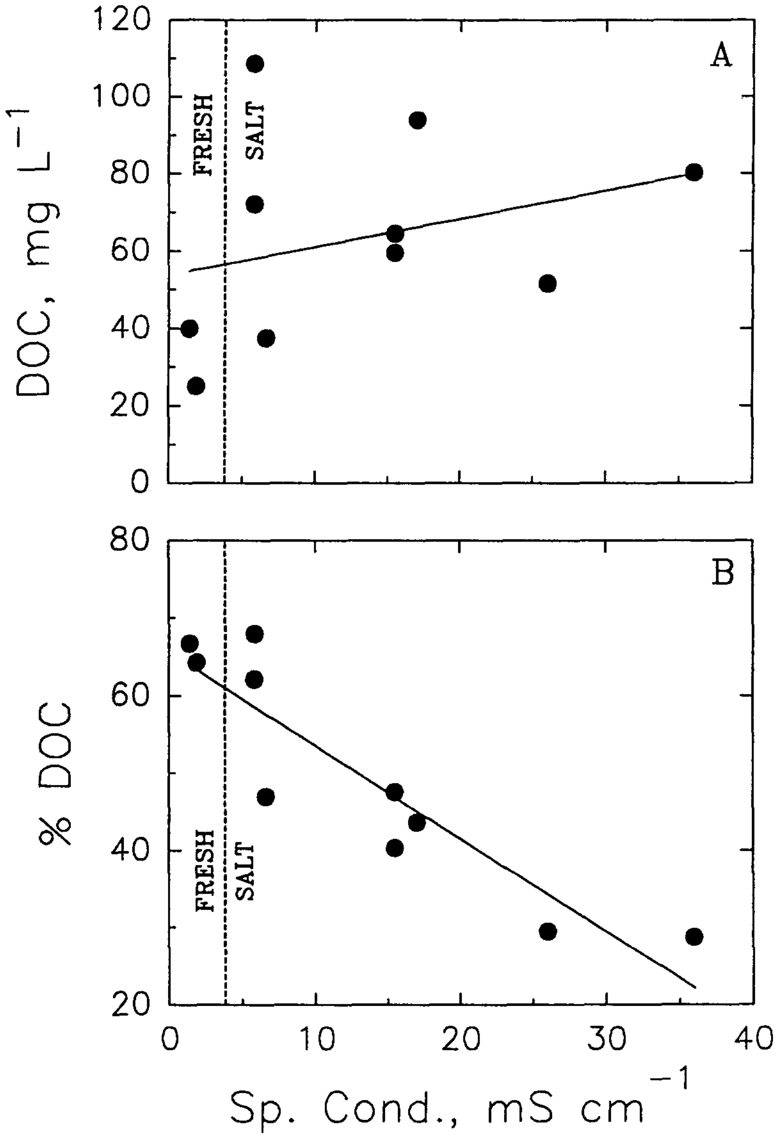

*Fig. 7.* a. Concentrations of DOC excluded by the ultrafilter normalized to total sample volume at measured specific conductance values ($R^2$ = 0.097). 7b. The proportion of total DOC excluded by the ultrafilter ($R^2$ = 0.83, $P < 0.01$). Solid lines are linear regressions and fresh and saltwater are separated by the dashed line as in Fig. 2.

for typical freshwater lakes in humid regions (Table 1). The pattern was the same as reported previously by Curtis & Prepas (1993) for 35 lakes in east-central Alberta. Concentrations of DOC in Alberta saline lakes (32–330 mg

*Table 1.* The range of selected characteristics of dissolved organic matter from different sites. Units are mg L$^{-1}$ for DOC and DON, atom ratio for C:N, percent of total DOC for humic substances (HS), units cm$^2$ (mg DOC)$^{-1}$ for specific absorption coefficient (SAC), and percent of total DOC less than molecular weight cuttoff indicated with < (molecular weight, MW, in kiloDaltons, kD).

| Site | DOC | DON | C:N | HS | SAC | MW, kD | Reference |
|---|---|---|---|---|---|---|---|
| Alberta, FW Lakes | 22–70 | 1–3.5 | 30–36 | 64 | 3–7 | <1kD, 33–35% | a |
| Alberta, SW Lakes | 32–330 | 2–14 | 26–75 | 64–73 | 1–6 | <1kD, 37–71% | a |
| Humid FW Lakes | 0.7–30 | 0.1–0.6 | 27–50 | 14–75 | 2–30 | <1kD, 7–40% | b |
| Other Salt Lakes | 15–90 | | | | | | c |
| Streams and Rivers | 1.5–95 | 0.16–0.4 | 9–48 | 50–85 | 22–41 | <1kD, 13% | d |
| Groundwater | 0.2–48 | 0.1–0.2 | 14–55 | 15–90 | | <10kD, 90% | e |
| Nearshore & Ocean | 0.6–3.6 | 0.03–0.25 | 7–15 | 5–15 | 9–18 | <1.8kD, 16–78% | f |
| Fulvic Acid | | | 20–200 | | 29–43 | 0.4–2 kD | g |

a) This study; Curtis & Prepas 1993. b) Tipping et al. 1988; Urban et al. 1990; Baron et al. 1991; Meili 1992; Curtis, unpublished data. c) Domagalski et al. 1989. d) Wetzel & Manny 1972; Meybeck 1982; Meyer & Tate 1983; Meyer 1986; Tipping et al. 1988; Lewis & Saunders 1989; Meili 1992; Trumbore et al. 1992; Vallentine & Zepp 1993; McDowell et al. 1994. e) Leenheer et al. 1974; Ford & Naiman 1989; Wassenaar et al. 1990; Schiff et al. 1990. f) Sugimura & Suzuki 1988; Yan et al. 1991; Hollibaugh et al. 1991; Hedges et al. 1992; Benner et al. 1992; Martin & Fitzwater 1992; Vallentine & Zepp 1993. g) Thurman et al. 1982; Thurman 1985; Aiken & Malcolm 1987; McKnight et al. 1988; McKnight et al. 1991; Vallentine & Zepp 1993.

L$^{-1}$, Fig. 2) were 6–10 times higher than for waters in humid regions, and about 100 times higher than for seawater (Table 1). Similarly, concentrations of dissolved organic nitrogen (DON) in Alberta freshwater and saline lakes (Fig. 4a) were about 10 and 100 times higher, respectively, than reported for freshwaters (Table 1). Concentrations of DON in seawater are generally lower than those reported for freshwater (Table 1).

Concentrations of DOC in east-central Alberta groundwater (13–30 mg L$^{-1}$, Fig. 2, excluding seepage between lakes) were similar to values reported for southern Alberta (Wassenaar et al. 1990). Concentrations of DOC measured in Alberta groundwater are much higher than values measured at most other sites throughout north America (generally less than 5 mg L$^{-1}$, Table 1). Concentrations were more typical of values measured in headwater lakes and low order streams in humid regions. Causes of elevated DOC concentration in Alberta groundwater are unknown. However, highest concentrations of DOC in Precambrian Shield streams are measured during droughts when fluxes are lowest (Schindler et al. 1992). Thus, low water yield might cause elevated DOC in Alberta groundwater.

Direct exchange of water between lakes and the atmosphere has opposite consequences on the concentration of catchment derived materials in humid and semi-arid regions. In humid regions, terrestrial inputs will be diluted with solute-poor precipitation because precipitation exceeds evaporation. Thus, concentrations of allochthonous materials in lakes cannot exceed the concentrations in catchment inputs. Conversely, in semi-arid regions, terrestrial inputs are concentrated in surface waters because evaporation exceeds precipitation and concentrations of solutes are limited by *in situ* loss processes (eg. solubility).

In humid regions, DOC concentrations decrease with increasing water residence time among (Meili 1992; Rasmussen et al. 1989) and within lakes (Schindler et al. 1992). In contrast, the concentrations of DOC in freshwater and saline lakes of sub-humid and semi-arid Alberta, increase with increasing water residence time (inferred from salinity). Thus, concentrations of DOC increase even though influxes of DOC decrease. These findings indicate that some part of DOC inputs can be evapoconcentrated or, alternatively, that internal sources of DOC tend to exceed internal sinks with increasing salinity. Seasonal changes in DOC concentration for eight Alberta saline lakes were directly proportional to evapoconcentration of total salts (Curtis & Prepas 1993), therefore a significant portion of DOC is probably highly refractory, given that the lakes have long water residence times.

The residence time for DOC to varies considerably among systems. For example, the half-time for loss of DOC (leaf leachate) in Bear Brook NH was only a few hours (McDowell 1985). Half-times of loss of streamwater DOC in limnetic enclosures was 90 days but was 350 days for lakewater DOC (calculated from Curtis 1993). Half-times for loss of riverine DOC in the ocean is thought to be on the order of 5–15 years (Kieber et al. 1990). The turnover time for DOC from the deep ocean is 6,000 years based on radiocarbon analysis of DOC (Williams & Druffel 1987). Thus, removal rates for DOC are site- and probably source–specific.

Even though most of the DOC pool in saline lakes is apparently refractory, a small dynamic portion could contribute significantly to ecosystem energy flow. Bacterial biomass can be very high in saline lakes (Kilham 1981) and appears to contribute significantly to the diet of grazers, often present in abundances far greater than predicted from the biomass of autotrophs (Campbell & Prepas 1986). Thus, the productivity of zooplankton might depend indirectly on microbially available DOC.

### *DOM quality*

The proportion of humic substances (humic DOC) in the DOC pools of Alberta surface waters (64–73%, Fig. 3b) is in the upper range of that encountered in surface and groundwaters (Table 1). High proportions of humic substances are typically associated with allochthonous sources because, the proportion of humic substances in autochthonous DOC is generally low (McKnight et al. 1994). High concentrations of humic-DOM are consistent with the sodium-dominance and high pH (Aiken & Malcolm 1987) of Alberta surface waters.

The relative importance of low molecular weight (LMW) DOC in lakes of semi-arid Alberta (32–71%, Fig. 6b) is much greater than for lakes from humid regions but similar to seawater. The high proportion of LMW DOC is consistent with the high proportion of humic DOC as fulvic acids (Aiken & Malcolm 1987), except that the proportion of LMW DOC was dependent on salinity whereas the proportion of humic DOC was not (Fig. 3b). Complicating these comparisons is the possible reduction of apparent molecular size with increasing ionic strength (DeHann et al. 1987). Interactions among the inorganic matrix and DOC separation techniques will require a great deal of attention to improve the quality of the comparisons made among systems using such operational characterizations.

C:N ratios in DOM from freshwater (29–35) and saline lakes (26–75, Fig. 4b) in Alberta are consistently higher than for seawater but were similar to values reported for freshwater from humid regions (Table 1). High C:N ratios, similar to high proportions of humic DOC, are also thought to indicate allochthonous sources (McKnight et al. 1994). Salinity-independent C:N ratios, in combination with large increases in DOC (Fig. 2) and DON (Fig. 4a) are consistent with evapoconcentration of refractory dissolved organic matter possibly of allochthonous origin. Low N-content of allochthonous DOM might limit its biodegradation because the nitrogen content of bacteria, plankton and particulate matter in general is much lower (Meybeck 1982; Heldal et al. 1985; Redfield et al. 1963).

The specific absorbance of DOM among Alberta lakes (excluding filtrates containing visible turbidity) was much lower than values determined for fulvic and humic acids for drainage lakes and streams and for coastal waters (Table 1). In general, specific absorbance by DOC in Alberta lakes was one quarter to one tenth of that calculated for other regions. Low specific color indicates low proportions of aromatic carbon in the DOC pool.

### *Possible mechanisms to explain shifts in DOM quality among lakes*

DOM in semi-arid Alberta is generally smaller (LMW), more humic, less colored, but with about the same N-content as DOM in lakes from humid regions (Table 1). Two hypothetical mechanisms are consistent with the differences in DOM quality between semi-arid Alberta lakes and waters from humid regions. These mechanisms are not mutually exclusive and may operate in combination. Both are amenable to experimental testing.

First, the relative abundance of materials in a complex mixture of DOM inputs could be altered by selective removal or transformation. Because, the color and molecular size of DOM in Alberta surface waters decrease with increasing water residence time, simple evapoconcentration of inputs is precluded. DOC in Alberta lakes could be the most refractory portion of the DOC mixture – resistant to scavenging, photolysis and biodegradation. Alternatively, a refractory pool could be created or enriched with materials transformed by incomplete reactions. For example, so-called photobleaching produces smaller, less colored DOC (Keiber et al. 1990). Similarly, incomplete biodegradation could reduce the size of organic molecules.

Second, DOC in Alberta Lakes could be from autochthonous sources. This hypothesis is consistent with the low color of autochthonous DOC (Tipping et al. 1988). However it is inconsistent with observations that refractory portion of autochthonous DOC is characterized as large molecular weight (Sundh 1992; Cole et al. 1984). Furthermore, autochthonous DOC is typically N-rich and humic-poor (McKnight et al. 1994) compared to the pool of DOM in east-central Alberta surface waters, making an internal source less likely in Alberta lakes.

Within humid and semi-arid regions, quantities of DOM are related empirically to proxies of water residence time – salinity in semi-arid regions and relative drainage area in humid regions. These relationships suggest that source and sink mechanisms are similar among lakes within regions. Between regions, opposite patterns of DOC concentration with water residence time were observed. The most likely explanation is that the most refractory DOC is diluted in humid regions and evapoconcentrated in arid regions. It is possible that mechanisms creating, consuming and transforming DOM quality are the same between regions. If so then the quality of DOM should depend similarly on water residence time between regions even though the pattern of DOC concentration is opposite. Whether sources and sinks are different *among* regions is the subject of our ongoing research.

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