Regulation of the nitrogen biogeochemistry of mountain lakes by subsidies of terrestrial dissolved organic matter and the implications for climate studies

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Abstract

Stable isotopes of nitrogen (N) were analyzed in modern sediments of mountain lakes, dissolved organic matter (DOM), and sediment cores spanning the past 12,000 yr to test the hypothesis that spatial and temporal (100–1000 yr) variation in the N content of mountain lakes is regulated by influx of allochthonous DOM. Analysis of spatial patterns in an elevation gradient of 75 mountain lakes revealed that most N was associated with DOM rather than inorganic N, particularly in subalpine lakes (< 1700 m above sea level). Similarly, analysis of N isotope ratios ($\delta^{15}N$) from 22 lakes showed that whole sediments of subalpine sites were significantly more depleted (0.74‰ ± 1.58‰) than were those of alpine lakes above 2200 m (3.04‰ ± 1.21‰), consistent with the depleted $\delta^{15}N$ of isolated DOM (~ 1.3‰). Sedimentary $\delta^{15}N$ values of Crowfoot Lake, presently near tree line, also varied greatly during the past 12,000 yr, with enriched values (~ 4‰) during the alpine phases of the lake's history and depleted values (~ 1‰) during the intervening subalpine phase (ca. 10,050–4160 ^{14}C yr before present) when DOM was abundant. In contrast, sedimentary $\delta^{15}N$ values remained constant (~ 2.5‰) at Snowflake Lake, an alpine reference site that never experienced a DOM-rich subalpine phase. These analyses suggest that climate regulates N influx and lake biogeochemistry by changing the subsidies of terrestrial DOM, and warn that future climate change may initially reduce N influx on a decadal scale by reducing hydrologic transfer before increasing N subsidies on a centennial scale by increasing terrestrial production of DOM.

Climate influences lake ecosystems by altering the influx of energy (E) and mass (*m*), both directly to the lake surface (Pham et al. 2008) and indirectly via the surrounding catchment (Blenckner 2005). Influx of E occurs mainly via the lake surface in the form of solar irradiance, atmospheric heat, and wind friction, whereas *m* enters the lake as water, dissolved substances, and transported particulate matter (Leavitt et al. 2009). To date, most limnological evidence demonstrates that effects of *m* transfer override the short-term effects of E on lake chemistry and biota (Pham et al. 2008; Dröscher et al. 2009), whereas long-term monitoring and paleoecological studies suggest that E transfer can also affect lakes indirectly by altering subsidies of organic and inorganic solutes to lakes (Leavitt et al. 2009). Further, hydrologic variability can affect lake ecosystems by altering the influx of allochthonous dissolved organic matter (DOM), a complex suite of molecules that regulate the physical (transparency, ultraviolet radiation [UVR], stratification), chemical (metal solubility, nutrients, O_2), and biological (microbial production) properties of lakes (Schindler et al. 1996; Vinebrooke and Leavitt 1998).

Most studies of boreal lakes demonstrate that the influx of allochthonous DOM to lakes is regulated by precipitation, soil flushing, and wetland release (Dillon and Molot 1997). Consequently, wetter conditions increase DOM inputs to these lentic systems (Forsberg 1992), whereas warmer and drier climates reduce DOM influx and increase in situ rates of removal (Schindler et al. 1990, 1992). Further, paleoecological studies suggest that extended periods of warm climate can promote development of terrestrial vegetation and soils in alpine environments (Luckman 1990; Hauer et al. 1997), which could increase export of terrestrially derived DOM and nutrients to lakes (Baron et al. 1991; McKnight et al. 1997). Mountain lakes near tree line may be particularly sensitive to climatically induced changes in DOM because they receive and often contain 10-fold less DOM than subalpine sites (Leavitt et al. 1997, 2003), and because changes in mean temperature (1–2°C) similar to those forecast for the next few decades (Field et al. 2007) can regulate development of high-elevation sources of terrestrial DOM (soils, coniferous forests) (Luckman 1990; Luckman et al. 1993).

Variation in DOM export from terrestrial systems as a result of soil development may have a pronounced effect on the biogeochemistry of N in mountain lakes, particularly for sites located at tree-line elevations. For example, studies of recently deglaciated terrains reveal that terrestrial ecosystems with poorly developed soils export little DOM or N to lakes until populations of N_2-fixing plants (e.g., *Alnus* spp.) and conifers have become established within the catchments (Hobbie et al. 1998; Engstrom et al. 2000; Engstrom and Fritz 2006). Before development of terrestrial DOM sources, dissolved N is present mainly as inorganic nitrate (NO_3^-), whereas organic N is more common in lakes with well-developed coniferous forests (Hood et al. 2005). Therefore, because DOM sources vary > 10-fold along an elevation gradient of mountain lakes (Leavitt et al. 1997, 2003), and because climate variability during the past 12,000 yr has caused an ~ 150-m shift in tree-line elevation (Luckman 1990; Luckman et al. 1993), we hypothesize that differences in N content of mountain lakes may arise primarily from spatial and temporal variation in subsidies of organic N from terrestrial sources.

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In this paper, we measured water chemistry, algal abundance, stable isotopes of DOM and sediments, and sediment geochemistry in two surveys of mountain lakes to quantify how DOM influx, N source, and N content of lake water and sediments varied as a function of lake position relative to tree line. Spatial patterns were then used to interpret historical changes in elemental composition and isotopic values of sediments both from Crowfoot Lake, a site presently near tree line but that has experienced a prolonged subalpine phase (10,050–4160 ^{14}C yr before present [BP]), and at high-elevation Snowflake Lake, a site that has remained in an alpine state for at least 6700 yr. Comparison of survey and core data revealed that spatial and temporal variation in N source and content among mountain lakes arose mainly because of differences in the influx of terrestrial DOM.

Methods

Study region—Study lakes are located mainly in Banff, Jasper, and Yoho National Parks (51°00′–53°30′N, 115°30′–119°30′W) of the Rocky Mountains, Alberta, Canada. Alpine water bodies generally occur above ~ 2100 m above sea level (a.s.l.), have short ice-free seasons (July–September), rapid water renewal times (< 30 d), and treeless catchments consisting of exposed rock, herbaceous vegetation, and patches of dwarf conifers. Tree line is not clearly defined in this region and ranges from 2100 to 2300 m a.s.l. depending on local topography and forest-fire history. Subalpine lakes are situated below tree line (1400–2100 m a.s.l.), have comparatively long ice-free seasons (early June to mid-Oct), and forested catchments containing mainly alpine fir (*Abies lasiocarpa*), Engelmann spruce (*Picea engelmannii*), and white spruce (*Picea glauca*). The geology of the region consists of stratified sedimentary rocks such as limestone, shale, and sandstone (Vinebrooke and Leavitt 1999).

Lake surveys—Mountain lakes were surveyed in August 1995 to quantify how water chemistry varied with lake elevation (1400–2400 m a.s.l.) and basin position relative to local tree line. In general, mean (±SD) conditions in these lakes include high pH (8.0 ± 0.3), low conductivity (193 ± 98 μS cm^{-1}), and low total phosphorus (TP) content (8 ± 19 μg P L^{-1}), but differ substantially in size (maximum depth = 21.5 ± 21.8 m). Although very large lakes are only common in valleys, mean lake size and chemistry are uncorrelated with basin elevation, with the exception of subalpine sites, which tend to have higher conductivity and dissolved organic carbon (DOC) content (*see* below).

Sampling and analytical techniques are described in Leavitt et al. (1997) and Vinebrooke and Leavitt (1999). Briefly, depth-integrated samples were collected from the uppermost 5 m within the epilimnia of 75 lakes, filtered through precombusted Whatman GF/F filters, and stored at ~ 0°C until analysis within 2 weeks for dissolved nutrient content (NO$_3^-$, NO$_2^-$, NH$_3$, DOC, PO$_4^{3-}$) using methods developed for dilute lakes, although not all analyses were available for all lakes. Unfiltered samples were analyzed for total Kjeldahl N (TKN) and TP following Stainton et al. (1977). Diffuse attenuation coefficients for UVR at 310 nm (Kd$_{310}$) were calculated

for each lake using filtered lake water, a Hewlett-Packard model 84452A photodiode array spectrophotometer, and a 10-cm quartz cuvette (Leavitt et al. 2003). At each site, the uppermost 2–3 cm of sediment was isolated with a hand-held mini-corer from an Ekman grab sampler collected at the deepest point in the lake. Sediments were frozen in the dark until analysis in the laboratory for fossil pigment and organic matter content (*see* below). Because these lakes are highly oligotrophic (Vinebrooke and Leavitt 1999), yet DOM is ~10-fold more abundant in subalpine lakes than in alpine sites (Leavitt et al. 1997), we hypothesized that total dissolved N content of mountain lakes would vary as a function of lake elevation and DOM content, with most dissolved N being derived from terrestrial organic matter in low-elevation lakes (i.e., DOM-N).

Sediments were collected from an additional 22 mountain lakes in July 2007 to quantify how sedimentary stable isotope ratios of N (δ^{15}N) and C (δ^{13}C) varied as a function of lake elevation and DOC content. As above, the uppermost 2–3 cm of sediment was isolated and frozen in the dark for duplicate laboratory analysis of elemental composition and stable isotope content, while water-column DOC and nutrient concentrations were determined for centrally located epilimnetic samples. Lakes were selected to lie along an elevation gradient of ~ 1000 m (1400–2400 m a.s.l.), although because of logistic constraints only six lakes were common to both 1995 and 2007 surveys. However, because both surveys encompassed the same national parks region, we assumed that there was no systematic difference in lake characteristics among lake sets. Instead, we hypothesized that both sedimentary δ^{15}N and δ^{13}C values should increase with lake elevation, reflecting the declining importance of isotopically depleted terrestrial DOM (Hood et al. 2005), the highly oligotrophic nature of alpine lakes (Leavitt et al. 1997), and the importance of mineral weathering in unvegetated landscapes in supplying ^{13}C-enriched carbonates to mountain lakes (Reasoner et al. 1994).

DOM isolation—DOM was isolated from two alpine lakes to determine its δ^{15}N and δ^{13}C values for comparison with sediments from survey lakes. Depth-integrated water-column samples were collected in 25-liter carboys from Middle Rowe (49°3′10″N, 114°3′21″W) (2163 m a.s.l.) and Pipit lakes (51°34′24″N, 115°49′45″W) (2217 m a.s.l.) during July and August 2007. These sites are located in Waterton Lakes and Banff National Parks, respectively. Samples were stored overnight at 10°C then sequentially screened (64-μm and 10-μm mesh), filtered (0.45-μm pore), and concentrated ~ 500-fold using a stainless-steel reverse osmosis (RO) unit manufactured by the Limnological Research Corporation (Winter et al. 2007). This system is equipped with a FilmTec FT30 US Filters thin composite RO membrane and has a molecular weight limit of 400 Da. Concentrated solutions were held at 5°C overnight to allow calcium carbonate to precipitate and separate from DOM by sedimentation. Isolated DOM was stored in acid-washed glass containers in darkness at 5°C until analysis for stable isotopes of N. In addition, DOM concentrates were washed with 0.3 mol L^{-1} HCl to remove carbonates and analyzed for δ^{15}N and δ^{13}C values. On the basis of

previous analysis of the isotopic composition of dissolved fulvic (δ^{15}N = 1.0‰ ± 0.3‰; δ^{13}C = −26.6‰ ± 0.6‰) and transphilic acids (δ^{15}N = 2.4‰ ± 0.2‰; δ^{13}C = −23.3‰ ± 0.9‰) isolated from other alpine catchments of the eastern Rocky Mountains (Hood et al. 2005), we anticipated that whole DOM would have a δ^{15}N signature of 1–2‰, whereas δ^{13}C would range from −24‰ to −27‰.

Holocene cores—Sediment cores spanning ca. 7000 to 12,000 yr were collected from two oligotrophic lakes near tree line to test the hypothesis that long-term (100–1000 yr) changes in climate regulate lentic biogeochemical cycles of N by altering influx of terrestrial DOM-N to mountain lakes. Crowfoot Lake (51°39′06″N, 116°25′27″W) is located at 1940 m a.s.l., just below the upper elevation of subalpine forest vegetation in the upper Bow Valley of Banff National Park (Holland and Coen 1982). At present, this lake receives drainage from predominantly alpine habitats, including ~ 30% of the catchment that is ice covered (Reasoner and Huber 1999; Leavitt et al. 2003). In contrast, Snowflake Lake is located ~ 40 km to the east (51°35′53″N, 115°49′57″W) in an undisturbed, unglaciated basin 200 m above permanent tree line (~ 2320 m a.s.l.) in the Canadian Rocky Mountain front ranges, 3 km south of Pipit Lake (Leavitt et al. 1994). Because of its elevation and steep catchment, Snowflake Lake has likely remained in an alpine state for most of the past 12,000 yr. In contrast, palynological, geochemical, and leaf macrofossil evidence shows that the Crowfoot Lake catchment was in a predominantly alpine state from ~ 12,880 to ~ 10,100 ^{14}C yr BP, and again from ~ 4160 ^{14}C yr BP to present, but that subalpine *Pinus* forests predominated during the warmer and drier intervening period (Reasoner et al. 1994; Reasoner and Huber 1999). Because this warm interval was also characterized by a 10-fold increase in the influx of terrestrially derived DOM to Crowfoot Lake (Leavitt et al. 2003), we hypothesized that historical change in sedimentary N and C isotope signatures during the Holocene mainly should reflect variations in terrestrial subsidies of organic matter and carbonates, but that these changes should not be evident in alpine Snowflake Lake.

Sediment cores from Crowfoot and Snowflake lakes were collected using the lightweight percussion corer as described by Reasoner et al. (1994). The two Crowfoot Lake cores included the 2.45-m-long CRW3 and the 2.05-m-long CRW6, whereas a 1.67-m-long core (SL1) was recovered from Snowflake Lake. Each core was cut into 1-m sections in the field, transported to University of Alberta, sectioned into 1-cm intervals, and frozen for subsequent analyses. Core CRW3 was analyzed for fossil pigments from algae and bacteria (Leavitt et al. 2003), organic matter content, fossil pollen, and leaf macrofossils (Reasoner et al. 1994; Reasoner and Huber 1999), whereas cores CRW6 and SL1 were analyzed for the elemental composition (%C, %N, organic matter) and stable isotopes signatures (δ^{13}C, δ^{15}N) of both whole sediments and those treated with 0.3 mol L^{-1} HCl to remove carbonates (Leng et al. 2005).

Chronological control for CRW3 was based on 18 accelerator mass spectrometric (AMS) analyses of ^{14}C activities of terrestrial macrofossils, the presence of charac-

teristic Holocene volcanic ash layers (tephra) from the Bridge River (~ 2360 ^{14}C yr BP), St. Helens Y$_n$ (~ 3400 ^{14}C yr BP), and Mazama (~ 6730 ^{14}C yr BP) eruptions, and AMS-dated lithostratigraphic transitions at the base of the core (Reasoner et al. 1994; Reasoner and Huber 1999). Sediment ages were approximated for CRW6 using a combination of lithrostratigraphic transitions and tephra layers, as well as statistical correlations between organic-matter content profiles from each core determined as percentage mass loss on ignition (LOI) at 500°C for 1 h (Leavitt et al. 2003). Sediment chronology of SL1 was approximated using only the three main regional tephra layers (*see* above), as the corer was unable to penetrate the > 10-cm-thick Mazama ash layer at this site, and leaf macrofossils were absent from the core. Although pollen was isolated from the base of SL1, the presence of detrital organic carbon (charcoal) from regional fires prevented reliable AMS determination of sediment age (basal date > 36,000 ^{14}C yr).

Laboratory analyses—Stable isotope ratios and elemental composition were determined using a ThermoQuest (F-MAT) Delta^{PLUS}XL isotope ratio mass spectrometer equipped with continuous flow (Con Flo II) unit and an automated Carlo Erba elemental analyzer as an inlet device (Savage et al. 2004). Stable N (δ^{15}N) and C (δ^{13}C) isotopic compositions were expressed in the conventional δ-notation in units of per mil (‰) deviation from atmospheric N$_2$ and organic C standards previously calibrated against authentic Vienna Pee Bee Belemnite. Sample reproducibility was < 0.25‰ and < 0.10‰ for δ^{15}N and δ^{13}C determinations, respectively.

Algal abundance and community composition was quantified from analysis of fossil pigments and their derivatives in sediments from the 1995 lake survey and the Crowfoot Lake CRW3 core. Pigments were extracted from freeze-dried sediments, filtered (0.2-μm pore), and dried under pure N$_2$ gas using the standard methods of Leavitt and Hodgson (2001). Carotenoids, chlorophylls (Chls), and derivatives were isolated, quantified, and calibrated using the high-performance liquid chromatography system of Leavitt and Hodgson (2001). Pigment analysis was restricted to taxonomically diagnostic carotenoids, including β-carotene (all algae), alloxanthin (cryptophytes), diatoxanthin (diatoms), myxoxanthophyll (colonial cyanobacteria), lutein–zeaxanthin (chlorophytes and cyanobacteria), and unique UVR-absorbing pigments from benthic cyanobacteria (Leavitt et al. 1997). Historical changes in UVR penetration (as UV-b radiation, 280–320 nm) were estimated for Crowfoot Lake as the ratio of UVR-absorbing pigments : algal carotenoids, an index that is linearly related to the depth of UVR-b penetration in whole-lake experiments (Leavitt et al. 1997). All pigment concentrations were expressed as nmol of pigment g^{-1} dry sediment, a metric that is linearly related to algal biomass in whole-lake calibrations (Leavitt and Findlay 1994).

Development of terrestrial sources of allochthonous N within the Crowfoot Lake region was quantified using the standard methods of Reasoner and Huber (1999) by estimating the accumulation rate of pollen (grain cm^{-2} yr^{-1}) derived from N$_2$-fixing terrestrial taxa (*Alnus* sp.,

Shepherdia canadensis), the main source of N to lakes in recently deglaciated and tree-line environments (Engstrom et al. 2000). Briefly, known concentrations of exotic *Lycopodium* pollen were added to whole sediment samples before the initial digestion in concentrated HCl to permit calculation of pollen concentrations and accumulation rates. Pollen identification was carried out at 400–1000X magnification with reference to appropriate taxonomic keys and the North American pollen reference collection at the Institute of Arctic and Alpine Research, Boulder, Colorado (Reasoner and Huber 1999).

Numeric analyses—Pearson correlation coefficients and unreplicated linear regressions were performed with SYSTAT v. 10.0 (SPSS Software) to quantify the statistical relationships between lake-water and sedimentary parameters measured in lake surveys. Statistical relationships between fossil time series from Crowfoot Lake were also quantified using Pearson correlation and regression analyses following sequential $\log_{10}$ and first-difference transformations to normalize the variance and remove temporal autocorrelations, if required. Cross-correlations were also calculated to determine whether there were significant lagged relationships between pairs of variables; however, as no significant lagged relationships were recorded, only lag = 0 relationships are reported herein. In instances where correlations were calculated between time series from different cores (e.g., CRW3, CRW6), time series were harmonized first to produce a common chronology and sampling interval using the procedures of Patoine and Leavitt (2006).

Past DOM concentrations (as mg DOC L^{-1}) and depths of UVR penetration were reconstructed for Crowfoot Lake as described by Leavitt et al. (2003). Briefly, because sediment organic matter content (as LOI) and water-column DOM concentrations are correlated strongly in these mountain lakes ($r = 0.77$, $p < 0.0001$, $n = 75$) (Leavitt et al. 1997), we used historical changes in core LOI to reconstruct past water-column concentrations of DOM. Past depths of UVR-b penetration were then calculated from reconstructed DOM concentrations using regional optical models (Leavitt et al. 2003). Depth of UVR-b penetration was compared with the mean lake depth derived from modern area-weighted basin morphometry to identify time periods in which the entire lake basin was subject to damaging levels of UVR (Leavitt et al. 2003).

Results

Lake surveys—Regression analysis of data from the 1995 survey showed that TKN content was correlated strongly and positively with that of DOC ($r^2 = 0.883$, $p < 0.0001$), whereas levels of dissolved inorganic N (DIN $= NO_3^- + NO_2^- + NH_3$) declined slightly with increased DOC concentration ($r^2 = 0.082$, $p = 0.01$) (Fig. 1a). As a result, organic N (TKN-NH_3) was the predominant component of TKN at most sites, with DIN achieving $> 50\%$ of TKN only in lakes with < 2.0 mg DOC L^{-1}. We infer that most of the organic N was present in the dissolved phase because algal biomass (as planktonic Chl *a*) was correlated only weakly with DOC concentration ($r^2 = 0.061$, $p = 0.032$)

and lake elevation ($r^2 = 0.036$, $p = 0.08$), and rarely exceeds 10 μg L^{-1} in these and other regional lakes (Vinebrooke and Leavitt 1999).

Water-column DOC concentrations were correlated negatively with lake elevation in both the 1995 ($r^2 = 0.225$, $p < 0.0001$) (Fig. 1b) and 2007 surveys ($r^2 = 0.640$, $p < 0.0001$) (not shown), although lakes with low DOC content were present at all elevations, and alpine lakes (> 2200 m a.s.l.) always had < 2.5 mg DOC L^{-1} (mean $\pm$ SD $= 1.06 \pm 0.70$ mg DOC L^{-1}). In addition, DOC concentrations were correlated positively ($r^2 = 0.579$, $p < 0.0001$) with the organic matter content of sediments (Fig. 1c) and integrated algal biomass (benthic + planktonic) ($r^2 = 0.197$, $p < 0.0001$) as measured by sedimentary concentrations of the ubiquitous algal pigment β-carotene (not shown). As reported by Leavitt et al. (2003), this metric of total algal abundance was also correlated strongly and positively with the ability of DOC to absorb UVR ($r^2 = 0.607$, $p < 0.0001$), as measured by Kd_{310} (Fig. 1d).

Sedimentary $\delta^{15}N$ values of both whole ($r^2 = 0.319$, $p = 0.004$) (Fig. 1e) and acidified sediments ($r^2 = 0.403$, $p = 0.003$) (not shown) were correlated positively with lake elevation in the 2007 survey. Although there was substantial variability at any given altitude, the mean ($\pm$SD) sediment $\delta^{15}N$ for alpine lakes above 2200 m a.s.l. ($\delta^{15}N_{whole}$ 3.04‰ $\pm$ 1.21‰; $\delta^{15}N_{acid}$ 3.85‰ $\pm$ 1.38‰) was ~ 2.5‰ greater than that for subalpine sites below 1700 m a.s.l. ($\delta^{15}N_{whole}$ 0.74‰ $\pm$ 1.58‰; $\delta^{15}N_{acid}$ 1.12‰ $\pm$ 1.17‰). Further, N isotope signatures of both whole ($r^2 = 0.449$, $p < 0.0001$) and acidified sediments ($r^2 = 0.395$, $p = 0.003$) were correlated negatively with the N content of the sediment (% dry weight [wt]). Similar positive correlations with elevation ($r^2_{whole} = 0.301$, $p = 0.004$; $r^2_{acid} = 0.252$, $p = 0.014$) and negative correlations with sedimentary N ($r^2_{whole} = 0.587$, $p < 0.0001$; $r^2_{acid} = 0.220$, $p = 0.021$) and C contents ($r^2_{whole} = 0.485$, $p < 0.0001$; $r^2_{acid} = 0.248$, $p = 0.015$) were also observed for C isotope ratios of whole and acidified sediments (Fig. 1f). However, although acidification had little consistent effect on absolute $\delta^{15}N$ values (< 0.5‰), treatment of sediments with HCl to remove carbonates reduced $\delta^{13}C$ values by an average of 8‰ for lakes below 1700 m a.s.l. and ~ 10‰ for those above 2200 m a.s.l. Residual $\delta^{13}C$ values of acidified sediments ranged from -21.7‰ to -32.6‰, whereas acid-treated C : N ratios averaged 11.7 ± 2.4, by mass. Finally, sedimentary $\delta^{15}N$ values were correlated negatively with water-column DOC concentration in the 2007 survey ($r^2_{whole} = 0.512$, $r^2_{acid} = 0.393$, both $p < 0.005$) (Fig. 1g), whereas DOC and sedimentary N content (% dry wt) were correlated positively ($r^2_{whole} = 0.659$, $r^2_{acid} = 0.612$, both $p < 0.0001$) (Fig. 1h).

Isolated DOM—Isotopic ratios of DOM purified and concentrated by reverse osmosis were similar for both Pipit and Middle Rowe lakes (Fig. 2). DOM exhibited a mean ($\pm$SD) $\delta^{15}N$ value of 1.38‰ $\pm$ 0.14‰ for whole and acidified samples from both lakes, whereas $\delta^{13}C$ values ranged from -23.4‰ to -25.2‰ for acidified samples from Middle Rowe and Pipit, respectively. The $\delta^{13}C$ value of whole-DOM concentrates (ca. -10‰) was not consid-

ered a reliable metric of pure DOM because these samples also include ^{13}C-enriched carbonates concentrated along with DOM during RO procedures (*see* Methods). Instead, it was noted that together, the $\delta^{15}N$ and $\delta^{13}C_{acid}$ values of pure DOM were similar to those obtained for acidified lake sediments (Fig. 1e,f).

Crowfoot Lake core—AMS dating described by Reasoner et al. (1994) and Leavitt et al. (2003) demonstrates that the CRW6 and CRW3 cores span over 11,000 ^{14}C yr. Within the CRW3 core, $\delta^{15}N$ values of whole sediments varied $\sim 4.5\permil$ during the Holocene ($\sim 11,330$ to 0 ^{14}C yr BP) (Fig. 3a), with values of $\sim 4.0\permil$ during cooler alpine phases ($\sim 11,330$ to 10,050 ^{14}C yr BP and ~ 4160 to 0 ^{14}C yr BP) known from previous analysis of fossil pollen and coniferous leaf remains (Reasoner et al. 1994; Reasoner and Huber 1999). In contrast, $\delta^{15}N$ ratios were generally $\sim 1.0\permil$ during the intervening subalpine phase with the exception of three enriched ($\sim 3–5\permil$) samples deposited between ~ 8000 and ~ 6500 ^{14}C yr BP. The $\delta^{15}N$ values of acidified samples (not shown) were correlated strongly with those of whole sediments ($r^2 = 0.773$, $p < 0.0001$), particularly for sediments older than 2000 yr ($r^2 = 0.944$, $p < 0.001$).

The $\delta^{13}C$ values of Crowfoot Lake whole sediments varied by over $25\permil$ during the Holocene, from enriched values of $\sim 0\permil$ to $-10\permil$ before 10,050 ^{14}C yr BP and after ~ 4160 ^{14}C yr BP, to depleted $\delta^{13}C$ of ca. $-25\permil$ to $-28\permil$ during the intervening ~ 6000-yr period (Fig. 3b). Isotope ratios were particularly enriched ($\sim 0\permil$ to $-5\permil$) both in the first and last 1000 yr of lake history, periods of time when glacial activity is thought to be greatest (Luckman et al. 1993; Reasoner and Huber 1999). In contrast, C isotope values of acidified sediments were more highly constrained (range $-25\permil$ to $-28\permil$) than those of whole sediments. Finally, historical changes in C and N isotopes were uncorrelated ($r^2 < 0.05$, $p > 0.05$) for time series of both whole and acidified sediments.

The $\delta^{15}N$ of whole sediments (Fig. 3a) was correlated inversely with both sediment N content (Fig. 3c) ($r^2 = 0.298$, $p < 0.0001$) and development of terrestrial N sources as recorded by the influx of pollen from N_2-fixing terrestrial plants (*Alnus* sp. and *S. canadensis*) (Fig. 3d) ($r^2 = 0.312$, $p < 0.0001$). Similarly, $\delta^{15}N$ was correlated negatively with total algal abundance (as fossil β-carotene) (Fig. 3e) during the Holocene ($r^2 = 0.600$, $p < 0.0001$), suggesting that either N subsidies regulated algal production, or that algae regulated N isotope composition. In contrast, there was a weak positive correlation between $\delta^{15}N_{whole}$ and the relative importance of colonial cyanobacteria recorded as either the proportion of sedimentary myxoxanthophyll to eukaryotic algal pigments (percentage cyanobacteria) ($r^2 = 0.063$, $p = 0.062$), or the relative concentration of photoprotective pigments from benthic cyanobacteria (UVR index) ($r^2 = 0.140$, $p = 0.004$) (both Fig. 3f). Together, these patterns suggest that in situ N_2 fixation by cyanobacteria did not regulate historical changes of sedimentary $\delta^{15}N_{whole}$.

As demonstrated by Leavitt et al. (2003), historical changes in total algal abundance within Crowfoot Lake (Fig. 3e) were correlated positively with past lake-water DOC concentration (Fig. 3g) reconstructed from sediment LOI ($r^2 = 0.637$, $p < 0.0001$) and inversely ($r^2 = 0.401$, $p < 0.0001$) with the maximum depth of UVR penetration (Fig. 3h). Specifically, algal abundance declined more than 10-fold during periods in which estimated water-column DOC declined to below ~ 1.75 mg C L^{-1} and UV-b radiation penetrated to the bottom of the lake.

Snowflake Lake core—Approximate dating of sediments using distinctive tephra layers demonstrated that core SL1 encompassed the period from the Mazama eruption (~ 6730 ^{14}C yr BP) to the present. In contrast to Crowfoot Lake, the isotopic composition of sediments from alpine Snowflake Lake did not vary substantially during the past ~ 6730 yr (Fig. 4). For example, $\delta^{15}N$ values of whole sediment ranged between $\sim 2\permil$ and $3\permil$ throughout the 1.7-m core (mean $\pm$ SD = $2.5\permil \pm 0.3\permil$), with only slightly depleted values recorded for sediments deposited between ~ 6730 and ~ 4000 ^{14}C yr BP (Fig. 4b). Similarly, there was little change in the $\delta^{13}C$ values of either whole or acidified Snowflake Lake sediments (Fig. 4d), with core-wide means ($\pm$SD) of $-23.9\permil \pm 1.4\permil$ and $-25.8\permil \pm 0.4\permil$, respectively. These isotope ratios are similar both to each other and to those values recorded at Crowfoot Lake ($-24.9\permil \pm 1.0\permil$) during the period ca. 7000–4000 ^{14}C yr BP (Fig. 4c), suggesting relatively little influence of ^{13}C-enriched carbonates at Snowflake Lake. Consistent with this view, there was little temporal variability in sedimentary content of N (Fig. 4f) and C (not shown) at Snowflake Lake. Together, these patterns are consistent with the absence of glaciers in the modern basin and further suggest that this site has been in an alpine state for at least 6700 yr.

Discussion

Analysis of modern lake-water and sediment chemistry, stable isotopes of DOM and sediments, and Holocene lake cores suggests that spatial and temporal variation in N content, N biogeochemistry, and N isotopic values of mountain lakes are all strongly influenced by terrestrial subsidies of N in DOM to lakes. For example, water-column concentrations of DOM were correlated positively with the dissolved and total N content of lakes (Fig. 1a), organic (%LOI) and elemental (%N, %C) content of sediments (Fig. 1c,h), and algal abundance (Fig. 1d), but were correlated negatively with sedimentary $\delta^{15}N$ (Fig. 1g) and lake elevation (Fig. 1b). Consequently, subalpine lakes generally exhibited depleted sedimentary N isotope ratios ($\delta^{15}N_{whole} = 0.74\permil \pm 1.58\permil$) relative to those of alpine sites ($3.04\permil \pm 1.21\permil$) because of elevated DOC concentrations (Fig. 1b), very low ratios of DIN : TKN ($< 1 : 10$) (Fig. 1a), and the isotopically depleted nature of pure DOM ($\sim 1.3\permil$) (Fig. 2) (Hood et al. 2005). Similarly, comparison of sediments from tree-line Crowfoot Lake (Figs. 3b, 4a) and alpine Snowflake Lake (Fig. 4b) revealed that Holocene transitions from alpine to subalpine status were also marked by 10-fold increases in the abundance of terrestrial N sources (Fig. 3d), water-column DOC concentrations (Fig. 3g), and sedimentary N content (Fig. 4c), while sedimentary $\delta^{15}N$ declined $\sim 3\permil$ to values consistent with those of modern DOM (Figs. 2, 3a) and sediments of
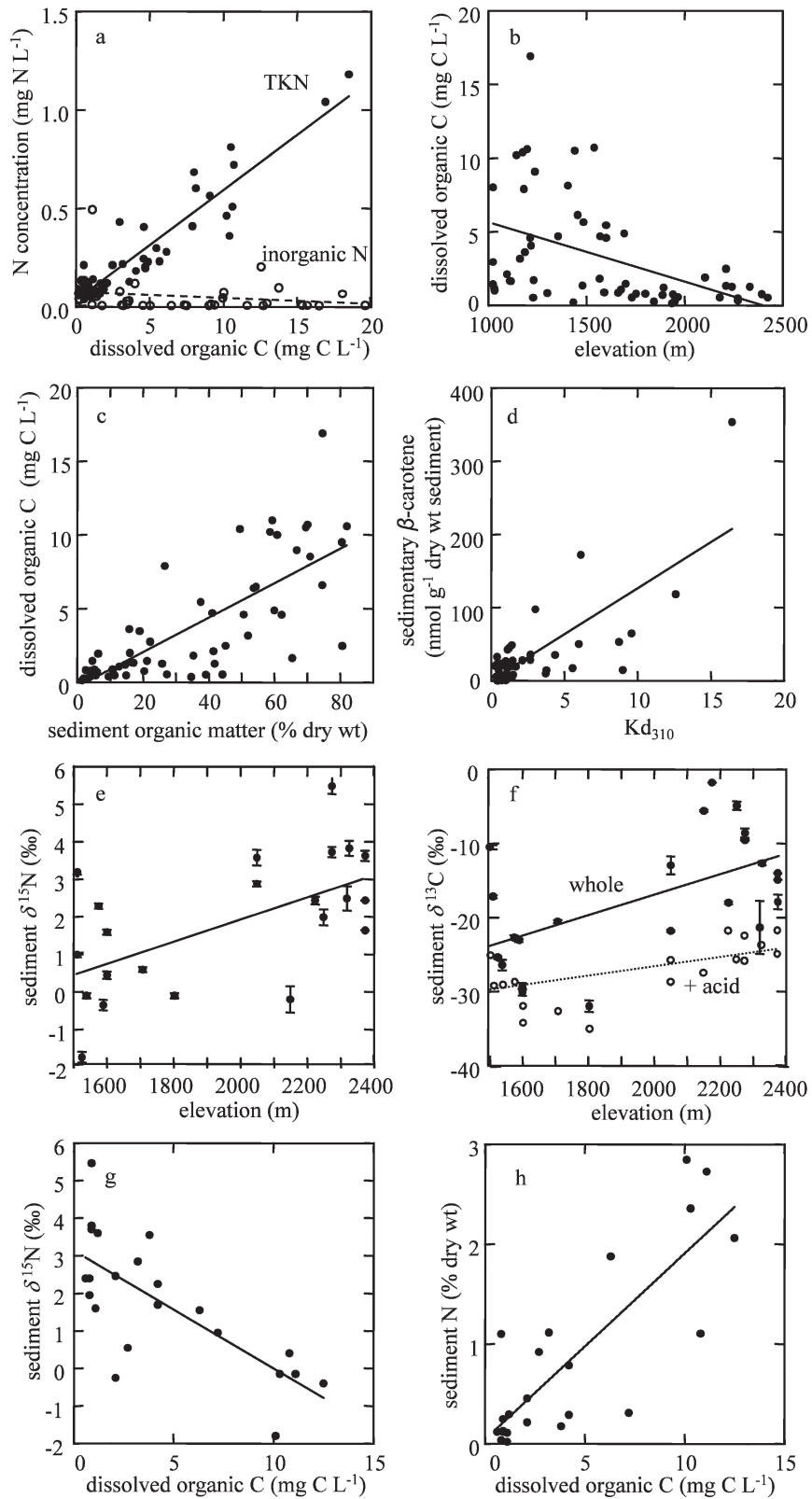
Fig. 1.   Gradients of water-column and sedimentary nitrogen (N) and carbon (C) revealed from surveys of mountain lakes near Banff National Park, Alberta, Canada during (a–d) 1995 and (e–h) 2007. (a) Relationship between total Kjeldahl N (TKN), total inorganic N (sum NO_3^-, NO_2^-, NH_3), and dissolved organic carbon (DOC) in epilimnetic waters. (b) Relationship between epilimnetic DOC and lake elevation. (c) Positive correlation between water-column DOC
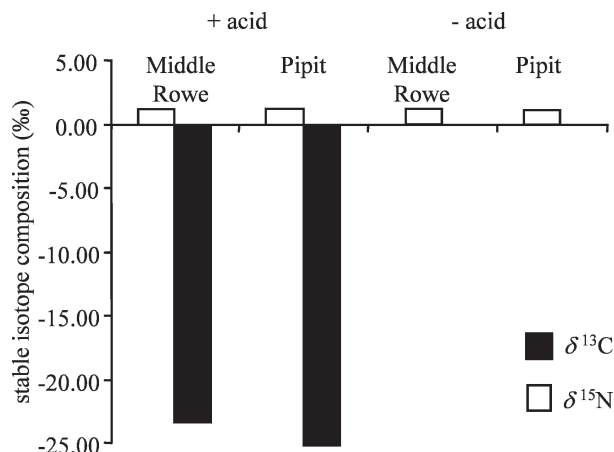
Fig. 2. Mean isotopic value of dissolved organic matter (DOM) concentrated by reverse-osmosis procedures from alpine Pipit and Middle lakes. Bars represent analysis of duplicate aliquots from each lake after pretreatment with acid to remove carbonates (+acid) or without pretreatment (−acid). White bars represent the $\delta^{15}N$ signature of whole or acidified DOM, whereas black bars represent $\delta^{13}C$ signature of acidified DOM. Unacidified C isotope values are not presented for whole DOM because ^{13}C-enriched carbonate was also concentrated during the DOM isolation procedure (*see* Methods).

subalpine lakes (Fig. 1e). Because both tree-line elevation and alpine soil development are known to be regulated by changes in mean temperature and precipitation (Luckman 1990; Reasoner and Huber 1999), these findings suggest that climate variability may regulate the chemistry and production of mountain lakes primarily by altering the influx of terrestrial DOM (Leavitt et al. 2003).

Source of N in mountain lakes—Several lines of evidence suggest that the influx of terrestrial DOM was the main factor regulating differences in N content among lakes along the elevation gradient. First, the total N content of both lake water ($r^2 = 0.883$, $p < 0.0001$) and sediments ($r^2 = 0.512$, $p < 0.0001$) were strongly correlated to water-column DOC concentrations, whereas water-column TKN content was uncorrelated with DIN concentrations ($r^2 = 0.030$, $p = 0.095$) and correlated only weakly with planktonic Chl a abundance ($r^2 = 0.087$, $p = 0.012$). Second, mean concentrations of DIN recorded during the 1995 survey (37 μg N L^{-1}) were sixfold less than mean TKN levels (230 μg N L^{-1}), demonstrating that organic N was the main fraction of N in the water column of many mountain lakes. Finally, although total dissolved N was not determined

directly, we infer that dissolved organic N (DON) was the predominant component of TKN because cell quotas of N : Chl are $\sim$ 6 : 1 by mass in live algae (Montagnes et al. 1994) and water-column Chl a rarely exceeds 10 μg L^{-1} in these lakes (Vinebrooke and Leavitt 1999), leading to a maximum particulate N content ($\sim$ 60 μg L^{-1}) that is at least fourfold less than mean water-column TKN concentrations (Fig. 1a). Consistent with the importance of terrestrial DOM influx in controlling the N content of lakes, the predominance of organic N was most marked in lakes below tree line, whereas alpine lakes above 2200 m a.s.l. consistently exhibited low DOM concentrations (1.06 $\pm$ 0.70 mg DOC L^{-1}) and DIN constituted > 50% of TKN.

The predominance of the DON fraction in the N budget of subalpine lakes is consistent with findings from other DOM-rich, low-elevation sites (Engstrom et al. 2000; Fritz et al. 2004; Leavitt et al. 2006), but contrasts lakes from the Colorado Rocky Mountains (Hood et al. 2003). In this latter case, TKN concentrations increase strongly ($r^2 = 0.82$, $p < 0.01$) as a function of the proportion of DIN : DON in lakes that are heavily polluted with NO$_3^-$ from anthropogenic sources (automobiles, agriculture). In addition, the magnitude of NO$_x$ deposition in precipitation, water-column concentrations of NO$_3^-$, and the proportion of NO$_3^-$ from human sources all increase with lake elevation in the U.S. Rocky Mountain sites (Nanus et al. 2008). Although some lakes of the Canadian National Parks should also be affected by atmospheric N pollution from cities (Calgary) and prairie farming, the observations that DIN varies little with elevation ($r^2 = 0.007$, $p = 0.238$) and that TKN is uncorrelated with DIN ($r^2 = 0.030$, $p = 0.095$) suggest that deposition of anthropogenic NO$_x$ is far less pronounced in many of these lakes than in comparable U.S. sites 400–1600 km to the southeast.

Strong correlations between DOM in the water column ([TKN-NH$_3$], DOC) and sedimentary content of N (Fig. 1h) or organic matter (Fig. 1c) may arise from several complementary mechanisms. First, both planktonic and sedimentary organic matter may be derived from inputs of terrestrial DOM from catchment soils and vegetation (McDowell and Likens 1988; Cole et al. 1989). In this instance, DOM can increase the organic matter content of sediments by suppressing microbial activity (Guildford et al. 1987), reducing the photic zone (Schindler et al. 1996), depleting deep-water oxygen content (Wetzel 1992), and being precipitated by UVR (Schindler et al. 1996). Second, organic N and C in the water column may be derived in part from release of degraded sedimentary organic matter

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concentration (mg C L^{-1}) and sedimentary organic matter content (% weight [wt] loss on ignition at 500°C). (d) Relationship between total algal abundance inferred from sedimentary concentrations of ubiquitous β-carotene and the diffuse attenuation coefficient (Kd) for UVR at 310 nm. (e) Variation in the $\delta^{15}N$ values of whole sediments (0–3-cm deep) and lake elevation. (f) Relationship between $\delta^{13}C$ signature of whole (solid symbol) and HCl-treated surface sediments (open symbol), and lake elevation. (g) Regression of the $\delta^{15}N$ value of whole surface sediments and the DOC content of epilimnetic waters. (h) Regression of the N content (% dry wt) of whole surface sediments and the DOC content of epilimnetic waters. Mean and standard deviation of duplicate isotope analyses presented in panels e and f.
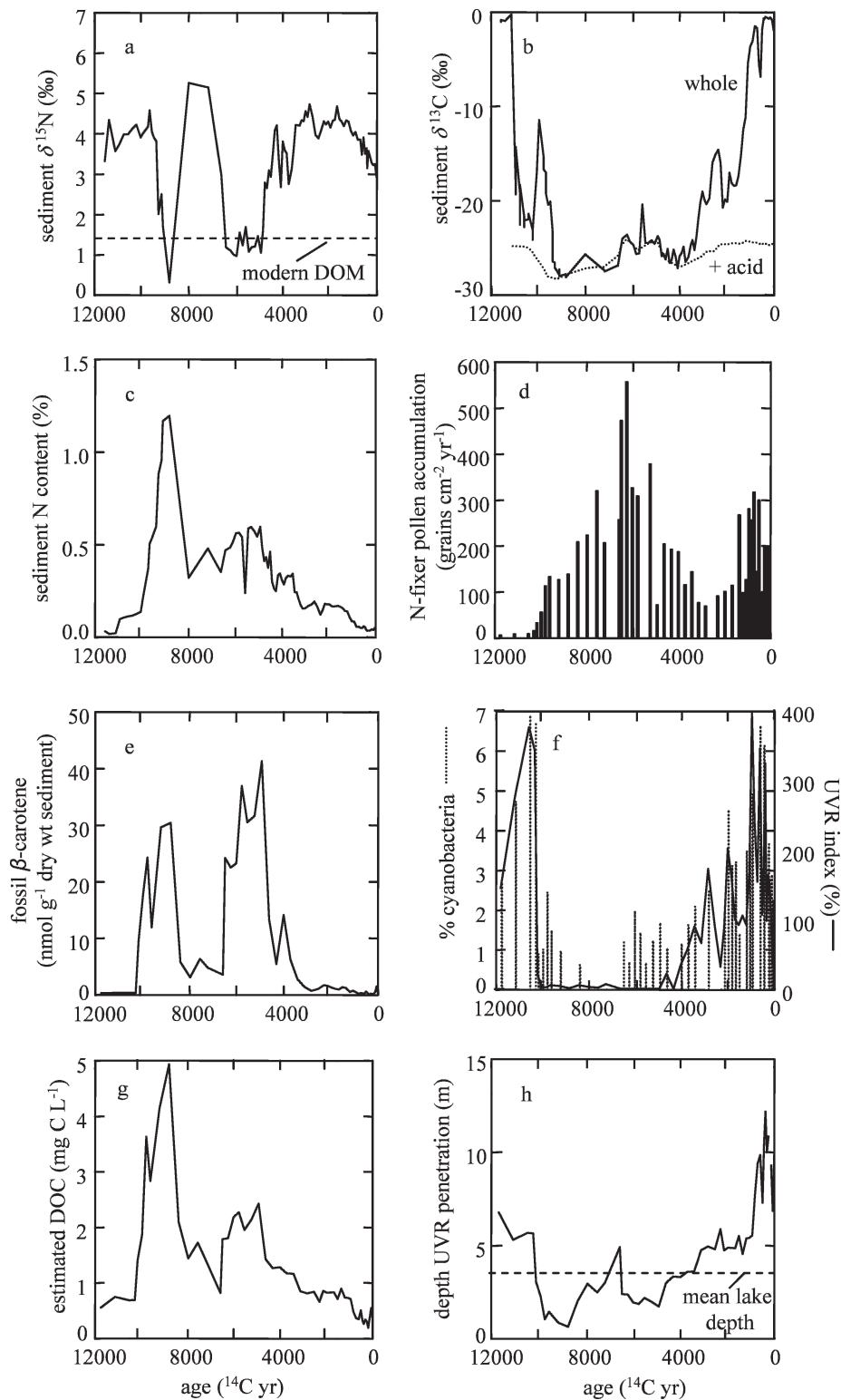
Fig. 3. Historical changes in sedimentary parameters and reconstructed water-column parameters at Crowfoot Lake, Alberta, Canada during the past ~ 12,000 yr. Time series include (a) $\delta^{15}N$ and (b) $\delta^{13}C$ values of whole (solid line) or HCl-acidified (dashed line) sediments (‰), (c) N content of sediments (% dry wt), (d) influx of pollen from regional N-fixing terrestrial plants (grains cm^{-2} yr^{-1}), (e) historical changes in total algal abundance recorded by fossil β-carotene (nmol pigment g^{-1} dry wt), (f) changes in the percentage relative abundance of fossil myxoxanthophyll (dashed histogram) and photoprotective pigments (solid line) from colonial

(Ishiwatari 1985; Aiken et al. 1991). Third, both water-column and sedimentary organic matter content could be affected by the influx of inorganic particulate matter from the landscape (Rowan et al. 1992), with small particles both scouring DOM from the water column (Leavitt et al. 2003; Luider et al. 2003) and diluting sedimentary organic matter content (Thornton et al. 1990). Finally, substantial differences in hydrology may affect deposition of organic matter such that rapidly flushed systems have both low DOM retention and minimal sedimentation of organic matter (den Heyer and Kalff 1998). At present, we do not favor any specific explanation and are conducting further research to identify the precise mechanisms linking water-column and sedimentary organic matter pools.

Analysis of stable isotope ratios of N in mountain lake sediments and DOM suggests that the δ^{15}N value of any given site reflects the local mixture of ^{15}N-enriched inorganic N ($\sim$ 3‰) and depleted organic N sources ($\sim$ 1‰), modified further by in-lake processes. For example, regression analysis of the relationship between sedimentary δ^{15}N values and DOC concentrations in the water column (Fig. 1g) suggests that lakes without significant DOM have mean N isotope ratios of 3.15‰ ± 0.39‰, irrespective of basin elevation. This value is similar to that recorded for both isolated NO_3^- (Nanus et al. 2008) and unpolluted preindustrial whole sediments (Wolfe et al. 2003) from alpine lakes of the U.S. Rocky Mountains. In contrast, sites in which DOM-N represents over 75% of N (i.e., > 2 mg DOC L^{-1}) (Fig. 1a) exhibited a mean $\delta^{15}N_{whole}$ of 0.97‰ ± 1.48‰, similar to values recorded for RO-isolated whole DOM in this study (1.38‰ ± 0.14‰) and terrestrial fulvic acids in alpine lakes of Colorado (1.0‰ ± 0.3‰) (Hood et al. 2005). Taken together, these analyses suggest that changes in sedimentary δ^{15}N values with elevation (Fig. 1e) arise in part because of the transition in N source from ^{15}N-depleted DOM-N in subalpine lakes to enriched NO_3^- at high elevation sites ($\sim$ 3‰) (Seastedt et al. 2004).

Substantial variability of sedimentary δ^{15}N values among lakes within a given elevation zone (Fig. 1e) or DOM content (Fig. 1g) likely arises from a combination of differences in N isotope ratios of source materials, in situ processing, and the influence of local catchment characteristics. For example, sites with low DOM content were present at all elevations (Fig. 1b), reflecting the effect of basin aspect and orientation on local tree-line elevation and soil development (Holland and Coen 1982) and the presence of active glaciers within individual catchments (Luckman et al. 1993; Reasoner and Huber 1999). Similarly, differences in proximity to point sources of atmospheric pollutants (e.g., Calgary) may introduce both

spatial (Nanus et al. 2008) and temporal (Wolfe et al. 2003) variation that affect sedimentary N isotope ratios. We also expect that δ^{15}N values will vary among sites because of differences in total N content of soils and vegetation (Brock et al. 2007; Bunting et al. 2007) and its effects on isotopic ratios (Hobbie et al. 1998), N_2 fixation by cyanobacteria (Patoine et al. 2006), nutrient limitation and N uptake by algae (Velinsky et al. 1991), and highly fractionating microbial processes (nitrification, denitrification), particularly in strongly stratified lakes (Kendall 1998; Teranes and Bernasconi 2000; Lehmann et al. 2004). Finally, variations in the proportion of microbial (2–3‰) and terrestrial (0–1‰) DOM may influence the isotope ratios of local organic matter (Hood et al. 2005). Unfortunately, more refined quantification of the relative importance of these factors will require catchment-scale mass balances of organic and inorganic N in contrasting catchment types, an approach that is beyond the scope of the present study.

Source of C in mountain lakes—Unexpectedly, analysis of C : N ratios did not clearly identify the provenance of organic matter inputs to lakes. For example, the C : N of RO-isolated DOM was low and ranged from 3 : 1 (Pipit Lake) to 13 : 1 (Middle Rowe Lake). Similarly, the very strong linear regression relationship (r^2 = 0.886, p < 0.0001) between putative DON content (as $TKN-NH_3$) and DOC concentration suggested an average C : N ratio of 14.5 for DOM. Finally, analysis of lake sediments demonstrated that C : N of acidified sedimentary material decreased only slightly with increased lake elevation (r^2 = 0.324, p = 0.005) such that even subalpine sites with DOC concentrations > 5 mg DOC L^{-1} and dense local forests exhibited low C : N ratios (14.1 ± 1.4). Because these low C : N ratios are similar to those of live algae, it is not possible to clearly identify the source of aquatic organic C without more sophisticated analysis of DOM fluorescence and ^{13}C-nuclear magnetic resonance spectroscopy (Hood et al. 2005).

Analysis of C stable isotope ratios suggested that the relative importance of organic and inorganic C in sediments varied widely among mountain lakes. For example, acidification of lake sediments reduced mean δ^{13}C signatures by $\sim$ 10‰ to −27.1‰ ± 4.0‰ (Fig. 1f) and C : N ratios from 48.9 ± 107.6 to 11.6 ± 2.4, both patterns consistent with the selective removal of highly enriched carbonates (ca. 0‰ to −10‰) by acidification to CO_2 (Leng et al. 2005). Although δ^{13}C values of acidified sediment increased modestly with lake elevation (r^2 = 0.301, p = 0.004), suggesting a change in C source, the isotope ratios of residual organic C could not be distinguished from those of terrestrial or aquatic plants (> −28‰), lacustrine algae (< −26‰) (Hodell and

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cyanobacteria, (g) water-column DOC concentration (mg C L^{-1}) estimated from sediment organic matter content, and (h) estimated depth of UVR penetration (m) in Crowfoot Lake relative to mean lake depth (dashed line). Previous analysis of fossil pollen, charcoal, and leaf fragments (Reasoner et al. 1994; Reasoner and Huber 1999) demonstrates that Crowfoot Lake was in an alpine state before ca. 10,050 ^{14}C yr before present (BP) and after 4160 ^{14}C yr BP, but was in a subalpine state during the intervening $\sim$ 6000 yr.
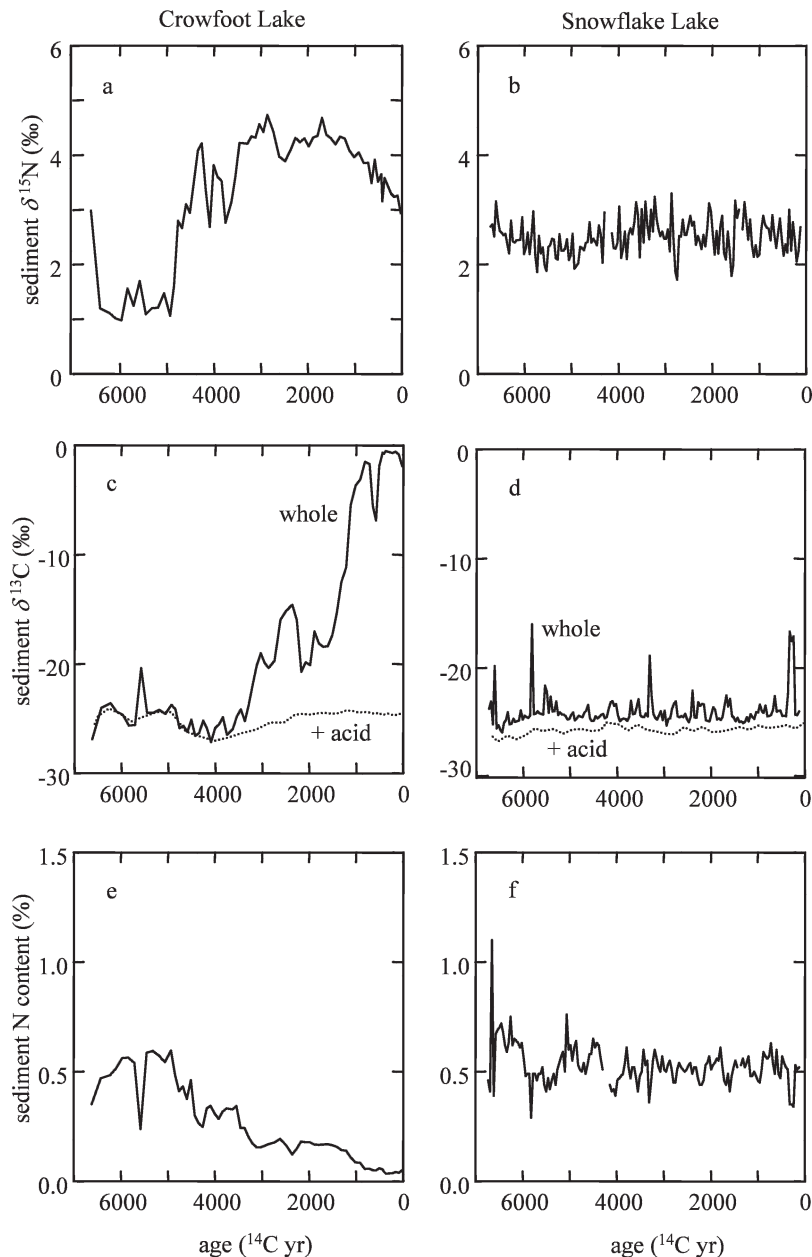
Fig. 4. Comparison of sedimentary profiles of (a, b) $\delta^{15}N$, (c, d) $\delta^{13}C$, and (e, f) N content (% dry wt) from Crowfoot (left column) and Snowflake lakes (right column). Isotope values for whole and acidified samples as in Fig. 3. Prior analysis by Reasoner and Huber (1999) showed that Crowfoot Lake (1940 m a.s.l.) transitioned from subalpine to alpine status ca. 4160 ^{14}C yr BP, whereas Snowflake Lake (2320 m a.s.l.) has been in a near-alpine state throughout the core period (ca. 6730 ^{14}C yr). Acidified samples are presented in dashed lines.

Schelske 1998; Meyers and Teranes 2001; Lehmann et al. 2004), or DOM isolated in this study ($-24.3 \pm 1.0‰$) (Fig. 2) or in the U.S. Rocky Mountains ($-24.5‰$ to $-27‰$) (Hood et al. 2005). Instead, these findings suggest that comparison of $\delta^{13}C$ values from whole and acidified sediments may be a useful metric of changes in the influx of inorganic C (e.g., glacial activity).

Temporal variation in N and C sources—Historical changes in sediment composition were consistent with climatic regulation of DOM influx to mountain lakes and its subsequent control of aquatic N biogeochemistry (Figs. 3, 4). For example, sediments deposited during the two alpine phases of Crowfoot Lake were characterized by the same enriched $\delta^{15}N$ and $\delta^{13}C$ values (Fig. 3a,b) and low concentrations of N (Fig. 3c) seen in modern alpine lakes (Fig. 1), whereas reconstructed concentrations of DOC (< 1 mg C L^{-1}) (Fig. 1g) were typical of high-elevation lakes in which most N is present as ^{15}N-enriched NO_3^-. In contrast, most sediments deposited during the subalpine

period ($\sim$ 10,100 to 4160 ^{14}C yr BP) at this site exhibited depleted δ^{15}N values (1–1.5‰) characteristic of both pure DOM (1.38 $\pm$ 0.14) (Fig. 2) and sediments from low-elevation lakes (0.74‰ $\pm$ 1.58‰) in which organic N accounted for the majority of water-column N (Fig. 1e). Because these stratigraphic changes were not recorded in the sediments of alpine Snowflake Lake during the past $\sim$ 6730 ^{14}C yr, despite the proximity (40 km) and similar orientation of both catchments (west side of broad north–south-oriented valleys), we infer that climatic variability affected Crowfoot Lake mainly via catchment development and transfer of solutes to lakes, rather than through direct effects on the lake itself (stratification, temperature, etc.) (Blenckner 2005; Leavitt et al. 2009). Similarly, because the timing of δ^{15}N enrichment preceded that of δ^{13}C in Crowfoot Lake by over 1000 yr (Fig. 4a, c), we infer that changes in δ^{15}N were not directly regulated by variation in glacier activity.

Climatically induced variations in the DOM influx also may have caused the period of enriched ($\sim$ 5‰) sedimentary δ^{15}N values (ca. 8000–6500 ^{14}C yr BP) recorded when Crowfoot Lake was in a subalpine phase (Fig. 3a). Specifically, we speculate that a period of intense aridity and reduced runoff during the subalpine phase may have temporarily reduced hydrologic export of ^{15}N-depleted DOM from the terrestrial environment, and favored recycling of inorganic N leading to enriched N isotope signatures. Consistent with this hypothesis, reconstructed DOC levels had declined fivefold by 8000 ^{14}C yr BP to concentrations typical of alpine lakes in which ^{15}N-enriched NO_3^- is the predominant N species within the water column (Fig. 3g), whereas sedimentary charcoal deposits reached their highest concentrations ca. 6800 ^{14}C yr BP, consistent with drier conditions and an elevated incidence of local fires (Reasoner and Huber 1999). As a result of a decrease in water-column DOM concentrations, algal abundance also declined ca. 10-fold (Fig. 3e) due to increased penetration of damaging UV-b radiation throughout most of the lake basin (Fig. 1h) (Leavitt et al. 2003). In addition, hydrologic influx is inferred to have been at an historical minimum during this transitory period because δ^{13}C values of whole and acidified sediments were both similar and highly depleted (Fig. 3b), a pattern that suggests minimal glacier activity within the catchment at this time. Overall, these changes are consistent with other limnological studies that demonstrate that DOM influx is regulated by hydrologic inputs (Dillon and Molot 1997), that in-lake concentrations of DOC decline during droughts (Schindler et al. 1996), and that the annual yield of terrestrial organic N is a linear function of runoff from mountain catchments (Sickman et al. 2001).

Implications for climatic effects on mountain lakes— Climate influences lake ecosystems by altering the influx of E and *m* both directly to the lake surface (Pham et al. 2009) and indirectly via the surrounding catchment (Blenckner 2005; Leavitt et al. 2009). To date, short-term (10–100 yr) studies suggest that transfer of solar and atmospheric E increases synchrony in chemical and physical properties among lakes (Dröscher et al. 2009; Pham et al. 2009), whereas *m* influx reduces the temporal coherence among

lakes because of higher spatial variation of precipitation and associated matter (Pham et al. 2008, 2009; Dröscher et al. 2009) and because differences in catchment characteristics (vegetation cover, topology, soils, etc.) act as a basin-specific filter of common forcing by climate (Blenckner 2005). The present study both confirms and extends these observations by demonstrating that E transfer can also indirectly regulate lake properties by altering the subsidies of DOM from land. This effect may be particularly pronounced for tree-line lakes with sparse vegetation in the upper reaches of their catchments, as these are sites where temperature rather than precipitation appears to regulate the rate of soil development (Luckman 1990). In addition, we infer that E and *m* transport mechanisms may interact to regulate the export of terrestrial DOM to lakes (e.g., Fig. 3a), as overland and shallow subsurface water flow is the main pathway by which terrestrial DOM reaches lakes (Schindler et al. 1992; Dillon and Molot 1997) and because droughts are known to immediately reduce DOM transport to lakes (Leavitt et al. 1997).

Modest extrapolation of our findings suggests that future global warming is likely to have nonlinear effects on the N biogeochemistry of lakes in the Canadian Rocky Mountains. At the scale of years to decades, increases in mean regional temperature favor increased melting of snowpack and glaciers, elevated hydrologic runoff (Schindler and Donahue 2006), and increased DOM export to mountain lakes as DOM is flushed from shallow soils (Hood et al. 2005). However, at the scale of decades to centuries, continuously higher temperatures lead to loss of permanent snowpack, widespread glacial wasting (Watson and Luckman 2004), higher evapotranspiration rates, and declines in lotic flows (Schindler and Donahue 2006), which should reduce export of terrestrial N into mountain lakes (Sickman et al. 2001). Finally, at the scale of centuries to millennia, continuously warmer conditions may increase the export of DOM to lakes after development of high-elevation sources of terrestrial organic matter (soils, coniferous forests), although this transfer also requires sufficient hydrologic runoff to deliver DOM to lakes.

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