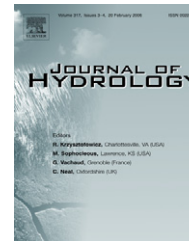




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# A coupled isotope tracer method to characterize input water to lakes

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**Summary** We develop a new coupled isotope tracer method for characterizing the isotopic composition of input water to lakes, and apply it in the context of ongoing hydrological process studies in the Peace-Athabasca Delta, a large, remote, riparian ecosystem in the boreal region of western Canada. The region has a highly seasonal climate, with floodplain lakes typically receiving input only during the 4–6 month open-water season from varying proportions of spring snowmelt, summer rains and river flooding. These possible input sources have distinct ranges of isotopic compositions that are strongly constrained to a well-defined local meteoric water line, thus affording the opportunity to derive lake-specific estimates of the integrated isotopic composition of input waters after accounting for the effects of secondary evaporative isotopic enrichment. As shown by comparison of the results of isotopic surveys of delta lakes prior to freeze-up in 2000 and 2005, this isotopic characterization of input waters can be combined with other data and field observations to provide new insight into spatial and temporal variability in delta lake recharge processes. This includes evidence that summer rainfall in 2000 played an important role in replenishing shallow basins delta-wide, especially in the central low-lying region, compensating for below-average snow accumulation during the previous winter. In contrast, 2005 was marked by greater relative contributions from both snowmelt and river flooding because of high winter snow accumulation and a spring ice-jam that caused river floodwaters to enter some basins in the southern part of the delta. The method is readily transferable to investigations in other remote regions that are sparsely monitored by conventional hydrometric networks.

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## Introduction

Understanding the relative roles of hydrological processes on lake water balances in large and remote freshwater ecosystems is important for their management but is challenging because approaches that depend upon conventional instrumentation may be impractical. Installation of hydro-metric devices in ecosystems with numerous aquatic basins requires substantial investment and assumptions are frequently required to, for instance, close lake water balances. For basins located in deltas and river floodplains that receive multiple sources of water, lake level recorders may provide useful knowledge of water level increases but other independent hydroclimatic information is needed to identify the cause of the observed change. Furthermore, results derived from only a few instrumented sites may be difficult or inappropriate to extrapolate over complex landscapes where the relative importance of hydrological processes is expected to vary spatially.

In recent years, water isotope tracers have become increasingly utilized as an alternative approach for providing assessments of lake water balances in remote regions. For example, Gibson and Edwards (2002) conducted a systematic survey of lake water isotope compositions to understand regional variations in evaporation losses and water budgets associated with the climate gradient across the northern boreal treeline in Canada. More recently, Mayr et al. (2007) used water isotope tracers in the southern Patagonia of Argentina to characterize contemporary lake water balances with the purpose of informing paleolimnological investigations. In these and other studies, quantitative estimates of lake water balances are derived using variations of the Craig and Gordon (1965) linear resistance model that describes isotopic evaporative enrichment. Applications commonly utilize lake water oxygen and hydrogen isotope compositions separately, which frequently generates small differences in water balance estimates using the individual tracers. Although these differences are often attributed to analytical or model uncertainties, mass conservation dictates that lake water balances calculated from lake water oxygen and hydrogen isotope compositions must agree.

Here we develop a new approach to the application of water isotope tracers that preserves the fundamental assumption of mass conservation allowing additional hydrological information to be derived regarding the nature of source waters to lakes in the Peace-Athabasca Delta (PAD), Canada. This large freshwater ecosystem contains hundreds of shallow (most are <2 m) basins where rainfall, snowmelt and river water are important to sustain aquatic habitat but their relative roles over space and time are not well characterized. These investigations build upon quantitative assessment of lake water balances across the PAD, which identified distinct landscape sectors of hydro-limnological conditions based on integration of isotope and chemistry analyses of lake water samples collected in October 2000 (Wolfe et al., 2007). In this previous study, as elsewhere, comparison of oxygen- and hydrogen-isotope estimated evaporation-to-inflow ratios revealed a small but systematic departure from the 1:1 line that was thought to emanate from uncertainties in model input values. We re-visit this dataset and utilize another from September

2005 to evaluate input water isotope compositions during two years characterized by very different hydrological and meteorological conditions. Results show marked spatial and temporal variability in input waters to lakes using our new coupled isotope tracer method, one that is readily transferable to other large, freshwater ecosystems.

## Study area

The Peace-Athabasca Delta (PAD) is a large (~3900 km<sup>2</sup>) wetland complex located at the convergence of the Peace, Athabasca and Birch rivers at the western end of Lake Athabasca, northern Alberta, Canada (Fig. 1). The PAD can be subdivided into three deltaic sectors: the Athabasca sector to the south (~1970 km<sup>2</sup>), the Peace sector to the north (~1680 km<sup>2</sup>) and the much smaller Birch sector to the west (~170 km<sup>2</sup>) (PADPG, 1973). Several large shallow lakes (Claire, Baril and Mamawi lakes) are located in the center of the PAD, where the three sectors coalesce (Fig. 1). The Peace sector lies to the north of these lakes and is a relict fluviodeltaic landscape that is covered by mature forests with bedrock inliers in the northeast. This sector is flooded only during major ice-jams that periodically develop on the Peace River. The southern Athabasca sector is an active delta of extremely low relief that frequently receives river floodwaters during both the spring thaw and open-water seasons. Ice-jam flooding of portions of the Athabasca sector occurred in the spring of 2005, preceding one of our lake water sample collections (September 2005).

There are numerous shallow lakes in the PAD, which span a broad hydrological spectrum. Based on previous studies incorporating water isotope tracers and limnological characteristics, basins have been categorized into four drainage types (Wolfe et al., 2007). Open-drainage basins are located mainly in the low-lying central portion of the delta where they frequently receive discharge from many of the rivers and creeks that constitute the complex channel network of the PAD. Closed-drainage basins are generally found in the Peace sector and receive widespread river water only during periodic ice-jam flood events on the Peace River, and thus, input from precipitation is an important source of water to these lakes. Restricted-drainage basins are located mainly in the Athabasca sector, where the input of river water is the primary hydrological process that controls lake water balances. Rainfall-influenced basins are found mainly in the central portion of the delta adjacent to the large open-drainage basins and occupy shallow depressions in the landscape (depth <50 cm). Their water balances are similar to those of restricted-drainage basins but their source waters are dominated by summer precipitation.

Climate in the PAD is strongly seasonal. According to 1971–2000 climate normals at Fort Chipewyan, Alberta, (Weather Station ID 3072658; Environment Canada, 2004), mean annual air temperature is  $-1.9^{\circ}\text{C}$ , mean January air temperature is  $-23.3^{\circ}\text{C}$  and mean July air temperature is  $16.7^{\circ}\text{C}$ . Precipitation averages 391.7 mm annually, with about 59% falling as rain during the May–September period. Meteorological conditions preceding the two sample collections (October 2000 and September 2005) were different (Fig. 2). The winter of 1999/2000 was warm and dry

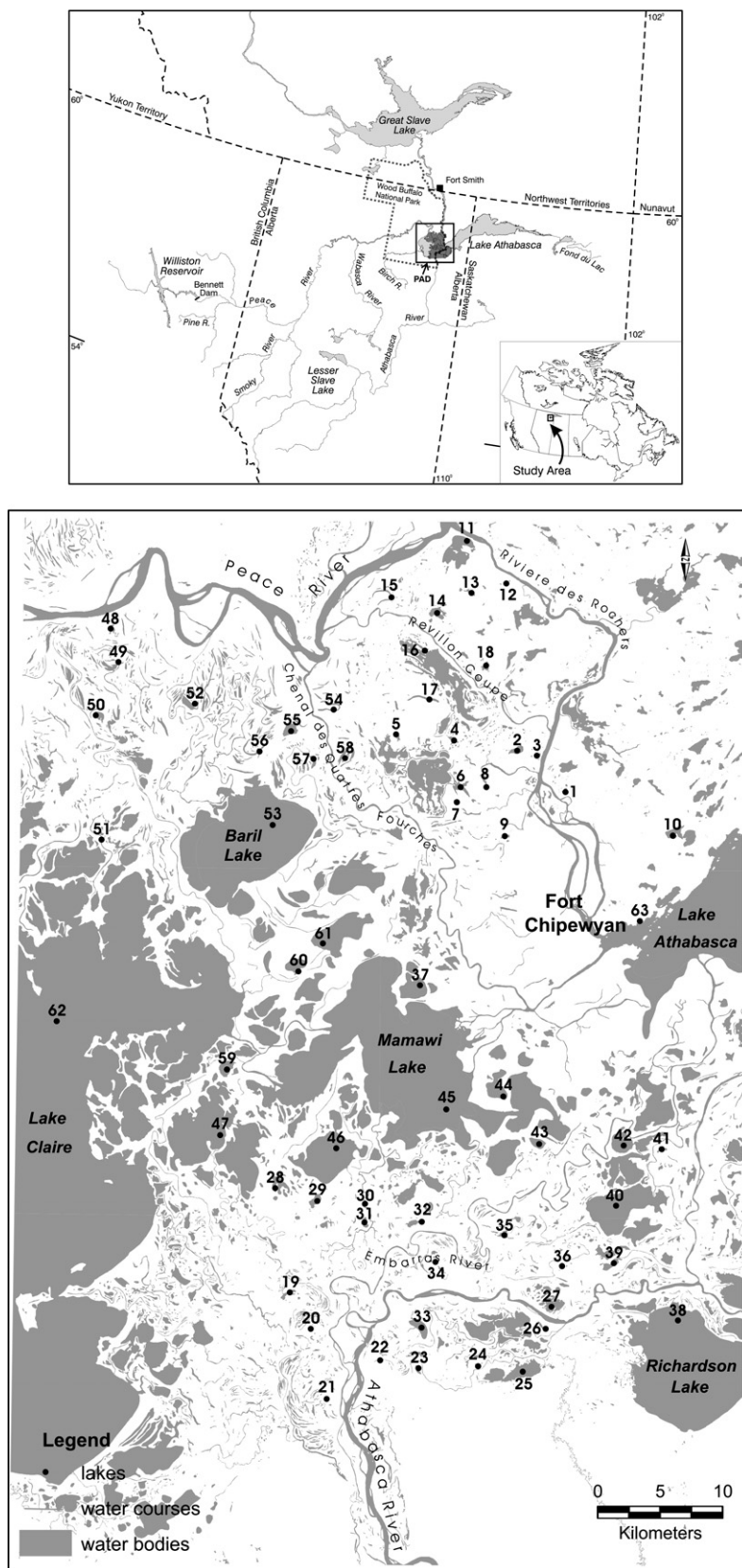
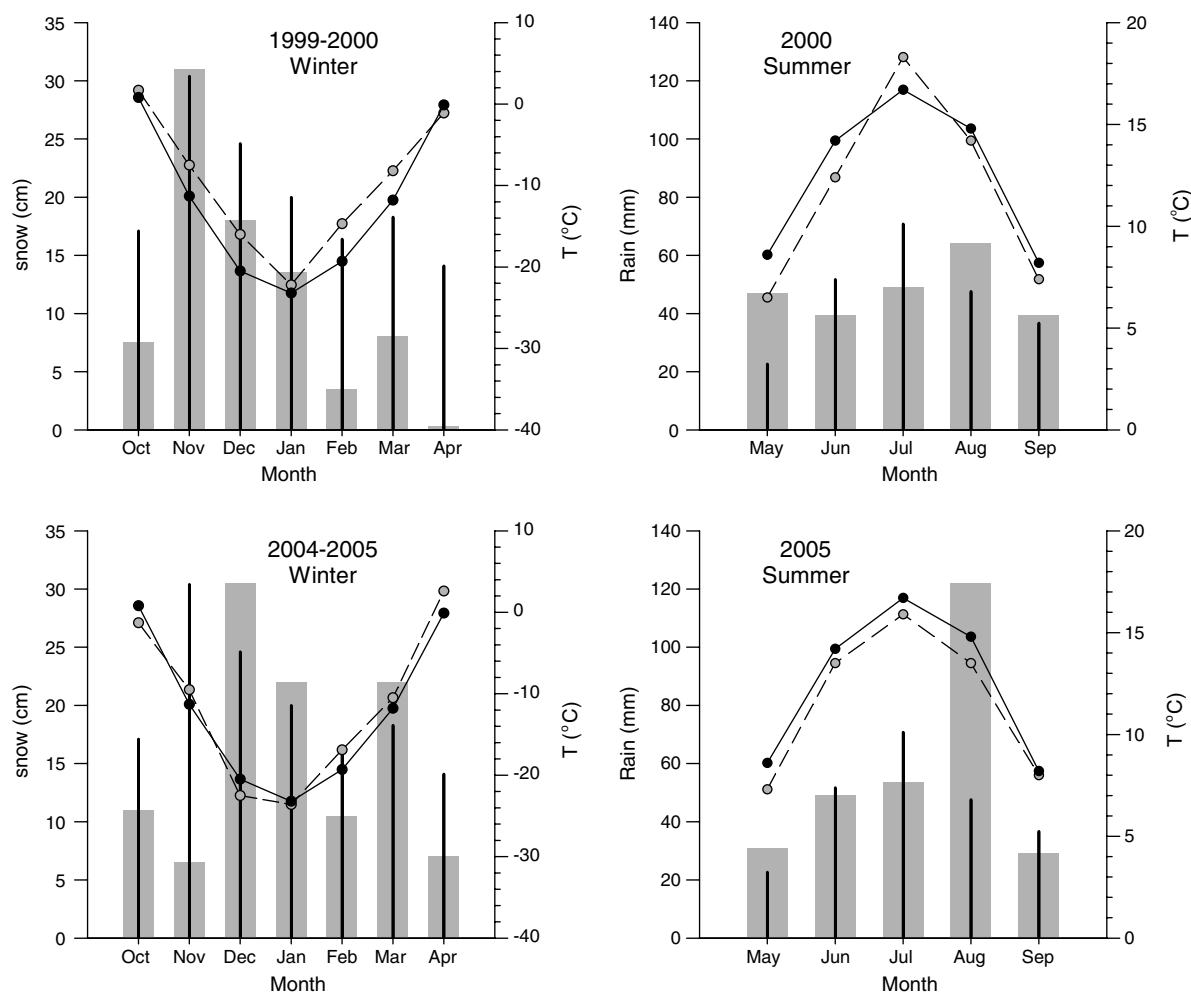


Figure 1 Location of the Peace-Athabasca Delta, Alberta, Canada and lake water sampling sites.

(especially from January to April); rainfall was evenly distributed throughout the following summer. In contrast, the win-

ter of 2004/2005 had much greater snowfall throughout the latter months of the season and greater rainfall in August.



**Figure 2** Winter and summer monthly mean temperature and accumulated precipitation for 1999/2000 and 2004/2005 compared to 1971–2000 climate normals. Climate normals are shown as solid circles (temperature) and bars (precipitation).

## Sampling and analysis

Water samples for oxygen and hydrogen isotope analysis were collected from 62 lakes on 25 October 2000 and most of these same lakes ( $n = 54$ ) were re-sampled on 14 September 2005 (Fig. 1). Samples were collected from approximately 10 cm below the surface at the center of each lake with the aid of a helicopter. All samples were sealed in 30 ml high-density polyethylene bottles and were analyzed at the University of Waterloo – Environmental Isotope Laboratory, where  $^{18}\text{O}/^{16}\text{O}$  and  $^2\text{H}/^1\text{H}$  ratios were measured using standard methods (Coleman et al., 1982; Drimmie and Heemskerk, 1993; Epstein and Mayeda, 1953). Results are reported in  $\delta$  values, representing deviations in per mil (‰) from VSMOW on a scale normalized to values of Standard Light Antarctic Precipitation ( $-55.5\text{‰}$  for  $\delta^{18}\text{O}$ ;  $-428\text{‰}$  for  $\delta^2\text{H}$ ; Coplen, 1996). Analytical uncertainties are  $\pm 0.2\text{‰}$  for  $\delta^{18}\text{O}$  and  $\pm 2.0\text{‰}$  for  $\delta^2\text{H}$ . To demonstrate the potential variability in input water isotope composition to lakes in the PAD, results from water samples collected over six years from two basins located in different hydrological settings (PAD 37 and PAD 54; Fig. 1) are also reported.

## Theory

### Isotopic labelling in the hydrological cycle

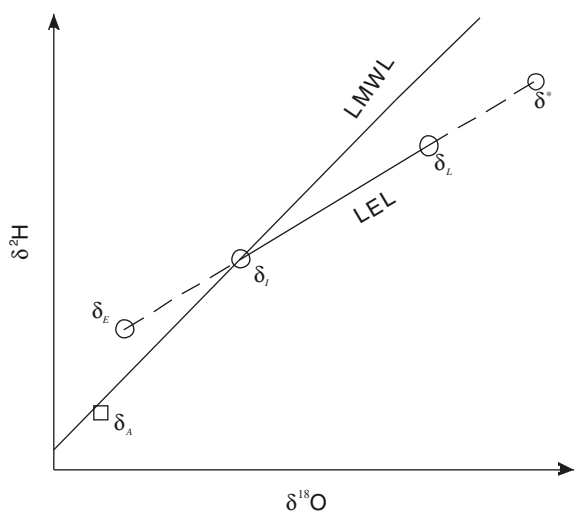
The distribution of water isotopes on the earth's surface (i.e., in precipitation and surface waters) is characterized by the existence of strong linear relations between  $\delta^2\text{H}$  and  $\delta^{18}\text{O}$  over a broad range of spatial and temporal scales, reflecting systematic mass-dependent isotope fractionation in the hydrological cycle (Rozanski et al., 1993; Gat, 1996; Gibson and Edwards, 2002). The most salient feature of the covariant behaviour between hydrogen and oxygen isotopes in the global water cycle is the global meteoric water line (GMWL), which is expressed by the linear function  $\delta^2\text{H} = 8\delta^{18}\text{O} + 10$  (Craig, 1961). The slope reflects the dominant influence of temperature-dependent equilibrium fractionation of heavy isotope species between atmospheric vapour and condensing precipitation, while the linearity is consistent with the notion that, at the global scale, atmospheric moisture primarily arises from one large water source (i.e., the subtropic ocean surface) and undergoes progressive distillation during poleward atmospheric transport. The GMWL has proven to be an especially useful reference line for understanding spatial patterns in the

variability of the isotopic composition of mean annual precipitation (Rozanski et al., 1993). Analogous labelling of precipitation occurs at local scale in the form of local meteoric water lines (LMWLs), reflecting temporal variability in the isotopic composition of local precipitation. LMWLs commonly lie close to the GMWL, usually with a lower slope because of raindrop re-evaporation and kinetic effects during snow formation (Rozanski et al., 1993).

The isotopic composition of water that has undergone evaporation diverges from the GMWL (or LMWL) and the variation of hydrogen- and oxygen-isotope signals in surface waters is systematic because of mass-dependent fractionation that occurs during isotopic enrichment. Typically, the isotopic compositions of neighbouring water bodies receiving input water similar in isotopic composition cluster along a well-defined linear trajectory termed a local evaporation line (LEL). In  $\delta^{18}\text{O}$ - $\delta^2\text{H}$  space, the slope of the LEL usually ranges between 4 and 6 and is primarily controlled by local atmospheric conditions including relative humidity ( $h$ ), temperature ( $T$ ) and the isotopic composition of atmospheric moisture ( $\delta_A$ ) (Gibson et al., in press). Moreover, the relative position of a given lake along the LEL is strongly associated with the water balance of the lake (Gonfiantini, 1986; Gat, 1996; Gibson and Edwards, 2002). Overall, the existence of two linear trends in  $\delta^{18}\text{O}$ - $\delta^2\text{H}$  space allows differentiation of meteoric water from surface water that has undergone secondary isotopic enrichment due to evaporation.

### Isotope-mass balance modelling

Quantitative investigations of lake water balances using water isotope tracers have been based on a conceptual framework incorporating individual water budget components in  $\delta^{18}\text{O}$ - $\delta^2\text{H}$  space (Fig. 3). For any given lake,  $\delta_L$ ,  $\delta_E$  and  $\delta_i$  represent the isotopic composition of the lake water, evaporative flux from the lake and the input water to the



**Figure 3** Schematic  $\delta^{18}\text{O}$ - $\delta^2\text{H}$  diagram identifying key isotopic parameters used in isotope-mass balance studies. These include lake water isotope composition ( $\delta_L$ ), input water isotope composition ( $\delta_i$ ), isotopic composition of evaporated vapour from the lake ( $\delta_E$ ) and the limiting isotopic composition ( $\delta^*$ ). The atmospheric moisture isotope composition ( $\delta_A$ ) is also shown.

lake, respectively.  $\delta_i$  is commonly estimated by the intersection of the LMWL with the best-fit line through a time-series of lake water isotope compositions or a spatial distribution of lake water isotope compositions in a watershed (e.g., Gibson et al., 1993). According to mass conservation,  $\delta_E$  should lie on the extension of the LEL to the left of the LMWL. Also shown in Fig. 3 is the isotopic composition of atmospheric moisture  $\delta_A$  and the limiting isotope composition  $\delta^*$ , which is the maximum isotopic enrichment attainable under local atmospheric conditions when a water body approaches desiccation.  $\delta^*$  is independent of hydrological conditions (i.e.,  $\delta_L$  and  $\delta_i$ ) but related to atmospheric conditions (i.e.,  $h$ ,  $T$  and  $\delta_A$ ; Welhan and Fritz, 1977; Allison and Leaney, 1982). Theoretically, regardless of the hydrological complexity of a landscape, the isotopic composition of all lakes in an area with similar atmospheric conditions will converge towards  $\delta^*$ .

### Isotopic composition of evaporated vapour ( $\delta_E$ )

Evaporated flux is a critical component for isotope mass-balance calculations but it is difficult to obtain samples directly for isotopic measurements. Studies have conventionally quantified  $\delta_E$  using the Craig and Gordon (1965) linear resistance model, which includes a molecular diffusion layer bounded below by a “virtually saturated” layer and above by a turbulent transportation layer. The fluxes through the molecular diffusion layer are linearly proportional to the vapour concentration difference between the upper and lower boundaries. As such, the vapour flux of  $^1\text{H}_2^{16}\text{O}$  (i.e., the common “light” water isotopologue) across the liquid–vapour interface is given by

$$E = (C_s - C_a)/\rho = C_s(1 - h)/\rho \quad (1)$$

where  $C_s$  is the vapour concentration in the “virtually saturated” layer,  $C_a$  is the vapour concentration in the turbulent transportation layer,  $h$  is the relative humidity of the turbulent layer normalized to the water surface temperature and  $\rho$  is a resistance coefficient (Gonfiantini, 1986; Gat, 1996). Similarly, the vapour flux of heavy water isotopologues (i.e.,  $^1\text{H}^2\text{H}^{16}\text{O}$  or  $^1\text{H}_2^{18}\text{O}$ ) across the same molecular diffusion layer is given by

$$E_i = (C_s R_s - C_a R_a)/\rho_i = C_s(R_s - h R_a)/\rho_i \quad (2)$$

Here, the subscript  $i$  designates water molecules bearing the heavy isotope species.  $R_s$  and  $R_a$  are the isotopic ratios of the water vapour in the saturated and turbulent layers, respectively. Since water vapour in the saturated layer should be in equilibrium with surface water at the saturated condition,  $R_s$  can be quantitatively related to the isotopic ratio of lake water ( $R_L$ ) by the definition of equilibrium fractionation:

$$R_L/R_s = \alpha^* \quad (3)$$

where  $\alpha^*$  is the liquid–vapour equilibrium fractionation factor (i.e.,  $\alpha^* > 1$ ). Isotopic fractionation is also commonly expressed by a separation term ( $\varepsilon^*$ ), where  $\varepsilon^* = \alpha^* - 1$  (for  $\varepsilon^*$  in decimal notation). The  $\varepsilon^*$  and  $\alpha^*$  parameters are dependent on temperature and can be calculated using empirical equations (Horita and Wesolowski, 1994).

According to the definition of the isotopic ratio,  $R_E$  can be expressed as  $R_E = E_i/E$ . By combining Eqs. (1)–(3),  $R_E$  can be expressed as

$$R_E = \frac{\rho}{\rho_i} \cdot \frac{R_L/\alpha^* - hR_A}{1 - h} \quad (4)$$

Introducing  $\varepsilon_k$ , the kinetic separation term according to Craig and Gordon (1965) and Gonfiantini (1986):

$$\varepsilon_k = (1 - h) \left( \frac{\rho_i}{\rho} - 1 \right) \quad (5)$$

Eq. (4) can then be rewritten in  $\delta$ -notation:

$$\delta_E = \frac{(\delta_L - \varepsilon^*)/\alpha^* - h\delta_A - \varepsilon_k}{1 - h + \varepsilon_k} \quad (6)$$

where  $\delta_L$  and  $\delta_A$  are the isotopic compositions of lake water and atmospheric moisture, and  $\varepsilon^*$  and  $\varepsilon_k$  are equilibrium and kinetic separation terms, as defined above.  $\varepsilon_k$  is estimated as a function of the relative humidity deficit, described elsewhere (Gonfiantini, 1986; Gibson and Edwards, 2002; Edwards et al., 2004). Note that Eq. (6) is formulated for  $\delta$ ,  $\varepsilon$  and  $h$  values in decimal notation. Eq. (6) can be further simplified on the basis of various assumptions (Welhan and Fritz, 1977; Allison and Leaney, 1982; Ferhi et al., 1983; Gat, 1996; Yakir and Sternberg, 2000; Gibson and Edwards, 2002; Vallet-Coulomb et al., 2006) but we apply Eq. (6) as it was presented by Gonfiantini (1986) and as shown here.

The quantitative evaluation of  $\delta_E$ , as expressed in Eq. (6), requires values for the isotopic composition of atmospheric moisture ( $\delta_A$ ), the isotopic composition of lake water ( $\delta_L$ ), relative humidity ( $h$ ) and temperature ( $T$ ). Of these parameters,  $\delta_L$ ,  $h$  and  $T$  can be measured routinely. Direct measurement of  $\delta_A$ , on the other hand, is difficult because sample collection is logistically challenging and can introduce unintended isotopic fractionation. Given the dynamic nature of the atmosphere, single sample of atmospheric moisture may not accurately represent an integrated isotopic mean value, especially in regions with strong seasonality (e.g., Jacob and Sonntag, 1991). Alternatively, several indirect methods have been used to estimate  $\delta_A$ . The most straightforward and frequently applied method is to assume isotopic equilibrium between evaporation-flux-weighted local precipitation and atmospheric moisture (Gibson, 2002). Although this method has been applied extensively in hydrological studies (Zuber, 1983; Gibson et al., 1993; Edwards et al., 2004), the assumption of equilibrium between precipitation and atmospheric moisture may not be valid if terrestrial recycling of evapotranspirative vapour is significant, as is the case in the Great Lakes region (Gat et al., 1994) and the Amazon basin (Gat and Matsui, 1991). A constant-volume pan can be used to derive  $\delta_A$  (Welhan and Fritz, 1977; Allison and Leaney, 1982; Gibson et al., 1999) but this requires careful maintenance of the pan over the investigation period, which may not be practical in remote regions. Where available, studies have used an index lake (e.g., Dinçer, 1968) as a natural analogue of a constant-volume pan to generate a time-integrated estimate of  $\delta_A$ .

Fortunately, our studies have identified a deep (>8 m) closed-drainage lake (PAD 18; see Fig. 1) that is an excellent index lake for the PAD region. The lake is perched well beyond the possible influence of river flooding, and hence is fed solely by precipitation. Several years (2000–2006) of field observation have revealed minimal interan-

nual variation in water level ( $\pm 0.2$  m) and near-constant isotopic composition ( $-9.1 \pm 0.5\text{‰}$  for  $\delta^{18}\text{O}$ ;  $-104 \pm 2\text{‰}$  for  $\delta^2\text{H}$ ), as expected if input is closely compensated by evaporation (i.e.,  $E/I \approx 1$ ) (Hall et al., 2004; Falcone, 2007; Wolfe et al., 2007). Based on catchment area and meteorological records, the lake water has an estimated mean residence time on the order of 10 years, which suggests that the system integrates precipitation inputs over multi-annual time-scales. The mean annual isotopic composition of precipitation based on the nearest monitoring station (Fort Smith, Northwest Territories,  $60^\circ\text{N}$   $112^\circ\text{W}$ ;  $-18.8\text{‰}$  for  $\delta^{18}\text{O}$ ;  $-147\text{‰}$  for  $\delta^2\text{H}$ , Birks et al., 2004) agrees very closely with shallow groundwaters sampled directly in the catchment of PAD 18 in 2003 ( $-18.5\text{‰}$  for  $\delta^{18}\text{O}$ ;  $-146\text{‰}$  for  $\delta^2\text{H}$ ). By solving Eq. (6) for both isotopes on the basis that the isotopic composition and mass of annual liquid input and thaw-season vapour output from PAD 18 are identical ( $\delta_E = \delta_i$ ), we obtain an estimate of  $\delta_A$  ( $-26.6\text{‰}$ ;  $-205\text{‰}$ ). Notably, this is very close to  $\delta_A$  ( $-25.4\text{‰}$ ;  $-211\text{‰}$ ) estimated previously by Wolfe et al. (2007) assuming equilibrium with thaw-season precipitation (Gibson and Edwards, 2002), and in good agreement with various ‘‘snapshot’’ estimates of  $\delta_A$  that we have obtained from field experiments using constant-volume pans (Falcone, 2007). As a result, we apply the index lake method to infer  $\delta_A$  in this paper.

### General lake water balance

The water-mass and isotope-mass balances of a well-mixed lake at hydrological and isotopic steady-state are (Gonfiantini, 1986; Gibson and Edwards, 2002):

$$I = Q + E \quad (7)$$

$$I\delta_i = Q\delta_Q + E\delta_E \quad (8)$$

where  $I$  is the inflow rate ( $\text{m}^3 \text{s}^{-1}$ ),  $Q$  is the outflow rate ( $\text{m}^3 \text{s}^{-1}$ ) and  $E$  is the evaporation rate ( $\text{m}^3 \text{s}^{-1}$ ).  $\delta_i$ ,  $\delta_Q$  and  $\delta_E$  represent the isotopic compositions of their corresponding hydrological components. Physical outflow does not cause isotopic fractionation, therefore outflow is isotopically equal to lake water (i.e.,  $\delta_Q = \delta_L$ ). By combining Eqs. (7) and (8), the evaporation-to-inflow ( $E/I$ ) ratio can be expressed using the isotopic compositions of each water budget component:

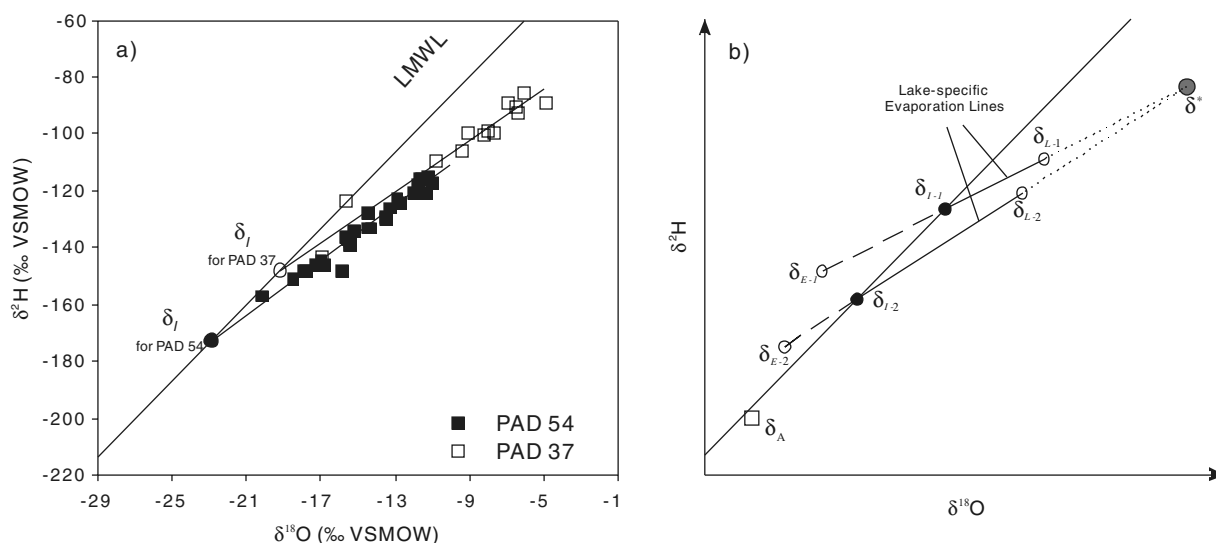
$$E/I = \frac{\delta_i - \delta_L}{\delta_E - \delta_L} \quad (9)$$

Eq. (9) has been used extensively to assess lake water balances (e.g., Dinçer, 1968; Gat and Levy, 1978; Gonfiantini, 1986; Gibson and Edwards, 2002; Vallet-Coulomb et al., 2006; Mayr et al., 2007).

## Results and discussion

### Developing a coupled isotope tracer method to characterize lake-specific input water

Isotopic composition of water samples collected from two lakes, PAD 37 and PAD 54, over a six-year period (2000–2005) show characteristic linear patterns when plotted in  $\delta^{18}\text{O}$ - $\delta^2\text{H}$  space (Fig. 4a). Although all samples plot below the LMWL ( $\delta^2\text{H} = 6.7\delta^{18}\text{O} - 19.2$ ; Birks et al., 2004), indicating varying degrees of the importance of evaporation to the



**Figure 4** (a) Lake water isotope compositions (2000–2005) for PAD 54 and PAD 37. Best-fit lines through these datasets intersect the LMWL at different points, suggesting that the lakes are fed by water of differing isotopic compositions. (b) Schematic  $\delta^{18}\text{O}$ - $\delta^2\text{H}$  diagram illustrating that the isotopic composition in a region that experiences similar atmospheric conditions will converge to the limiting isotopic composition ( $\delta^*$ ) with increasing evaporation, independent of  $\delta_i$ .

lake water balances, their respective trajectories are subtly offset, suggesting these two lakes are fed by input waters with slightly different time-integrated isotopic compositions ( $\delta_i$ ). For PAD 54, the estimated  $\delta_i$  ( $-22.9\text{‰}$  for  $\delta^{18}\text{O}$  and  $-173\text{‰}$  for  $\delta^2\text{H}$ ) is more depleted than that for PAD 37 ( $\delta^{18}\text{O} = -19.2\text{‰}$ ;  $\delta^2\text{H} = -148\text{‰}$ ), which is consistent with their different hydrological settings. PAD 54 is a deep ( $\sim 4$  m) oxbow lake near the Peace River, which is known to receive isotopically-depleted input from both occasional river floodwater and snowmelt runoff generated from its large contributing area. On the other hand, PAD 37 is a shallow basin ( $< 1$  m deep) in the central part of the PAD having minimal contributing catchment, and is thus more dependent on the input from rainfall, which is typically isotopically-enriched relative to snowmelt and river water. Indeed, at least one sampling episode appears to have captured almost complete replenishment of the lake by recent rainfall, judging by the sample point lying on the LMWL (Fig. 4a).

Variability in  $\delta_i$  can also affect the accuracy of isotope-based water balance assessments (see Eq. (9)). In this environment, individual lakes (such as PAD 37 and PAD 54) possess their own evaporation lines anchored by lake-specific input water isotope compositions. Moreover, in a region with similar, well-mixed atmospheric conditions (i.e.,  $h$ ,  $T$  and  $\delta_A$ ), the limiting isotope compositions ( $\delta^*$ ) for different lakes, regardless of lake water balance status (including  $\delta_i$ ), are predicted to be the same. As a result, all lake-specific LELs of varying  $\delta_i$  and slope will converge towards a common  $\delta^*$  (Fig. 4b).

In previous isotope-mass balance studies of lakes in the PAD (Wolfe et al., 2007), variability in  $\delta_i$  was recognized and estimated from the intersection of lake-specific LELs, drawn parallel to a regional predicted LEL, and the LMWL. This resulted in conservative estimates of  $\delta_i$  with respect to variability about the mean annual isotope composition

of precipitation and generated reasonable water balance estimates largely supported by limnological parameters. However, this approach is not consistent with the notion that all lakes in a region should tend towards the same  $\delta^*$ . Below, we present a more rigorous mathematical solution for  $\delta_i$  by assuming equivalent  $E/I$  ratios derived from both  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  signatures, thereby conserving both mass and isotopes.

As illustrated in Fig. 4b,  $\delta_i$  is expected to lie on the line between  $\delta_L$  and  $\delta_E$ .  $\delta_i$  can be further constrained to the LMWL, since the input water is primarily meteoric water that has not undergone evaporation. Rain and snow samples collected during 2000–2005 showed good agreement with this LMWL (Falcone, 2007). This leads to the expression below that directly couples oxygen and hydrogen isotope signatures from Eq. (9):

$$\frac{\delta^2 H_i - \delta^2 H_L}{\delta^2 H_E - \delta^2 H_L} = \frac{\delta^{18} O_i - \delta^{18} O_L}{\delta^{18} O_E - \delta^{18} O_L} \quad (10)$$

With calculation of  $\delta_E$  from Eq. (6) and measurement of  $\delta_L$ , Eq. (10) can be used to solve for a unique pair of  $\delta^{18}\text{O}_i$  and  $\delta^2\text{H}_i$  values by incorporating the LMWL. Herein, we refer to this approach as a coupled isotope tracer method for estimating input water isotope composition to lakes. In the following section, we test this approach by characterizing input waters to lakes in the PAD sampled at the end of two thaw seasons having contrasting hydrological and meteorological conditions.

#### Lake water input composition in the Peace-Athabasca Delta for 2000 and 2005

Isotopic compositions of lake water sampled in October 2000 and September 2005 show similar linear trends offset from the LMWL in  $\delta^{18}\text{O}$ - $\delta^2\text{H}$  space but different distribution patterns (Fig. 5a and b). For lake waters sampled in 2000 (Fig. 5a), the isotopic compositions range from  $-13.9\text{‰}$  to

–6.7‰ in  $\delta^{18}\text{O}$  (–124‰ to –89‰ in  $\delta^2\text{H}$ ) with the exception of PAD 45, which has a low isotopic composition (–17.3‰ and –139‰ for  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$ , respectively) and lies close to the LMWL. For 2005 (Fig. 5b), lake water isotope compositions range from –16.9‰ to –9.6‰ in  $\delta^{18}\text{O}$  (–137‰ to –105‰ in  $\delta^2\text{H}$ ). In general, lake water isotope compositions in 2005 are more depleted than for 2000 and exhibit less scatter ( $R^2$  is 0.88 for 2000 and 0.96 for 2005).

Applying the coupled isotope tracer method (Eq. (10)) to the 2000 and 2005 lake water isotope composition datasets yields lake-specific  $\delta_i$  values, integrated over the thaw season, for each corresponding  $\delta_L$  (Fig. 5a and b). Calculations revealed that  $\delta_i$  values for the two years span discrete (though overlapping) spectra along the LMWL. In 2000,  $\delta_i$  values vary from –22.7‰ to –12.0‰ in  $\delta^{18}\text{O}$  (–171‰ to –100‰ in  $\delta^2\text{H}$ ), while  $\delta_i$  values in 2005 range from –26.2‰ to –16.1‰ in  $\delta^{18}\text{O}$  (–195‰ to –127‰ in  $\delta^2\text{H}$ ).

Snowmelt, rainfall and river water are the three main sources of water to lakes in the PAD (Peters, 2003; Peters et al., 2006). It is expected that snowmelt and rainfall will affect all delta lakes to some extent, while influence from river water occurs perennially for lakes in direct connection with the channel network (i.e., open-drainage basins) and only during flood events for other lakes. These different types of source waters also bear distinct isotopic signatures based on extensive sampling of snow, rain and river water from 2000–2005. The isotopic composition of rain ranges from –20.5‰ to –13.9‰ for  $\delta^{18}\text{O}$  (–168‰ to –112‰ for  $\delta^2\text{H}$ ), whereas the isotopic composition of snow ranges from –30.6‰ to –19.4‰ for  $\delta^{18}\text{O}$  (–233‰ to –141‰ for  $\delta^2\text{H}$ ). While a wide range of isotopic compositions have been obtained for rain and snow, the isotopic composition of river water spans a much narrower range. The isotopic composition of the Athabasca River ranges from –19.3‰ to –17.5‰ for  $\delta^{18}\text{O}$  (–154‰ to –142‰ for  $\delta^2\text{H}$ ). The Peace River is isotopically depleted compared to the Athabasca River but also brackets a narrow range of values (–20.1‰ to –19.0‰ for  $\delta^{18}\text{O}$ ; –159‰ to –150‰ for  $\delta^2\text{H}$ ). For the convenience of illustration and discussion, we present the isotopic ranges of potential source waters to lakes (i.e., rain, snow and river water) as gray lines in Fig. 5.

The relative influence of the three main source waters to lakes can be evaluated by the position of  $\delta_i$  along the defined segments of the LMWL. Based on mixing, input

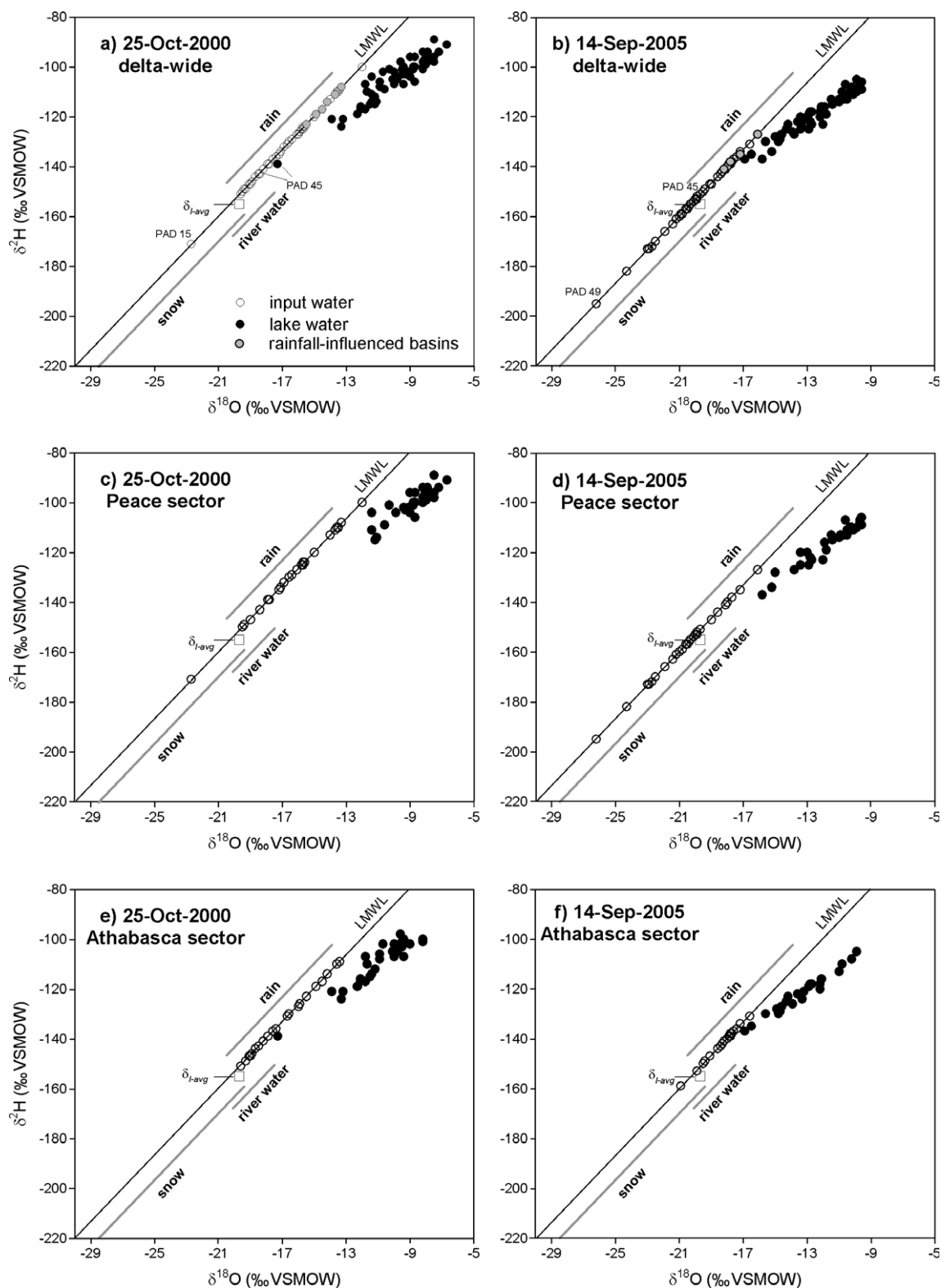
water with a high proportion of snowmelt will tend to be more isotopically depleted than input water composed of a high proportion of rainfall. Therefore, lakes with low  $\delta_i$  values, within the isotopic range of snow, would suggest that snowmelt plays an important role in the lake's water budget for that particular year, while higher  $\delta_i$  values would suggest a greater proportion of rainfall contributing to these lake basins. Because of the narrow isotopic variability of river water,  $\delta_i$  values for flooded lakes are expected to be similar to that of river water. This concept is readily demonstrated in the  $\delta_i$  results for lakes with well-understood and relatively simple water budgets. For instance, several lakes ( $n = 10$ ) that have previously been identified as rainfall-influenced basins based on their hydrolimnological characteristics and others, such as PAD 45 (Mamawi Lake; Fig. 1), are open-drainage basins that have permanent channel connections with major rivers (see Appendix D in Wolfe et al., 2007). The input to these basins is dominated by a single source of water (i.e., summer rainfall for the rainfall-influenced lakes and river water for PAD 45) and they have straightforward water budgets.  $\delta_i$  results for rainfall-influenced basins are positioned at the enriched end of the isotopic spectrum of input waters for both 2000 and 2005, whereas  $\delta_i$  for PAD 45 averages –18.7‰ in  $\delta^{18}\text{O}$  (–144‰ in  $\delta^2\text{H}$ ) over the two sample collection years, which is well within the range of its primary source of water, the Athabasca River (–19.3‰ to –17.5‰ for  $\delta^{18}\text{O}$ ; –154‰ to –142‰ for  $\delta^2\text{H}$ ) (Fig. 5a and b).

At the other end of the input water isotope spectrum, some lakes are primarily influenced by snowmelt runoff during the spring thaw. Based on the  $\delta_i$  results, PAD 15 and PAD 49 appear to be strongly influenced by snowmelt in 2000 and 2005, respectively (Fig. 5a and b). PAD 15 is a deep (~4 m) oxbow lake possessing catchment attributes similar to PAD 54 described above. Its large contributing area likely also provides substantial isotopically-depleted snowmelt runoff to the lake. PAD 49 is a small, shallow (~0.6 m), closed-drainage basin. Both of these lakes are surrounded by mature forests, which tend to accumulate snow during the winter, while the forest canopy intercepts direct rainfall during the thaw season. As a result, locally accumulated winter snowpack recharges these lakes rapidly during spring snowmelt and more gradually by sustained shallow groundwater flow over the thaw season. Examination of the spatial distribution pattern of  $\delta_i$

**Table 1** Input parameters used in Eqs. (6) and (10) to calculate lake-specific input water isotope composition

|  | 2000           | 2005           | Reference   |
|--|----------------|----------------|---|
| $h(\%)$  | 65.7           | 69.7           | Environment Canada (2004)   |
| $T$ (°C)   | 12.0           | 11.4           |   |
| $\alpha^*$ ( $^{18}\text{O}$ , $^2\text{H}$ )        | 1.0105, 1.0943 | 1.0106, 1.0951 | Horita and Wesolowski (1994)  |
| $\varepsilon^*$ ( $^{18}\text{O}$ , $^2\text{H}$ ) ‰ | 10.5, 94.3     | 10.6, 95.1     |   |
| $\varepsilon_k$ ( $^{18}\text{O}$ , $^2\text{H}$ ) ‰ | 4.9, 4.3       | 4.3, 3.7       | Gonfiantini (1986)  |
| $\delta_A$ ( $^{18}\text{O}$ , $^2\text{H}$ ) ‰      | –26.6, –205    | –26.6, –205    | Index lake method (based on average $h$ and $T$ of 67.7% and 11.7 °C) |

Thaw season temperature ( $T$ ) and relative humidity ( $h$ ) are based on evaporation flux-weighted mean daily values measured at Fort Chipewyan, Alberta. Note, all parameters are in decimal expression when applied in Eq. (6). For example, 60% = 0.6 for  $h$  values; –18‰ = –0.018 for  $\delta$  values and 10‰ = 0.01 for  $\varepsilon$  values.



**Figure 5**  $\delta^{18}\text{O}$ - $\delta^2\text{H}$  diagrams with measured lake water isotope compositions and calculated lake-specific input water isotope compositions using Eqs. (6) and (10) (see Table 1 for input parameters). (a) and (b) Delta-wide results for 25-October-2000 and 14-September-2005. The gray circles are for previously identified rainfall-influenced basins (Wolfe et al., 2007). Labelled sites are mentioned in the text. (c) and (d) Peace sector results for 25-October-2000 and 14-September-2005. (e) and (f) Athabasca sector results for 25-October-2000 and 14-September-2005. Note that drier and more variable conditions in 2000 led to greater overall enrichment and scatter in isotopic composition of lake waters. Isotopic ranges for snow, rain and river water are also shown (gray lines), as well as the average isotope composition ( $\delta_{i\text{-avg}}$ ) for the three water sources (Falcone, 2007). Note that these ranges lie on or are close to the LMWL, but are offset for graphic purposes only.

reveals that lakes with the lowest  $\delta_i$  values for the two sample collection years are all located in the northern Peace sector where closed-drainage basins are dominant and mature forest covers the landscape (Fig. 5c and d). Topographic relief from outcropping bedrock knolls in the northeast likely also captures snowdrifts. In comparison,  $\delta_i$  results indicate that no basins from the southern Athabasca sector were strongly influenced by snowmelt in either 2000 or 2005 (Fig. 5e and f), consistent with the wetland-covered, active floodplain landscape containing mainly restricted-drainage basins in this portion of the PAD. Instead, a small number of lakes in the Athabasca sector may have received river floodwaters from an ice-jam on the Athabasca River during the spring of 2005, which likely also accounts for the narrower range of  $\delta_i$  for these basins (of composition similar to the Athabasca River) compared to 2000 (Fig. 5e and f).

There are also notable differences in the distribution patterns of  $\delta_i$  for Peace sector basins between collection years 2000 and 2005 (Fig. 5c and d). The average isotopic composition of the three main sources of water (i.e., rain, snow and river water) is  $-19.7\text{‰}$  for  $\delta^{18}\text{O}$  and  $-155\text{‰}$  for  $\delta^2\text{H}$  (Falcone, 2007; also shown in Fig. 5). For lakes sampled in 2000, 30 of 31 (97%) have  $\delta_i$  values isotopically-enriched relative to this reference point (Fig. 5c). In contrast, only 9 of 30 (30%) lakes in 2005 have  $\delta_i$  values that are higher compared to this reference point (Fig. 5d). As shown in Fig. 2, the winter of 1999–2000 was relatively warm with less-than-average snowfall (especially from January to April) and summer 2000 rainfall was near-normal. In contrast, snowfall in 2004–2005 was much higher than 1999–2000, although subsequent total summer rainfall was similar. Consistent with these meteorological conditions, our  $\delta_i$  results indicate far greater relative importance of snowmelt to Peace sector lake water budgets in 2005 compared to 2000 (Fig. 5c and d).

## Conclusions

A coupled isotope tracer method is described to characterize the dual-isotope composition of input water ( $\delta_i$ ) to individual lake basins. Estimation of  $\delta_i$  is based on the

assumption that input waters fall along a definable LMWL in  $\delta^{18}\text{O}$ - $\delta^2\text{H}$  space and that mass and isotopes are conserved during evaporation, which is described by the linear resistance model of Craig and Gordon (1965). We demonstrate the viability of this technique by applying it in the Peace-Athabasca Delta to show that lakes receive input derived from varying proportions of rainfall, snowmelt and river water, which can be systematically related to lake catchment characteristics and hydrological settings, as well as antecedent meteorological conditions.

Our case study is drawn from late thaw-season lake water sample collections during two years of contrasting hydrological and meteorological conditions. Lake sat this time are likely to closely approach hydrologic and isotopic steady-state, although a similar approach could be applied under non-steady-state conditions. The method has the added advantage of potentially combining the identification of  $\delta_i$  with basin evaporation-to-inflow ratio assessments over large, hydrologically complex landscapes like deltas and floodplains from single point-in-time measurements, where widespread time-series isotopic sampling of individual basins is not feasible.

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## Appendix I

Water isotope composition of PAD lakes from two regional sampling campaigns

| Lake                    | Date            | $\delta^{18}\text{O}_L$ | $\delta^2\text{H}_L$ | Lake   | Date              | $\delta^{18}\text{O}_L$ | $\delta^2\text{H}_L$ |
|-------------------------|-----------------|-------------------------|----------------------|--------|-------------------|-------------------------|----------------------|
| <i>Athabasca sector</i> |                 |                         |                      |        |                   |                         |                      |
| PAD 19                  | 25-October-2000 | -8.2                    | -100                 | PAD 19 |                   |                         |                      |
| PAD 20                  | 25-October-2000 | -11.5                   | -115                 | PAD 20 | 14-September-2005 | -12.2                   | -120                 |
| PAD 21                  | 25-October-2000 | -9.4                    | -107                 | PAD 21 |                   |                         |                      |
| PAD 22                  | 25-October-2000 | -9.8                    | -105                 | PAD 22 | 14-September-2005 | -13.3                   | -124                 |
| PAD 23                  | 25-October-2000 | -10.1                   | -105                 | PAD 23 | 14-September-2005 | -11.0                   | -113                 |
| PAD 24                  | 25-October-2000 | -10.0                   | -107                 | PAD 24 | 14-September-2005 | -13.9                   | -126                 |
| PAD 25                  | 25-October-2000 | -13.3                   | -124                 | PAD 25 | 14-September-2005 | -14.8                   | -130                 |
| PAD 26                  | 25-October-2000 | -12.3                   | -119                 | PAD 26 | 14-September-2005 | -14.9                   | -128                 |
| PAD 27                  | 25-October-2000 | -9.5                    | -101                 | PAD 27 | 14-September-2005 | -9.9                    | -105                 |

## Appendix I (continued)

| Lake                | Date            | $\delta^{18}\text{O}_L$ | $\delta^2\text{H}_L$ | Lake     | Date              | $\delta^{18}\text{O}_L$ | $\delta^2\text{H}_L$ |
|---------------------|-----------------|-------------------------|----------------------|----------|-------------------|-------------------------|----------------------|
| PAD 28              | 25-October-2000 | -9.4                    | -100                 | PAD 28   | 14-September-2005 | -12.1                   | -116                 |
| PAD 29              | 25-October-2000 | -12.1                   | -117                 | PAD 29   | 14-September-2005 | -12.7                   | -118                 |
| PAD 30              | 25-October-2000 | -11.2                   | -112                 | PAD 30   | 14-September-2005 | -14.6                   | -127                 |
| PAD 31              | 25-October-2000 | -11.4                   | -114                 | PAD 31   | 14-September-2005 | -15.6                   | -130                 |
| PAD 32              | 25-October-2000 | -9.0                    | -102                 | PAD 32   | 14-September-2005 | -12.2                   | -118                 |
| PAD 33              | 25-October-2000 | -12.1                   | -116                 | PAD 33   | 14-September-2005 | -14.2                   | -123                 |
| PAD 34              | 25-October-2000 | -11.8                   | -117                 | PAD 34   | 14-September-2005 | -12.8                   | -118                 |
| PAD 35              | 25-October-2000 | -9.0                    | -102                 | PAD 35   | 14-September-2005 | -10.2                   | -108                 |
| PAD 36              | 25-October-2000 | -9.5                    | -103                 | PAD 36   |                   |                         |                      |
| PAD 37              | 25-October-2000 | -8.2                    | -101                 | PAD 37   | 14-September-2005 | -10.8                   | -110                 |
| PAD 38              | 25-October-2000 | -13.2                   | -121                 | PAD 38   | 14-September-2005 | -14.3                   | -125                 |
| PAD 39              | 25-October-2000 | -9.6                    | -98                  | PAD 39   | 14-September-2005 | -10.8                   | -110                 |
| PAD 40              | 25-October-2000 | -10.9                   | -108                 | PAD 40   | 14-September-2005 | -13.6                   | -122                 |
| PAD 41              | 25-October-2000 | -11.7                   | -110                 | PAD 41   | 14-September-2005 | -12.9                   | -119                 |
| PAD 42              | 25-October-2000 | -11.8                   | -107                 | PAD 42   | 14-September-2005 | -13.2                   | -121                 |
| PAD 43              | 25-October-2000 | -9.4                    | -100                 | PAD 43   |                   |                         |                      |
| PAD 44              | 25-October-2000 | -10.9                   | -106                 | PAD 44   |                   |                         |                      |
| PAD 45              | 25-October-2000 | -17.3                   | -139                 | PAD 45   | 14-September-2005 | -16.9                   | -137                 |
| PAD 46              | 25-October-2000 | -11.7                   | -110                 | PAD 46   | 14-September-2005 | -16.5                   | -135                 |
| PAD 47              | 25-October-2000 | -10.7                   | -102                 | PAD 47   |                   |                         |                      |
| PAD 59              | 25-October-2000 | -10.0                   | -102                 | PAD 59   |                   |                         |                      |
| PAD 62              | 25-October-2000 | -13.9                   | -121                 | PAD 62   | 14-September-2005 | -14.7                   | -129                 |
| <i>Peace sector</i> |                 |                         |                      |          |                   |                         |                      |
| PAD 1               | 25-October-2000 | -8.0                    | -94                  | PAD 1    | 14-September-2005 | -12.8                   | -122                 |
| PAD 2               | 25-October-2000 | -7.9                    | -98                  | PAD 2    | 14-September-2005 | -11.0                   | -114                 |
| PAD 3               | 25-October-2000 | -8.0                    | -99                  | PAD 3    | 14-September-2005 | -10.7                   | -113                 |
| PAD 4               | 25-October-2000 | -7.2                    | -94                  | PAD 4    | 14-September-2005 | -10.3                   | -110                 |
| PAD 5               | 25-October-2000 | -6.7                    | -91                  | PAD 5    | 14-September-2005 | -10.1                   | -111                 |
| PAD 6               | 25-October-2000 | -7.8                    | -96                  | PAD 6    | 14-September-2005 | -10.2                   | -110                 |
| PAD 7               | 25-October-2000 | -8.2                    | -98                  | PAD 7    | 14-September-2005 | -10.5                   | -111                 |
| PAD 8               | 25-October-2000 | -11.4                   | -111                 | PAD 8    | 14-September-2005 | -13.8                   | -127                 |
| PAD 9               | 25-October-2000 | -7.5                    | -96                  | PAD 9    | 14-September-2005 | -10.9                   | -113                 |
| PAD 10              | 25-October-2000 | -9.9                    | -104                 | PAD 10   |                   |                         |                      |
| PAD 11              | 25-October-2000 | -8.7                    | -100                 | PAD 11   | 14-September-2005 | -11.5                   | -113                 |
| PAD 12              | 25-October-2000 | -8.1                    | -97                  | PAD 12   | 14-September-2005 | -9.9                    | -110                 |
| PAD 13              | 25-October-2000 | -8.8                    | -100                 | PAD 13   | 14-September-2005 | -12.7                   | -123                 |
| PAD 14              | 25-October-2000 | -9.1                    | -102                 | PAD 14   | 14-September-2005 | -10.9                   | -113                 |
| PAD 15              | 25-October-2000 | -8.7                    | -106                 | PAD 15   | 14-September-2005 | -12.9                   | -125                 |
| PAD 16              | 25-October-2000 | -8.8                    | -101                 | PAD 16   | 14-September-2005 | -11.9                   | -116                 |
| PAD 17              | 25-October-2000 | -9.4                    | -102                 | PAD 17   | 14-September-2005 | -11.8                   | -119                 |
| PAD 18              | 25-October-2000 | -8.2                    | -100                 | PAD 18   | 14-September-2005 | -9.6                    | -106                 |
| PAD 48              | 25-October-2000 | -11.1                   | -114                 | PAD 48   | 14-September-2005 | -13.4                   | -125                 |
| PAD 49              | 25-October-2000 | -9.0                    | -104                 | PAD 49   | 14-September-2005 | -12.0                   | -123                 |
| PAD 50              | 25-October-2000 | -7.5                    | -98                  | PAD 50   | 14-September-2005 | -9.6                    | -109                 |
| PAD 51              | 25-October-2000 | -8.7                    | -96                  | PAD 51   | 14-September-2005 | -13.0                   | -120                 |
| PAD 52              | 25-October-2000 | -9.3                    | -103                 | PAD 52   | 14-September-2005 | -10.5                   | -113                 |
| PAD 53              | 25-October-2000 | -11.4                   | -104                 | PAD 53   | 14-September-2005 | -10.6                   | -107                 |
| PAD 54              | 25-October-2000 | -11.2                   | -115                 | PAD 54   | 14-September-2005 | -15.2                   | -134                 |
| PAD 55              | 25-October-2000 | -10.6                   | -109                 | PAD 55   |                   |                         |                      |
| PAD 56              | 25-October-2000 | -8.8                    | -101                 | PAD 56   | 14-September-2005 | -9.7                    | -107                 |
| PAD 57              | 25-October-2000 | -7.5                    | -89                  | PAD 57   | 14-September-2005 | -9.7                    | -108                 |
| PAD 58              | 25-October-2000 | -8.2                    | -94                  | PAD 58   | 14-September-2005 | -11.4                   | -115                 |
| PAD 60              | 25-October-2000 | -9.0                    | -96                  | PAD 60   |                   |                         |                      |
| PAD 61              | 25-October-2000 | -10.3                   | -101                 | PAD 61   | 14-September-2005 | -13.4                   | -120                 |
| PAD 13 A            |                 |                         |                      | PAD 13 A | 14-September-2005 | -15.8                   | -137                 |

## Appendix II

Water isotope composition of PAD 54 and PAD 37 between 2000 and 2006

| PAD 54            |                         |                      | PAD 37            |                         |                      |
|-------------------|-------------------------|----------------------|-------------------|-------------------------|----------------------|
| Date              | $\delta^{18}\text{O}_L$ | $\delta^2\text{H}_L$ | Date              | $\delta^{18}\text{O}_L$ | $\delta^2\text{H}_L$ |
| 25-October-2000   | -11.2                   | -115                 | 25-October-2000   | -8.2                    | -101                 |
| 04-March-2001     | -11.3                   | -121                 | 10-June-2001      | -8.0                    | -99                  |
| 04-June-2001      | -11.0                   | -117                 | 27-August-2001    | -6.5                    | -91                  |
| 27-August-2001    | -12.0                   | -121                 | 03-June-2002      | -9.4                    | -106                 |
| 03-June-2002      | -12.9                   | -123                 | 21-September-2002 | -9.0                    | -100                 |
| 24-July-2002      | -11.8                   | -118                 | 02-May-2004       | -16.9                   | -143                 |
| 24-August-2002    | -11.6                   | -116                 | 17-July-2004      | -4.9                    | -89                  |
| 21-September-2002 | -11.7                   | -116                 | 19-September-2004 | -15.7                   | -124                 |
| 01-May-2003       | -20.2                   | -157                 | 16-May-2005       | -15.5                   | -139                 |
| 22-May-2003       | -18.5                   | -151                 | 18-July-2005      | -7.7                    | -99                  |
| 04-June-2003      | -17.9                   | -148                 | 14-September-2005 | -10.8                   | -110                 |
| 16-June-2003      | -17.8                   | -148                 | 01-June-2006      | -6.4                    | -92                  |
| 03-July-2003      | -17.3                   | -146                 | 20-July-2006      | -6.1                    | -85                  |
| 15-July-2003      | -16.8                   | -146                 | 01-September-2006 | -7.0                    | -89                  |
| 23-September-2003 | -15.6                   | -137                 |                   |                         |                      |
| 02-May-2004       | -15.4                   | -138                 |                   |                         |                      |
| 11-June-2004      | -15.4                   | -139                 |                   |                         |                      |
| 16-July-2004      | -14.5                   | -128                 |                   |                         |                      |
| 28-July-2004      | -14.3                   | -133                 |                   |                         |                      |
| 17-September-2004 | -13.5                   | -130                 |                   |                         |                      |
| 19-September-2004 | -13.5                   | -129                 |                   |                         |                      |
| 12-March-2005     | -15.9                   | -148                 |                   |                         |                      |
| 16-May-2005       | -17.0                   | -145                 |                   |                         |                      |
| 18-July-2005      | -15.7                   | -136                 |                   |                         |                      |
| 28-July-2005      | -15.6                   | -137                 |                   |                         |                      |
| 14-September-2005 | -15.2                   | -134                 |                   |                         |                      |
| 01-June-2006      | -14.5                   | -133                 |                   |                         |                      |
| 20-July-2006      | -13.3                   | -126                 |                   |                         |                      |
| 01-September-2006 | -12.7                   | -124                 |                   |                         |                      |

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