HOCHAUFLÖSENDE ISOTOPENBASIERTE MODELLIERUNG DER GRUNDWASSERANBINDUNG MEROMIKTISCHER TAGEBAUSEEN

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Diplom Geographin Anne Gretje Seebach

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Hochauflösende isotopenbasierte Modellierung der Grundwasseranbindung meromiktischer Tagebauseen

vorgelegt von Anne Gretje Seebach

1. Gutachter:	Prof. Dr. Asaf Pekdeger Freie Universität Berlin
2. Gutachterin:	Prof. Dr. Gudrun Massmann Carl von Ossietzky Universität Oldenburg

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Lebenslauf

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Eidesstattliche Erklärung

Hiermit erkläre ich, Anne Gretje Seebach, die vorliegende Dissertation selbst verfasst und keine anderen als die von mir angegebenen Quellen und Hilfsmittel verwendet zu haben. Den benutzten Werken wörtlich oder inhaltlich entnommene Stellen sind als solche gekennzeichnet.

Anne Gretje Seebach

Berlin, im Sommer 2010

Warum sind der Himmel blau und die Wolken weiß?

Woher kommt der Regen?

Wie kommt das Salz in unsere Ozeane?

Warum gibt es die Alpen?

Was ist ein Isotop?

Wie viel Grundwasser fließt monatlich in meine drei Tagebaurestseen?

… dies ist nur ein Auszug der Fragen und Gedanken, den der eine oder andere Geowissenschaftler sich wohl von klein auf gestellt hat.

Einige Fragen beantworten sich durchs Erklärenlassen und Zuhören – anderen muss man selbst auf den Grund gehen. Das habe ich versucht. Hier ist der Beginn einer Antwort.

Vorwort und Dankeschön

Die vorliegende Arbeit wurde zwischen Dezember 2005 und März 2009 im Department Isotopenhydrologie am Helmholtz Zentrum für Umweltforschung UFZ unter der Betreuung von Dr. Kay Knöller erstellt. Sie basiert auf Untersuchungen, die innerhalb des von der Deutschen Forschungsgemeinschaft (DFG) geförderten Projektes „Hochauflösende Isotopenbasierte Modellierung der Grundwasseranbindung meromiktischer Tagebauseen“ als Teilprojekt im Verbundprojekt „Schichtungsmodellierung in Tagebauseen unter Berücksichtigung geochemischer Prozesse sowie von Vertikaltransport und Grundwasseranbindung“ durchgeführt wurden. Entsprechend der Projektvorgaben lag der Hauptschwerpunkt der Untersuchungen auf der Modellierung monatlicher Grundwassereinstromraten an drei Tagebaurestseen in der Niederlausitz. Hierzu erfolgten die isotopische Charakterisierung sämtlicher wasserbilanzrelevanten Komponenten und die Herleitung eines isotopenhydrologischen Wasserbilanzmodells, das für geschichtete Seen allgemein gültig ist.

Diese Arbeit konnte nur durch das Zusammenspiel glücklicher Zufälle, helfender Hände und dem unermüdlichen Glauben an die Übertragbarkeit „alltäglicher“ Isotopenbilanzmodelle auf permanent geschichtete Seen zustande kommen. So möchte ich mich an erster Stelle bei meinem Betreuer und Initiator des Projektes Kay Knöller für die unaufhörliche Geduld und das Vertrauen in meine Forschungsfähigkeit bedanken, die er mir in den vergangenen Jahren entgegen gebracht hat. Außerdem danke ich ihm für die Kreativität, die er aus mir heraus kitzeln konnte und für das Abenteuer Lausitz sowieso.
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Stephan Rübben und meiner Familie danke ich für die Liebe und Unterstützung, die sie mir in den Jahren entgegengebracht haben.

Zusammenfassung

An drei Untersuchungsstandorten in der Niederlausitz wurde versucht, der monatliche Grundwassereintrag isotopenhydrologisch (Sauerstoff-18, Deuterium) zu berechnen. Die Untersuchungen wurden an zwei meromiktischen sowie einem polymiktischen Tagebausee durchgeführt. Ziel der Untersuchungen war es, die für durchmischte Seen etablierte isotopenhydrologische Bilanzgleichung zu abzuwandeln und damit die Bilanzierung geschichteter Seen zu ermöglichen. Ein weiterer Aspekt waren Studien zur Modifizierung des Craig & Gordon Modells von 1965 (Craig und Gordon, 1965) und dessen Anpassung an Gebiete hoher Luftfeuchtigkeit.

Hierzu wurden über einen Untersuchungszeitraum von 2,5 Jahren zeitlich und räumlich hochaufgelöste Proben aus Grundwasser, Seewasser, Verdunstungspfannen, Niederschlag und Luftfeuchte genommen und isotopisch ($\delta^{18}O$, D) untersucht. Ferner wurden die meteorologischen Parameter Luftfeuchte und Lufttemperatur sowie die Oberflächenwassertemperatur der untersuchten Seen aufgezeichnet. Diese Daten wurden in einem Bilanzmodell zusammengeführt. Unglücklicherweise traten während des Untersuchungszeitraumes aufgrund der unbekannten hydrogeologischen Situation unvorhersehbare Schwierigkeiten mit der Bestimmung eines lokalen Backgroundwertes für die Isotopensignaturen des Grundwassers auf. Außerdem stellten die hydrodynamischen Rahmenbedingungen an allen drei Standorten die Bilanzierung vor eine Herausforderung. So schränken starke Seespiegelschwankungen durch den begleitenden Wechsel von effluenten und influenten Strömungsbedingungen die Anwendung der Isotopenbilanz enorm ein. Des Weiteren verliefen die Verdunstungsexperimente unter den lokalen meteorologischen Bedingungen nicht zufrieden stellend, was leider erst gegen ende des Untersuchungszeitraums erkannt wurde. Die Ergebnisse dieser Studie zeigen, dass die Anwendbarkeit des Isotopenbilanzmodells bezüglich der Quantifizierung von Grundwassereintragsraten durch die lokalen hydrogeologischen Verhältnisse stark limitiert werden kann. Dennoch konnte durch die umfassenden Gelände- und Laborarbeiten neben einem nahezu lückenlosen Datensatz für die drei Untersuchungsgebiete wertvolle Erkenntnisse zur Weiterführung dieses Projektes sowie zur Umsetzung bei zukünftigen ähnlichen Untersuchungen gewonnen werden. Hierzu zählen u.a. die Entwicklung einer Methode zur direkten isotopischen

Bestimmung von Wasserdampf bzw. Evaporat. Ferner konnten für die untersuchten Standorte hochaufgelöste Local Meteoric Water Lines (LMWL) und Evaporation Lines (EL) generiert werden. Darüber hinaus konnten Erkenntnisse über die heterogenen Verteilungsmuster von Isotopensignaturen im Grund- und Seewasser gewonnen werden.

1 Ausgangsfragen und Zielsetzungen des Projekts

Im Rahmen des Projektes „Hochauflösende isotopenbasierte Modellierung der Grundwasseranbindung meromiktischer Tagebauseen“ als Teil des Paketantrages „Schichtungsmodellierung in Tagebauseen unter Berücksichtigung geochemischer Prozesse sowie von Vertikaltransport und Grundwasseranbindung“ wurden Grundwasser – Seewasser – Wechselwirkungen an drei Tagebaurestseen in der Niederlausitz untersucht.

Die wesentlichen Zielstellungen waren:

- Monitoring und Erstellung der Datengrundlage für das Bilanzmodell,
- Erfassung der seeinternen räumlichen Differenzierung von Isotopenverteilungsmustern,
- Ermittlung von Gesetzmäßigkeiten des Isotopenaustauschs an Chemoklinen als Basis für die isotopenhydrologische Bilanzierung des Monimolimnions meromiktischer Seen,
- Isotopenhydrologische See-Bilanzierungen und Ableitung von Kenngrößen, die sich allgemein unter den klimatischen Bedingungen Mitteleuropas als Modellbasis anwenden lassen.

Hierbei lag der Schwerpunkt bei der Betrachtung der beiden meromiktischen Seen Waldsee und Moritzteich im Muskauer Faltenbogen [Fig.1 (Seebach und Knöller, 2010)]. Parallel dazu wurde der polymiktische Tagebaurestsee 107 (TBS107) bei Plessa als Kalibrierungsobjekt genutzt.
Der theoretische Ansatz der isotopenhydrologischen Seebilanzierung basiert auf der Gleichung

$$\frac{\Delta V_L \delta L + \Delta \delta_L V_L}{\Delta t} = P\delta_P - E\delta_E + I_S\delta_S - O_S\delta_L + I_{GW}\delta_{GW} - O_{GW}\delta_L \qquad (1)$$

mit δ als Isotopenverhältnis des Sees δ_L, des Niederschlages P, der Evaporation E, des Oberflächenzuflusses I_S und Oberflächenabflusses O_S sowie des Grundwassereintrages I_{GW}, des Grundwasserabstromes O_{GW} und des Volumens des Sees V über das Probennahmeintervall Δt.

2 Durchgeführte Feld- und Laborarbeiten

Probennahme

An den drei untersuchten Seen wurden zwischen Projektbeginn und Spätsommer 2008 Niederschlags-, Grundwasser- und Seewasserproben zur Untersuchung der stabilen Sauerstoff- und Wasserstoffisotope gesammelt. Die Probennahme erfolgte am Moritzteich und am Waldsee monatlich. Hierbei wurde der Chemoklinenbereich besonders hoch aufgelöst (10 cm). Das Seewasser des TBS107 wurde nur zweimonatlich beprobt, da davon auszugehen war, dass die Wassersäule stets gut durchmischt ist. Über dies hinaus wurden an allen drei Untersuchungsstandorten Verdunstungsexperimente durchgeführt; hierzu erfolgte die monatliche Beprobung von im See schwimmenden Verdunstungspfannen. Zur Bestimmung des relevantesten – da bislang unsichersten – Parameter der Isotopenbilanz, der Isotopensignatur des Evaporates δ_E, wurde eine Konstruktion entworfen, die die direkte Messung ermöglicht (Seebach et al., 2010). Am Waldsee und Moritzteich konnte dadurch eine (lückenhafte) Zeitreihe der Isotopie des Evaporates erhoben werden. Hierzu wurde alle 6 Stunden 30 Minuten lang mit einer Peristaltikpumpe die Luft oberhalb der Wasserfläche in Gasanalysebeutel gepumpt. Weitere zur isotopischen Seebilanzierung notwendige Parameter wurden im Rahmen des Teilprojektes „Untersuchungen zur Schichtungsentwicklung und komplexe Schichtungsmodellierung in Tagebauseen“ erhoben und zur Verfügung gestellt. Hierzu zählen die meteorologischen Parameter Luftfeuchte und Lufttemperatur sowie die Oberflächentemperatur des Seewassers. Da am TBS keine meteorologischen Daten aufgenommen wurden, konnte auf vergleichbare Werte vom 2 km entfernten TBS111 zurückgegriffen werden.

Laborarbeiten

Im Labor erfolgte die Evakuierung der Luftproben über eine Vakuumapparatur, wobei die Luftfeuchte in Kühlfallen ausgefroren werden konnte und isotopisch untersucht wurde. Die Isotopenanalyse aller Wasser- sowie der Luftfeuchteproben wurde im Department Isotopenhydrologie am Helmholtz Zentrum für Umweltforschung – UFZ durchgeführt.

3 Ergebnisse und Interpretation

Seewasser

Um die saisonale Variation in der Seewasserisotopie übersichtlicher darzustellen, wurden die gemessenen Isotopenprofile in 10 cm-Schritten interpoliert und die Schwankungsbreiten grafisch dargestellt [Fig.3 (Seebach und Knöller, 2010)]. Die gemessenen Profile gaben Auskunft über das Schichtungsverhalten der beiden meromiktischen Seen, bzw. belegten die Existenz des von der Atmosphäre abgeschlossenen Monimolimnions. Für $\delta^{18}O$ wurden im Moritzteich saisonale Schwankungen zwischen -7,77 ‰ und -2,44 ‰ im Mixolimnion bzw. -8,83 ‰ und -3,51 ‰ im Monimolimnion gemessen. Der Waldsee zeigte saisonale $\delta^{18}O$-Isotopenvariationen von -4,65 ‰ im Oberflächenwasser und -4,68 ‰ unterhalb der Chemokline. Auch am TBS107 waren $\delta^{18}O$-Isotopenvariationen im Jahresverlauf zu beobachten. Hier schwankten die Werte zwischen -4,16 ‰ im Seetiefsten und +1,17 ‰ in sommerlichem Oberflächenwasser.

Die Leitfähigkeitsmessungen, die im Rahmen des Teilprojektes „Untersuchung von Vertikaltransport und Grundwasseranbindung in Tagebauseen mittels Schwefelhexafluorid (SF6)“ durchgeführt wurden, zeigten, dass sich die Tiefenlage des Chemoklinenbereiches in Waldsee und Moritzteich im Jahresverlauf verändert. Am Waldsee sank die Chemokline in den Frühjahrsmonaten um etwa 0,6 m durch konvektive Erosion ab. Dies lässt sich durch die extremen Temperatursprünge zwischen Tag und Nacht erklären, die mit fortschreitendem Sommer immer größer werden und die Vertikalmischung des Mixolimnions bis an die Chemokline hinab reichen lassen (Von Rohden et al., 2009). Zum Winter hin wurde ein allmählicher Anstieg der Chemokline unterschiedlicher Dimensionen beobachtet, der auf jährlich unterschiedlich ausfallenden Grundwasserzustrom zurückgeführt wird. Am Moritzteich sank die Chemokline zu den Wintermonaten hin ab. Das im Vergleich mit dem Waldsee sehr große Mixolimnion kühlt zum Herbst hin ab und führt durch seine winterliche Vertikalmischung und windinduzierte Konvektion zur Chemoklinenerosion. Der sommerliche Anstieg der Chemoklinenregion um etwa 0,5 m pro Jahr ist auf einströmendes Grundwasser zurückzuführen.

Grundwasser

Die untersuchten Grundwassermessstellen wiesen innerhalb des Untersuchungszeitraumes ein weites Spektrum von $\delta^{18}O$-Signaturen auf. So ließen sich bei den sieben Pegeln am Waldsee Werte zwischen -9,59 ‰ (MWS3) und -5,53 ‰ (MWS4) nachweisen. Pegel MWS6 (Mittelwert -8,28 ‰) und MWS7 (Mittelwert -8,41 ‰) am Westufer zeigten die leichtesten Isotopensignaturen im Gegensatz zu Pegel MWS4 am Südzipfel des Waldsees. Die $\delta^{18}O$-Isotopenverteilungsmuster im Grundwasser am Moritzteich streute zwischen -9,73 ‰ (MMT3) und -5,76 ‰ (MMT9). Anhand der gemessenen Werte konnten die 17 Beobachtungspegel in drei Gruppen unterteilt werden. Die erste Gruppe fasst Pegel zusammen, in denen stets Isotopenwerte bestimmt wurden, die isotopisch leichter waren als das Seewasser. In einer zweiten Gruppe konnten Pegel geclustert werden, deren Isotopenwerte schwerer als das des Sees waren. Die dritte Gruppe umfasst die übrigen Pegel, deren Isotopenwerte durch signifikante Schwankungen geprägt waren. Am TBS107 variierten die $\delta^{18}O$-Werte während des Untersuchungszeitraums zwischen -0,93 ‰ (M107-1) und -9,69 ‰ (M107-4). Hierbei grenzten sich die Pegel M107-3, M107-4 und MPL-11 mit isotopisch leichten Grundwasserproben ($\delta^{18}O$ -9,34 ‰ bis -8,47 ‰) deutlich von den isotopisch angereicherten Wasserproben ($\delta^{18}O$ -5,38 ‰ bis -1,85 ‰) aus den Pegeln M107-1, M107-2 und MPL-1 ab.

Niederschläge

Die an den drei Standorten gesammelten Niederschlagsproben wiesen große Ähnlichkeiten bezüglich ihrer Isotopie als auch ihrer jährlichen Verteilung auf. So differierten die $\delta^{18}O$-Werte am Waldsee zwischen -1,32 ‰ und -13,82 ‰, am Moritzteich zwischen -1,70 ‰ und -13,00 ‰ und am TBS107 zwischen -3,69 ‰ und -15,90 ‰. Monatliche Niederschlagshöhen schwankten zwischen 7 mm am Moritzteich im Mai 2008 und 136 mm am Waldsee im Mai 2007.

Evaporation

Anhand der Verdunstungspfannen konnten für alle Untersuchungsgebiete die monatlichen Evaporationshöhen ermittelt werden. Am TBS107 schwankten die Werte zwischen 3 mm im Dezember 2007 und 124 mm in Juni 2008. Am Waldsee lagen die

Verdunstungshöhen zwischen 10 mm im Dezember 2006 und 124 mm in Juli 2006. Für den Moritzteich wurden Werte zwischen 7 mm (Februar 2007) und 111 mm (März 2007) berechnet. An allen Seen korrelierten die meteorologischen Bedingungen sehr gut mit den aufgezeichneten Seespiegelschwankungen.

An Waldsee und Moritzteich konnten für zahlreiche Probennahmeintervalle Datensätze für die Isotopie des Evaporates gewonnen werden. So schwanken die $\delta^{18}O$-Werte für das gewonnene Waldsee-Evaporat zwischen -24,07 ‰ und -16,36 ‰. Für den Moritzteich konnten Werte zwischen -25,41 ‰ und -7,36 ‰ ermittelt werden.
Für Zeiträume, für die keine Werte erhoben werden konnten, wurde ein best-fit-model mit gängigen Gleichungen [Fig.5a (Seebach und Knöller, 2010)] zur rechnerischen Bestimmung der Isotopenverhältnisse des Evaporates abgeleitet. Hierzu wurden die berechneten δD- und $\delta^{18}O$-Werte des Evaporates in einem D/O-Diagramm aufgetragen und die korrespondierenden Evaporationslinien (EL) generiert. Die aus Gleichung (8) [aus (Seebach und Knöller, 2010)] erhaltene EL liegt auf derjenigen, die aus den gemessenen Seewasserproben resultiert. Aus diesem Grunde wurde Gleichung (8) für die Berechnung der unvollständigen Datensätze der Isotopensignatur des Evaporates ausgewählt. Selbst wenn die im Rahmen dieses Projektes entwickelte Methode zur direkten isotopischen Bestimmung des Evaporates Unsicherheiten hinsichtlich der technischen Durchführung birgt, liegen die ermittelten Werte in einem realistischeren Bereich als die durch das Craig & Gordon Modell erhaltenen. Fig.. 5b [aus (Seebach und Knöller, 2010)] zeigt den Vergleich zwischen berechneten und gemessenen Datensätzen. Die Streuung der berechneten Datensätze weit in die positiven X/Y-Achsenabschnitte hinein wird durch den Parameter Luftfeuchte (%) hervorgerufen, der bei Werten >~80 % große Unsicherheiten in der rechnerischen Bestimmung verursachen kann.
Fig. 6 [aus (Seebach und Knöller, 2010)] vermittelt einen Eindruck der unterschiedlichen Größenordnungen zwischen direkt gemessenen und berechneten Werten im zeitlichen Verlauf des Untersuchungszeitraums. Es ist offensichtlich, dass die Abweichung zwischen den Datensätzen zu Perioden hoher Luftfeuchtigkeit zunimmt. Trotzdem werden fehlende Messwerte in der folgenden Seebilanzierung durch berechnete Daten substituiert.

Isotopenbilanz

Um bei den geschichteten Seen Mixolimnion und Monimolimnion separat voneinander bilanzieren zu können, wurden unten stehende Gleichungen aufgestellt. Bei diesem Ansatz wurde davon ausgegangen, dass während des saisonalen Absinkens der Chemokline Wasserpakete vom Monimolimnion ins Mixolimnion eingebracht werden. Der Wiederanstieg der Chemokline hingegen, der maßgeblich durch den Grundwasseranstrom ins Monimolimnion induziert wird, wirkt sich theoretisch nur auf die Bilanzgleichung des Monimolimnions aus.

$$I_{GW_MIX} = \frac{\Delta\delta_{MIX} V_{MIX} + E\delta_E - P\delta_P - I_S\delta_S + \delta_{MIX}(P - E + I_S) + V_C(\delta_{MIX} - \delta_C)}{\delta_{GW} - \delta_{MIX}} \quad (2a)$$

$$I_{GW_MON} = \frac{\Delta\delta_{MON} V_{MON} - \Delta V_C(\delta_{MON} - \delta_C)}{\delta_{GW} - \delta_{MON}} \quad (2b)$$

V_C und δ_C stellen hierbei die Wassermenge im schwankenden Chemoklinenbereich dar, die entsprechend der Chemoklinenfluktuation vom Monimolimnion ins Mixolimnion übergeht.

Fig. 7 [aus (Seebach und Knöller, 2010)] zeigt die berechneten monatlichen Grundwassereintragsraten. Als Eingangsparameter für den isotopischen lokalen Backgroundwert des Grundwassers in die Bilanzmodelle des Mixolimnions bzw. Monimolimnions wurden die in Tab.3 [aus(Seebach und Knöller, 2010)] aufgeführten Isotopenwerte berücksichtigt.

Die in Fig. 7 [aus (Seebach und Knöller, 2010)] dargestellten rechnerischen Ergebnisse des Grundwassereintrages zeigen, dass die Bilanzierung von Seen nur erfolgreich durchgeführt werden kann, wenn zuströmendes Grundwasser einem isotopisch definierbaren Grundwasserleiter zugeordnet werden kann. Da die beprobten Grundwasserbeobachtungsrohre in den Untersuchungsgebieten allerdings hinsichtlich der Isotopie des Grundwassers sehr heterogene Verteilungsmuster aufweisen, ist davon auszugehen, dass es sich um mehrere unterschiedliche Grundwasserleiter handelt, die die Seen speisen. Damit scheint es sich bei den analysierten Seewasserproben um Mischungswässer unterschiedlicher Herkunft zu handeln. Fragwürdig sind auch die negativen Eintragsraten. Auch in Arbeiten von Welhan & Fritz (Welhan und Fritz, 1975) und Yehdegho (Yehdegho et al., 1997)

werden negative Grundwassereintragsraten präsentiert. Wie diese Daten zu bewerten sind, konnte von Welhan & Fritz nicht begründet werden. Yehdegho et al. erhielten für ihre Studien am Schwarzl See in Österreich negative Werte nur für Phasen der Eisbedeckung (Januar bis Februar). Durch Modifikation der berücksichtigten Seewasserisotopenwerte wurden allerdings Grundwassereintrags-raten bestimmt, die gering positive Werte aufwiesen. Während die Mehrzahl der Autoren die Unsicherheit des rechnerisch bestimmten Isotopenwertes des Evaporates als limitierende Größe hervorheben, betonen Yehdegho et al., dass in ihren Berechnungen die Isotopenverhältnisse des Seewassers als Eingangsgröße kritisch zu betrachten sei.

4 Grenzen des Isotopenbilanzmodells

Hydrogeologische Rahmenbedingungen

Einen wesentlichen Eingangsparameter des isotopenhydrologischen Bilanzmodells stellt die Isotopensignatur des lokalen Grundwassers dar. Wie in Kapitel 2 beschrieben, sind die Isotopenverteilungsmuster im Untersuchungsgebiet Muskauer Faltenbogen sehr heterogen. Zudem war die hydrogeologische Situation im Untersuchungsgebiet zu Projektbeginn unbekannt. Die Auswahl der zu bilanzierenden Seen richtete sich vorrangig nach dem Schichtungsverhalten der Gewässer. Erst im Rahmen der geologischen Erkundung (Pechstein, 2008) konnte eine kleinskalige Abschätzung des geologischen Aufbaus vorgenommen werden. In diesem Prozess wurde erstmalig ein Versuch unternommen, die beprobten Grundwassermessstellen sowohl am Moritzteich als auch am Waldsee den unterschiedlichen Grundwasserleitern zuzuordnen. Die Verschneidung der Isotopendaten mit den hydrodynamischen Aspekten (Einmessung der Pegel zu m NHN und Relation zu den Seewasserspiegeln) zeigte, dass am Waldsee aus hydrodynamischer Perspektive nur 3 Pegelstandorte als Grundwasser-zuflussrichtungen in Frage kommen. Hierzu zählen die Pegel MWS2, MWS3 und MWS4. Die isotopenhydrologische Betrachtung hingegen lässt darauf schließen, dass der Grundwasserzustrom lediglich aus der Richtung von ostsüdöstlicher MWS3 erfolgen kann. Entscheidend ist aber die Tatsache, dass dem Waldsee etwa 100 m weiter südlich ein weiterer Tagebaurestsee vorgelagert ist. Da der Gradient zwischen

dem Grundwasserspiegel in MWS4 und dem Seespiegel zu Beginn der Untersuchungen konstant in Richtung Waldsee verlief, könnte davon ausgegangen werden, dass bereits isotopisch angereichertes Wasser aus dem vorgelagerten See über den Pegel MWS4 in den Waldsee hinein fließt.

Die Situation am Moritzteich verhält sich anders. Dort erweckt die hydrodynamische Kartierung den Eindruck, der Grundwasserzustrom erfolge nahezu restloch-umlaufend. Aus isotopenhydrologischer Perspektive stellt sich ähnlich wie beim Waldsee die Frage, inwiefern Pegel MMT5 die Isotopie des Seewassers beeinflusst. Das Grundwasser aus diesem Pegel ist hinsichtlich $\delta^{18}O$ durchschnittlich um 1,26 ‰ schwerer als das Seewasser im Monimolimnion und um 2,70 ‰ leichter als das Mixolimnionwasser. Ebenso fragwürdig ist Pegel MMT9 am Westufer des Moritzteiches. Hier könnte bereits angereichertes Wasser aus dem nahe gelegenen Grabensystem infiltriert sein.

Eine weitere Unsicherheit im Bilanzmodell stellen die erheblichen saisonalen Seespiegelschwankungen (WS 0,19 m; MT 0,41 m) dar. Mindestens in den Sommermonaten kommt es zu einer Umstellung der hydraulischen Gradienten. Herrschen in den Grundwasserneubildungsmonaten effluente Strömungsverhältnisse vor, kehren sich diese zu Beginn der Vegetationsperiode um, wobei das Seewasser nun in die Grundwasserleiter infiltriert. Allein dieser Aspekt verhindert die zuverlässige Bilanzierung von Grundwassereinstromraten.

Meteorologische Rahmenbedingungen

Publizierte Untersuchungen, die sich mit der isotopenhydrologischen See-bilanzierung befassen, wurden vornehmlich in Gebieten mit niedrigen Luftfeuchtigkeiten und oder zur Erhebung der Evaporationshöhe durchgeführt. Hierzu zählen maßgeblich die Arbeiten von Gibson et al. (Edwards et al., 2002; Gibson, 2001; Gibson, 2002; Gibson und Edwards, 1999; Gibson et al., 1993a; Gibson et al., 1993b; Gibson et al., 1996a; Gibson et al., 1999; Gibson et al., 1996b; Gibson et al., 1998), Dinçer (Dinçer, 1968) und Gat (Gat, 1970). Jedes untersuchte Gebiet hatte durch lokale Rahmenbedingungen den Vorteil, dass einige wichtige Parameter vernachlässigt werden konnten So erlaubten bspw. die Untersuchungen von Gibson, den Grundwassereinstrom in den Wintermonaten zu ignorieren (Permafrostbedingungen). Lediglich Welhan (Welhan und Fritz, 1975), Yehdegho et

al. (Yehdegho et al., 1997) und Knöller (Knöller und Strauch, 2002) verfolgten den Ansatz, anhand des Bilanzmodells Grundwassereintragsraten zu bestimmen.

Technische Rahmenbedingungen

Die erfolgreiche Durchführung von Langzeitfeldexperimenten ist maßgeblich von den Probennahmeintervallen abhängig. So konnte auf anfängliche technische Probleme mit der Luftfeuchtesammeleinheit nicht kurzfristig reagiert werden. Oftmals ging durch diese unvorhersehbaren Komplikationen nicht nur der Datensatz des vergangenen Probennahmeintervalls sondern auch der Datensatz des Folgemonats verloren. Über dies hinaus wurde erst während der Datenauswertung ersichtlich, dass die zeitliche Auflösung Probennahmeintervalle für die Verdunstungs-experimente nicht hoch genug war. Um detaillierte Aussagen über den Verdunstungsprozess aus isotopenhydrologischer Perspektive treffen zu können, sind tägliche Beprobungen in langen niederschlagsfreien Intervallen unabdingbar. Dies zu realisieren wäre im Rahmen dieses Projektes technisch und logistisch nicht möglich gewesen.

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Estimation of lake water – groundwater interactions in meromictic mining lakes by modelling isotope signatures of lake water

ANNE SEEBACH*†, SEVERINE DIETZ‡, DIETER LESSMANN‡ and KAY KNOELLER†

†Department of Isotope Hydrology (ISOHYD), Helmholtz Centre for Environmental Research – UFZ, Theodor-Lieser-Street 4, 06120 Halle/Saale, Germany
‡Department of Freshwater Conservation, Brandenburg University of Technology, PO Box 101344, 03013 Cottbus, Germany

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A method is presented to assess lake water–groundwater interactions by modelling isotope signatures of lake water using meteorological parameters and field data. The modelling of δ^{18}O and δD variations offers information about the groundwater influx into a meromictic Lusatian mining lake. Therefore, a water balance model is combined with an isotope water balance model to estimate analogies between simulated and measured isotope signatures within the lake water body. The model is operated with different evaporation rates to predict δ^{18}O and δD values in a lake that is only controlled by weather conditions with neither groundwater inflow nor outflow. Comparisons between modelled and measured isotope values show whether the lake is fed by the groundwater or not. Furthermore, our investigations show that an adaptation of the Craig and Gordon model [H. Craig, L.I. Gordon. Deuterium and oxygen-18 variations in the ocean and the marine atmosphere. In *Stable Isotopes in Oceanographic Studies and Paleotemperature, Spoleto*, E. Tongiorgi (Ed.), pp. 9–130, Consiglio Nazionale delle Ricerche, Laboratorio di Geologia Nucleare, Pisa (1965).] to specific conditions in temperate regions seems necessary.

Keywords: Craig–Gordon model; Deuterium; Evaporation; Hydrological modelling; Isotope mass balance; Mining lake; Oxygen-18; Water balance

1. Introduction

Widespread surface mining activities in the Lusatian Mining District in Germany caused significant changes to the aquatic systems. In combination with the flooding of former opencast pit areas, the occurrence of pyrite oxidation in dump sediments subjected the lakes to continuous solute inflow and acidification. Pyrite present in the lignite-bearing sediment strata is aerated and oxidised when the surrounding sediment layers are removed and mixed. The stored

*Corresponding author. Email: anne.seebach@ufz.de

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oxidation products are mobilised by seeping rainwater and/or by the rising groundwater table and lead to acidic conditions and density gradients in mine pit lakes [2]. The common range of pH values in acidic mining lakes of Lusatia varies between 2 and 4 [3].

Oxygen-18 and deuterium are environmental tracers that are used to follow the hydrological cycle at different scales [4]. The stable water isotopes not only allow the determination of precipitation origin [5] but also the relative contributions of different sources to the groundwater [6] – in addition, the applicability of the isotope mass balance approach has been well proven within the last decades for the estimation of water balances of lakes and catchments all over the world [7]. The range of study sites contains artificial lakes [8–12] as well as natural lakes like sinkholes [7] or wetland-dominated river basins [13, 14]. Since the appliance of that tool has been growing so successfully, many different approaches of sampling methods have been established both on different spatial and time scales (e.g. daily [15, 16] to monthly [10] time steps). The combination of diverse conventional methods (e.g. chemical mass balance) and the isotope water balance [9, 10] may help to evaluate both water budgets and environmental impacts like the contamination or acidification of water bodies. However, for all that, there are still limitations to the isotope mass balance model in particular for determining the isotopic composition of atmospheric water vapour [17].

In this context, the mixing dynamics of mining lakes ought to be investigated. A present research project is concerned with the stratification of water bodies, in particular of meromictic mining lakes. The application of stable oxygen isotope methods will be used to understand the mixing dynamics within the lakes and the interactions between groundwater and lake water. At the study site, however, the exchange between lake water and groundwater is still undefined. Therefore, the dimension of groundwater inflow and outflow has to be estimated. Usually, for lakes in this region, net evaporation is higher than precipitation plus surface inflow. In those cases, it is obvious that the lake could never exist without any groundwater inflow. However, due to specific local conditions, net evaporation rates can differ considerably from the established mean value of the region. On the other hand, the geological setting is complex and the occurrence of clay and lignite layers may indicate that the lake basin is separated from the groundwater aquifer. These aspects were the background for the idea to run a simulation for isotope signatures of the lake water as a preliminary assessment of the groundwater situation in the study area and for answering the question if this lake can even be an adequate object for the actual research project.

Therefore, a simplified model based on meteorological variables was used. The aim of that approach was to show the theoretical long-term development of the isotope composition within the lake water and the simplified calculation of the amount of inflowing groundwater. The model was run at two different levels. At the beginning, the isotope development of the lake water was simulated with different evaporation rates. This procedure based on the assumption that there is neither groundwater inflow nor outflow to predict the isotope signatures of a lake that is only driven by meteorological parameters. The calculations depended on varying evaporation rates with a monthly time step. The comparison between the simulated δ^{18}O and δD signatures and a data set of δ^{18}O and δD values measured in 2006 and 2007 was used to decide whether the lake water body is interacting with the groundwater.

2. Study site

The study site Lake Waldsee is located within the Lower Lusatian Lignite Mining District, Germany. This region is an ice-pushed ridge of a moraine formed by the Middle Pleistocene Elster-glacial, called Muskauer Faltenbogen (figure 1). Horizontal tertiary deposits have been

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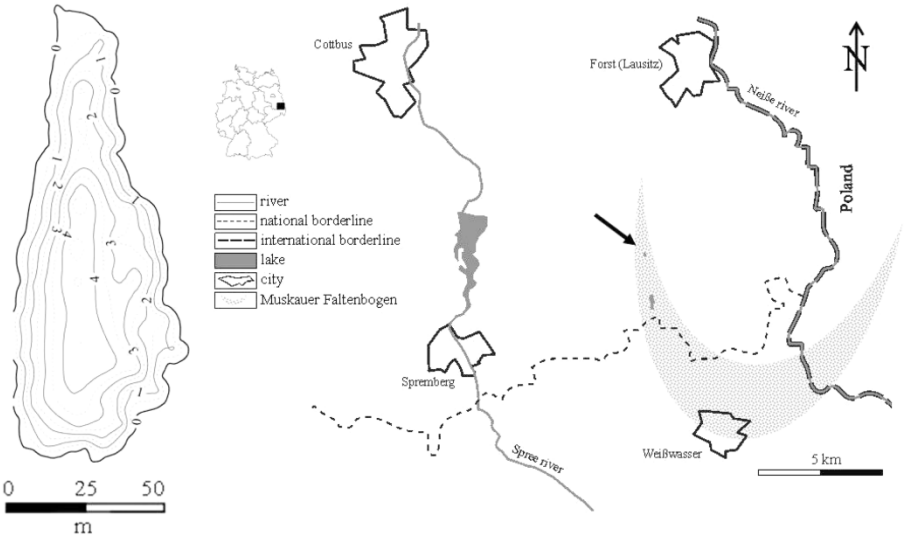

Figure 1. Bathymetry of Lake Waldsee and a generalised map of the study area with its location in Germany.

deformed and tilted glaciotectonically. The exploration of the uplifted lignite layers started during the early 1860s and lasted until the late 1950s [18].

The climate of the study area is characterized by an annual temperature range between −1.3 and 17.9 °C with an average temperature of 8.4 °C [19] and a mean humidity close to 80% [20]. Since no current values of precipitation and evaporation were available, long-term average values were taken from the literature. The annual rainfall averages 627 mm [21], the potential evaporation rate of open surfaces water bodies with an average depth of 4–9 m amounts to 752 mm [22] (table 1). Due to its protected and shaded location, the evaporation rate of Lake Waldsee is likely to be significantly smaller. Lake Waldsee is a meromictic pond with a surface area of approximately 3400 m^2, a volume of 7500 m^3 and a maximum depth of 4.9 m. A tributary does not feed the lake even though the assumed surface catchment area of the ca. 5 m wide slope around the lake is estimated to amount 1558 m^2. At a certain lake water level, a small rill drains the lake. Unfortunately, hydrologic data of the area are limited and information about the elevation of the groundwater table in the vicinity is not available.

3. Methodological background

3.1 *Isotope mass balance*

Based on the general hydrologic water balance [equation (1)]:

$$dv/dt = I_G + I_Z + P - E - O \tag{1}$$

the equation of the isotope water balance [equation (2)] can be written as

$$\frac{V\,d\delta_L + \delta_L dV}{dt} = \delta_P(I_Z + P) + \delta_G I_G - \delta_E E - \delta_O O, \tag{2}$$

where V is the volume of the lake, I_G the groundwater inflow, I_Z the surface inflow from the surrounding slopes, P the precipitation, E the evaporation and O the outflow. The δ terms δ_L,

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Table 1. Monthly meteorological and isotope data used for the calculation of the isotope signature of Lake Waldsee.

Month	Temperature (air, °C) [19]	Precipitation (mm) [21]	Humidity (%) [20]	$\alpha^{18}O$ (T) [24]	αD (T) [24]	$\delta^{18}O_p$ (‰)$_{VSMOW}$ [10]	δD_p (‰)$_{VSMOW}$ [10]	$\varepsilon_k^{18}O$ (‰) (calc.)	$\varepsilon_k D$ (‰) (calc.)	$\delta^{18}O_A$ (‰)$_{VSMOW}$ (calc.)	δD_A (‰)$_{VSMOW}$ (calc.)
January	−1.3	44	86	1.01243	1.10456	−11.27	−73.0	2.0	1.8	−22.39	−174.3
February	−0.2	37	84	1.01226	1.10529	−11.79	−69.8	2.3	2.0	−21.93	−169.0
March	3.3	37	77	1.01176	1.10109	−8.88	−61.7	3.3	2.9	−20.56	−158.0
April	7.7	47	73	1.01119	1.09600	−9.19	−60.8	3.8	3.4	−19.59	−152.5
May	13.0	58	70	1.01057	1.09012	−7.86	−49.3	4.3	3.8	−17.52	−135.6
June	16.4	60	69	1.01021	1.08649	−7.06	−41.7	4.4	3.9	−15.97	−124.6
July	17.9	74	71	1.01006	1.08492	−6.63	−43.7	4.1	3.6	−16.24	−125.2
August	17.2	73	74	1.01013	1.08565	−6.82	−46.6	3.7	3.3	−16.81	−128.8
September	13.5	51	79	1.01051	1.08958	−7.22	−51.5	3.0	2.6	−17.94	−137.3
October	9.0	48	84	1.01103	1.09453	−8.53	−55.2	2.3	2.0	−19.19	−145.5
November	4.1	45	85	1.01165	1.10015	−9.83	−71.9	2.1	1.9	−21.75	−167.3
December	0.4	53	87	1.01218	1.10456	−11.37	−80.8	1.8	1.6	−23.38	−180.2

Temperature, precipitation, humidity, the isotope signature of the precipitation and the equilibrium isotopic fractionation factor (α) were taken from the literature [10, 19–21, 24]. Kinetic enrichment (ε_κ) and the isotopic composition of atmospheric water vapour were calculated referring to [23] and [17], respectively.

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δ_P, δ_G, δ_E and δ_O refer to the isotope composition of the lake water, precipitation, groundwater, evaporate and subsurface outflow relative to Vienna Standard Mean Ocean Water (VSMOW), respectively.

For the first step – the simulation of the lake water's isotope signature – a number of conditions were defined to keep the range of variations in a moderate order.

- The catchment area of the lake is equal in size to its surface area plus a 5-m margin around the lake providing surface runoff at rain events.
- There is neither groundwater inflow nor groundwater outflow.
- The mixolimnion of the lake water is of homogenous isotope composition.
- After reaching the maximum lake level, the effluent of the lake is activated.

Acting on these assumptions, changes in the lake volume and isotope composition will be driven only by precipitation and evaporation. The surface runoff of the slopes are characterised by the same isotope composition like the precipitation, respectively. The calculations start at the initial state of the lake system. During the early period of flooding, it was supposed that the lake underwent polymictic conditions until it reached a defined volume that enabled the development of stratification. Due to this continuous increase in lake volume, the lake's steady state was regulated by surface outflow, which would have had the same isotope signature as the lake water. Following these requirements, equation (2) can be shortened to equation (3)

$$\delta_{(P+I_Z-E)} = \frac{\delta_P(I_Z + P) - \delta_E E}{I_Z + P - E}. \tag{3}$$

And finally equation (4)

$$\delta_L = x\delta_{(I_Z+P-E)} + y\delta_{L(0)}, \tag{4}$$

where $\delta_{L(0)}$ is the preceding oxygen isotope composition of the lake water, and x and y are weighted portions of the temporary lake volume. Except δ_E, all parameters were taken from literature [10, 21, 22] where long-term average meteorological and isotope data sets are quoted on a monthly base (table 1). Unfortunately, no actual data for evaporation rates were available; the value of 752 mm was evaluated between 1951 and 1975 [22]. For modelling the isotope signatures of the lake water, it seemed to be suitable to reduce the real evaporation rate from 752 to 600 mm taking into account that the lake is located mostly shaded within a forest. The annual precipitation height of 627 mm [21] offers uncertainties as well, since the meteorological conditions vary in short distances due to the hilly geomorphology of the study area.

As the isotopic composition of the evaporation flux δ_E could not be determined directly, it had to be estimated according to equation (5) derived from the resistance model of the evaporation process proposed by Craig and Gordon [1]

$$\delta_E = \frac{(\delta_L - h\delta_A - \varepsilon)}{(1 - h + \varepsilon_k/1000)} \tag{5}$$

with the ambient relative humidity h [20] and the total isotopic fractionation factor $\varepsilon = \varepsilon^* + \varepsilon_\kappa$, including both equilibrium ε^* and the additional kinetic ε_κ fractionation during evaporation of open of surface water. The calculations followed equations (6) and (7) [23]

$$\varepsilon_{kbl-v(^{18}O)} = 14.2(1 - h), \tag{6}$$

$$\varepsilon_{kbl-v(^{18}D)} = 12.5(1 - h). \tag{7}$$

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The isotopic content of the atmospheric moisture δ_A was calculated by supposing isotopic equilibrium between local precipitation and atmospheric water vapour following equation (8) [17]

$$\delta_A = \delta_P \left(\frac{1}{\alpha}\right) - \left(1 - \frac{1}{\alpha}\right), \tag{8}$$

with the equilibrium isotope fractionation factor at the temperature of the air–water interface α [24].

3.2 *Measurement of in situ parameter and isotope analysis*

Vertical profiles of temperature and electrical conductance were determined with a multi-parameter sonde (YSI 6820) during four field samplings in 2006 and at the beginning of 2007. The dates of the measurements represented the altering stages of the lake's seasonal circulation.

Water samples for isotope analysis were collected on four occasions during 2006 and 2007. Stable isotope analysis of oxygen-18 and deuterium was carried out using a CO_2 equilibration unit coupled with an IRMS (Finnigan MAT Delta S). The isotope ratios of $^{18}O/^{16}O$ and $D/^{1}H$ are conventionally expressed in delta notations of their relative abundances as deviations in per mil (‰) from the international standard VSMOW. The analytical uncertainty of the $\delta^{18}O$ and δD measurements is ± 0.1 and ± 0.8 ‰, respectively.

4. Results and discussion

4.1 *Field data*

Investigations since 1995 have shown that the position of the chemocline ranges between 0.5 and 1.5 m. Profiles of temperature and electrical conductance (figure 2) indicated that the lake is meromictic. Even though the temperature gradient between the mixolimnion (well-mixed surface water body) and the monimolimnion (deep water body) was minimal during certain periods of the year, the gradient in the electrical conductance due to the high iron content of the deep water maintained stratification. The water body was characterised by an average pH value of 6.3 in the monimolimnion. In 1999, the mean pH in the well-mixed surface layer shifted from 3.2 to 6.5 [25].

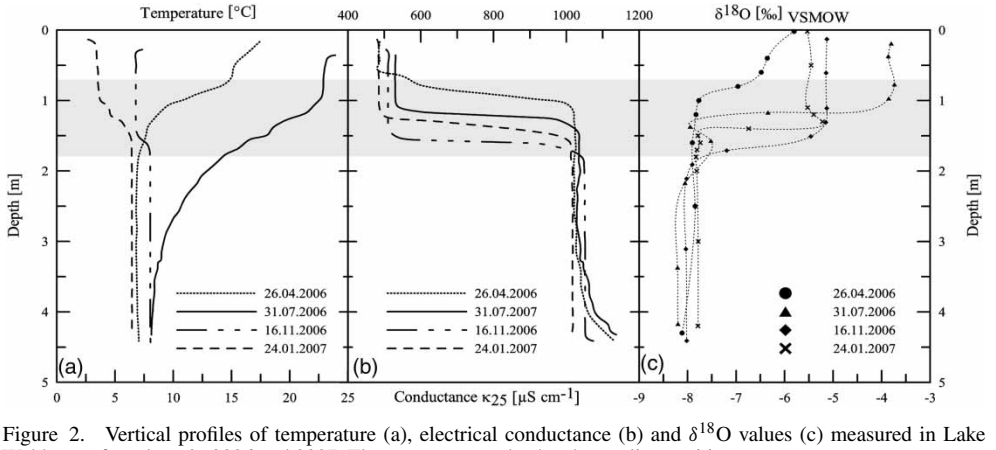

Figure 2. Vertical profiles of temperature (a), electrical conductance (b) and $\delta^{18}O$ values (c) measured in Lake Waldsee at four dates in 2006 and 2007. The grey zone marks the chemocline position.

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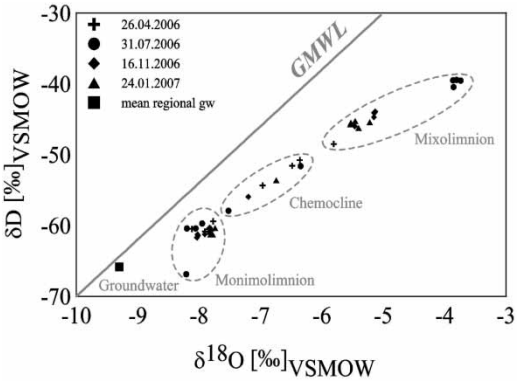

Figure 3. $D/^{18}O$ diagram of isotope composition of obtained lake water samples and the background value of regional mean groundwater signatures.

During 2006 and 2007, four data sets of $\delta^{18}O$ and δD values were obtained from Lake Waldsee. Figure 2 represents the depth profiles of oxygen-18 isotopes. The stable isotope values varied in the mixolimnion between -3.74 and -5.81 ‰ for $\delta^{18}O$ and -39.4 and -48.5 ‰ for δD, respectively. In the monimolimnion, the values varied between -7.52 and -8.21 ‰ and -57.9 and -66.9 ‰ for $\delta^{18}O$ and δD, respectively. Observed mean annual values of the stable isotope composition in regional groundwater aquifers averaged -9.30 ‰ for $\delta^{18}O$ and -65.9 ‰ for δD, respectively [10]. Figure 3 displays a $D/^{18}O$ diagram of the lake water samples and the background value of the regional groundwater aquifer. The mixolimnion water naturally shows the highest influence of evaporation effects and can be considered as the end member in the mixing process between lake water and the meteoric characterised groundwater.

4.2 *Modelling of lake water isotopes*

The simulation of the $\delta^{18}O$ signatures of the lake water was calculated with four different evaporation rates ($E = 1, 100, 300, 600$ mm) on a monthly basis for a period of 10 years, respectively. Figure 4 demonstrates the generated development of the $\delta^{18}O_L$ values during one decade. The mine pit was filled with precipitation water and after attaining a volume of about 3400 m^3, the water body may have established a permanent stratification and formed a monimolimnion with a volume of about 1500 m^3 of size. The separated water in the monimolimnion preserved the $\delta^{18}O_{L(MIX)}$ signature that was marked by the well-mixed water body prior to the permanent stratification. The time needed to stabilise the $\delta^{18}O_{L(MIX)}$ values on a certain level depends on the evaporation rate. The influence of the evaporation rate is also reflected in the range of variations of the calculated $\delta^{18}O_{L(MIX)}$ values and correlates positively with the span of the $\delta^{18}O_{L(MIX)}$ signatures. The higher the evaporation rate the higher the span of the $\delta^{18}O_{L(MIX)}$ variations. Table 2 demonstrates the range of the modelled $\delta^{18}O_{L(MIX)}$ values from the moment of the stratification. The seasonal variations of the surface water isotope signatures are well known from many investigated lakes [5, 6, 26]. These fluctuations are regulated by the meteorological parameters, e.g. precipitation amount and temperature. In each of the simulated cases, the initial $\delta^{18}O_L$ composition is regulated by the $\delta^{18}O_P$ signature of the precipitation and therefore underlies identical oscillations. Depending on the change of the evaporation rates, the $\delta^{18}O_{L(MIX)}$ values show different intensities of the evaporation-effected enrichment of oxygen-18 due to kinetic isotope fractionation. Except the $E = 600$ mm model, the $\delta^{18}O_{L(MON)}$ values become heavier with increasing evaporation rate due to the length of

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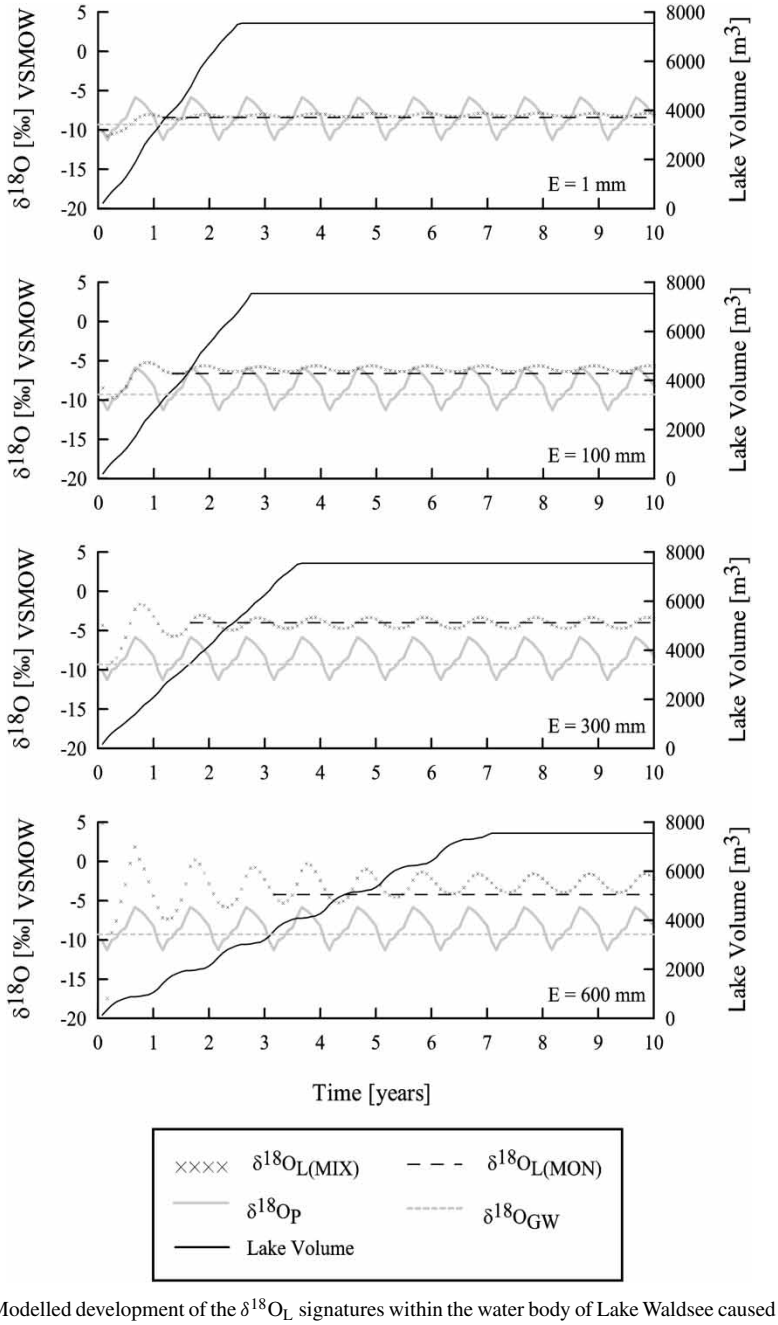

Figure 4. Modelled development of the $\delta^{18}O_L$ signatures within the water body of Lake Waldsee caused by varying evaporation rates. The solid black line shows the increasing lake volume, the crosses, the dashed line and the solid grey line display the oxygen isotope composition of the mixolimnion water $\delta^{18}O_{L(MIX)}$, the monimolimnion water $\delta^{18}O_{L(MON)}$ and the precipitation $\delta^{18}O_P$, respectively.

time that is needed to reach the required volume for developing stratification (table 2). The reason for the relatively light $\delta^{18}O_{L(MON)}$ value of the $E = 600$ mm model is the big span of the $\delta^{18}O_{L(MIX)}$ variations as the isotope signature of the depth water is controlled by the point of time when the monimolimnion was separated from the mixolimnion and therefore depends on the isotopic composition of the mixolimnion water.

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Table 2. Range of the modelled $\delta^{18}O_{L(MIX)}$ values from the moment of the stratification, the isotope composition of the depth water and the point of time of stratification.

Evaporation rate (mm)	Min. $\delta^{18}O_{(MIX)}$ (‰)$_{VSMOW}$	Max. $\delta^{18}O_{(MIX)}$ (‰)$_{VSMOW}$	Range $\delta^{18}O_{(MIX)}$ (‰)$_{VSMOW}$	$\delta^{18}O_{(MON)}$ (‰)$_{VSMOW}$	Time needed for stratification (months)
1.00	−8.58	−7,86	0.72	−8.41	14
100	−6.65	−5.63	1.02	−6.62	16
300	−4.90	−3.04	1.86	−3.98	20
600	−5.31	−0.29	5.02	−4.22	38

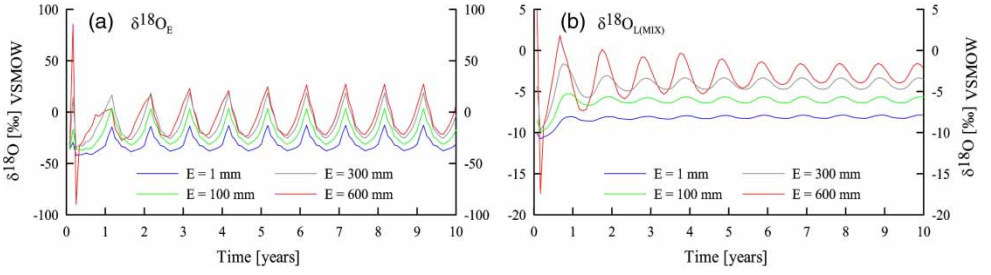

Figure 5. Modelled development of the $\delta^{18}O_E$ signatures of the surface water (a) and the $\delta^{18}O_{L(MIX)}$ signatures within the water body of Lake Waldsee (b) caused by varying evaporation rates.

However, as δ_E cannot be determined directly, this parameter shows the biggest uncertainty in the model. Figure 5 concludes the development of the modelled $\delta^{18}O_E$ and the resulting $\delta^{18}O_{L(MIX)}$ signatures. These strong variations are controlled by equation (5), which is based on the kinetic fractionation factor ε_κ and the humidity h, respectively. The high span of the modelled isotope signatures of the evaporate in the $E = 600$ mm model is due to the higher impact of the evaporation amount in equation (3) than at the low evaporation models and therefore leads to the bigger variation of the surface water's isotope signatures. Craig and Gordon [1] already detected that the humidity and the isotopic composition of the moisture of the 'free air' are responsible for the isotope ratio of the evaporate. The higher the humidity the lower the evaporation fractionation effect, because at humidity that is equal to 100%, only pure isotope exchange between surface water and air moisture exists, and no evaporative flux from the lake surface to the free atmosphere occurs. Increasing humidity induces a decreasing ε_κ, thus the resulting δ_E signature becomes lower than at low humidity. The explanation for this statement is that under conditions of high humidity diffusion between the mixed atmosphere and the water surface is equal, and no net diffusive fractionation emerges. Furthermore, at low humidity the water surface will be more enriched in the heavy isotopes.

Figure 6 shows the evaporation lines of the modelled isotope signatures for the mixolimnion water and the isotope signatures of the depth water considering different evaporation rates. The image displays that only the monimolimnion values of the $E = 100$ mm model fit approximately with the measured data. Even if the evaporation line of the $E = 600$ mm model fits with the real surface waters, the calculated isotope composition of the depth water is too heavy in comparison with real terms.

Looking back to figure 2, all four data sets indicate a chemocline between 0.8 and 1.5 m below the water surface. The $\delta_{L(MIX)}$ values in the surface water layer are enormously affected by evaporation in contrast to the isotope signatures of the monimolimnion water. The simulated $\delta_{L(MIX)}$ signatures in the $E = 600$ mm model showed values between -5.00 and -1.00 ‰ for oxygen-18 and are consequently heavier than in real terms. The total quantity of simulated

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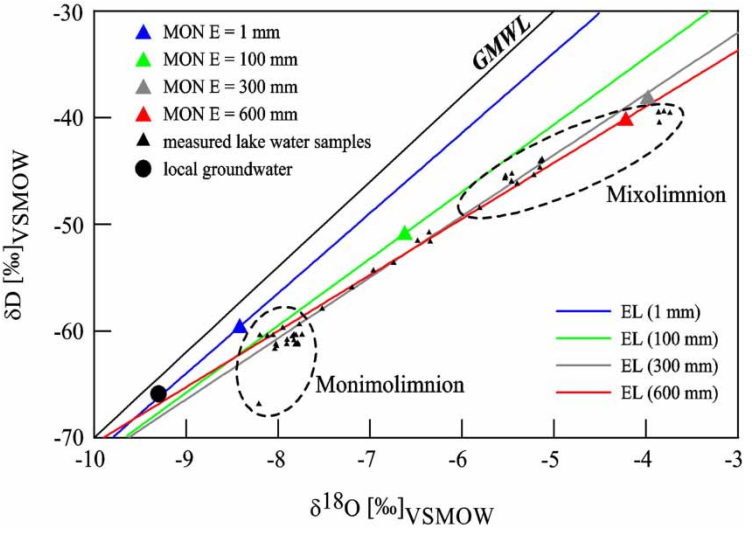

Figure 6. $D/^{18}O$ diagram of the measured lake water samples: the modelled evaporation lines for the mixolimnion water and the isotope signatures of the modelled monimolimnion water for different evaporation rates, respectively.

inflow, which is composed of the precipitation over the lake's surface area and the pit-wall runoff, affects the isotope signature of the mixolimnion water towards an enrichment of the lighter isotopes. On the other hand, evaporation contrasts this progress in the direction of enrichment of the heavier isotopes. Nevertheless, both, precipitation and evaporation only control the mixolimnion water, while the modelled isotope composition of the monimolimnion water was set at the time of stratification and shows a $\delta^{18}O$ value of -4.22 ‰. The measured mean $\delta_{L(MON)}$ values (-7.90 ‰ for ^{18}O and -60.9 ‰ for D, respectively) of the monimolimnion water and the observed mean groundwater values (-9.30 ‰ for ^{18}O and -65.9 ‰ for D, respectively) are much lighter in isotope composition and could be a link to the dilution of the lake water by groundwater inflow. Taking into account that one of the frame conditions for the modelling was zero groundwater inflow, the comparison between the modelled and the measured isotope data of the monimolimnion water shows that in real terms the lake must be fed by groundwater inflow.

5. Conclusions

The modelling of isotope signatures proves to be a useful tool to assess the interactions between a lake water body and the surrounding groundwater. The simulated $\delta^{18}O_L$ values of meromictic Lake Waldsee showed the differences between the deep-water layer (monimolimnion) and the well-mixed surface water layer (mixolimnion). As expected, the surface water was characterised by an evaporative enrichment of the oxygen-18 in contrast to the isotopic more lightly monimolimnion water. The measured values for oxygen-18 of the depth water (figure 2) showed a much lighter isotope composition (minimum -8.21 ‰, maximum -7.52 ‰) than the modelled isotope signatures of the monimolimnion water (~ -4.22 ‰) in the $E = 600$ mm model (figure 4). A comparison of the modelled evaporation lines, the calculated isotope values of the monimolimnion water and the measured lake water samples, shows that the slope of the $E = 600$ mm model fits the real evaporation line even if the monimolimnion value of the $E = 1$ mm model corresponds to the measured data. Taking into account the frame conditions,

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which were set for the modelling, this comparison shows that the deep water of Lake Waldsee must be diluted by groundwater inflow.

6. Future studies

Unfortunately, the Craig and Gordon model [1] displays uncertainties for determining δ_E in regions with seasonally high humidity, because in these cases the evaporative fractionation effect is interfering with diffusive isotope exchange between the lake water surface and the free atmosphere. From this point of view, further investigations are needed [27]. For this reason, the model should be adapted to the conditions of temperate climates. For ongoing studies, several groundwater wells were installed around Lake Waldsee and a weather station was set up at the study site. In addition, an in-lake evaporation pan of 200 L volume was placed into the lake. The investigation activities also include a method that allows to sample monthly average samples of the evaporate for the measurement of oxygen-18 and deuterium.

Acknowledgements

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Dating problems with selected mining lakes and the adjacent groundwater body in Lusatia, Germany

A. Seebach[a]*, C. von Rohden[b], J. Ilmberger[b], S.M. Weise[a] and K. Knöller[a]

[a]*Department of Isotope Hydrology, Helmholtz Centre for Environmental Research – UFZ, Halle (Saale), 06120 Germany;* [b]*Institute of Environmental Physics, University of Heidelberg, Heidelberg, 69120 Germany*

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This study presents selected results, applying environmental tracers to investigate lake water–groundwater interactions at two study sites located in Lusatia, Germany. The focus of the investigations were two meromictic pit lakes and their adjacent aquifers. In order to follow hydrodynamic processes between lake and groundwater, mixing patterns within the lakes as well as ages of lake and groundwater, water samples of ground- and lake water were collected at three occasions, representing summer and winter conditions in the aquatic systems. The water samples were analysed for stable isotopes (deuterium, oxygen-18) and tritium and sulphurhexafluoride (SF_6 concentration). Lake water profiles of conductivity and ^{18}O could validate the permanent stratification pattern of both the lakes. Groundwater data sets showed a heterogeneous local distribution in stable isotope values between rain and lake water. A two-component mixing model had been adopted only from ^{18}O data to determine lake water proportions in the surrounding groundwater wells in order to correct measured tritium and SF_6 concentrations in groundwater samples. This procedure had been hampered by upstream-located wells indicating strong ^{18}O enrichment in groundwater samples. However, rough groundwater ages were estimated. For both study sites, Piston flow ages between 12.9 and 27.7 years were calculated. The investigations showed the good agreement between two different environmental dating tools, considering the marginal data sets.

Keywords: deuterium; groundwater dating; oxygen-18; stable isotope tracer techniques; sulphurhexafluoride; tritium

1. Introduction

Widespread surface lignite mining activities in Lusatia, Germany caused dramatic changes to the aquatic systems. Related to pyrite oxidation occurring in the aerated dump sediments, ground- and surface water bodies underlie continuous acidification. Thus, these waters are characterised by high concentrations of dissolved ions – e.g. sulphur and iron – and therefore density gradients can induce specific stratification patterns in lakes. In this context, different investigations concerning hydrogeochemistry, lake water budgets, and tracer hydrology were performed at two selected mining lakes. For the assessment of groundwater flow directions as well as groundwater–surface

*Corresponding author. Email: anneseebach@gmx.net

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water interactions, the patterns of the stable environmental isotopes oxygen-18 and deuterium were investigated. In addition, both tritium analysis of lake and groundwater and SF_6 measurement of groundwater samples were conducted to classify lake and groundwater ages. The scope of this article is to present a qualitative contribution derived from stable isotopes about the effects of lake water admixtures for dating of ground- and lake water with tritium and SF_6.

2. The study site

The area of interest is located approx. 130 km south of Berlin (Figure 1). Both investigated lakes (Lake Moritzteich and Lake Waldsee) are embedded in an ice-trusted ridge of a moraine formed by the Middle Pleistocene Elsterian glaciation. The high pressure of the glacier generated the deformation and uplift of Tertiary lignite, clay, silt, and sand layers [1]. Therefore, an intricate small-scale structured hydrogeology is present in the study area. In the study region, the groundwater flow direction is – as far as known – orientated from south to north [2]. Besides the two study lakes, there are many other small lakes in this region, resulting from lignite mining activities.

Lake Moritzteich has a surface area of 16.5 ha and a maximum depth of ≈17.4 m; a small ditch with seasonal flow conditions feeds Lake Moritzteich during the winter half of the year. On the contrary, Lake Waldsee shows a surface area of ≈3300 m^2 and a maximum depth of ≈5.0 m. There is no surface inflow into Lake Waldsee. After mine drainage was stopped in the 1960s, the lakes reached their definite volume in the late 1970s. Because of the particular hydrogeochemistry in this area, both study lakes show meromictic stratification. Due to density gradients resulting from high content of dissolved ions, the water column is separated into two horizontal layers, the well-mixed surface water with seasonal overturn (mixolimnion), and the deep-water body (monimolimnion), which is disconnected from atmospheric impacts [3]. The transition zone between the aerobic surface water and the anoxic deep-water is called chemocline. The height of the chemocline region in both lakes changes within a seasonal cycle. During the investigation period, seasonal lake level changes were observed at both study sites. At Lake Waldsee, the difference between summer and winter surface water level was about 20 cm; for Lake Moritzteich, the lake level changes amounted to 40 cm.

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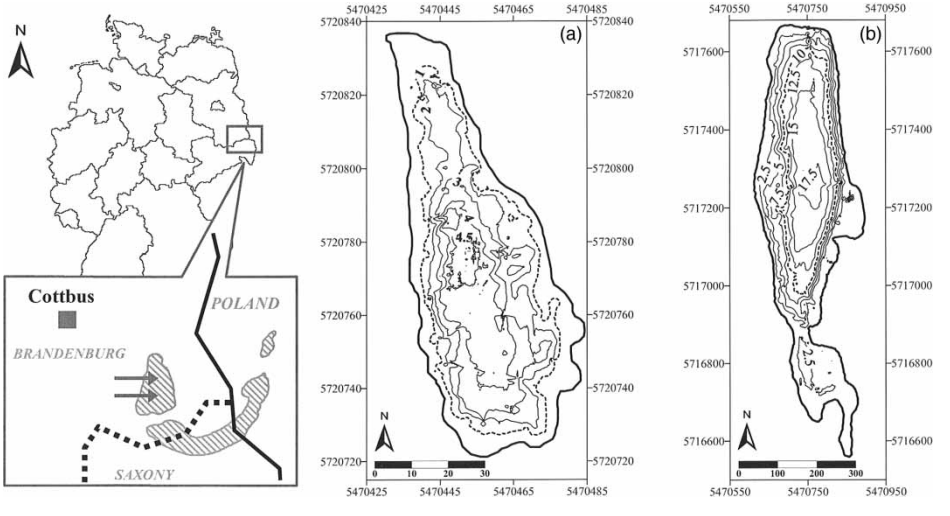

Figure 1. Location of the study site and bathymetric maps of Lake Waldsee (a) and Lake Moritzteich (b).

Table 1. Sampling schedule for the investigated areas.

Sample		Summer 2006	Winter 2006	Summer 2007
Waldsee	Lake water	$D/^{18}O/T$	$D/^{18}O/T$	
	Groundwater		$D/^{18}O/T$	$D/^{18}O/SF_6$
Moritzteich	Lake water	$D/^{18}O/T$		
	Groundwater		$D/^{18}O/T$	$D/^{18}O/SF_6$

3. Fieldwork and laboratory measurements

At two field trips in 2006, samples were taken from both lakes. Lake water sampling was carried out at the deepest point of the lakes, respectively. A specially designed submerged pump admitted the precise water sampling from a certain depth. The resolution of the lake water profiles for stable isotope analysis was about 10 cm in the chemocline region of the lakes in order to capture the exchange between the mixolimnion and the monimolimnion of the respective lakes. Additionally, electrical conductivity (κ_{25}) profiles of both lakes were taken with an automatic probe (Idronaut). Groundwater sampling was executed with a peristaltic pump in winter 2006 and summer 2007. During clear pumping of the groundwater wells surrounding each lake, field parameters (pH, κ_{25}) were measured continuously until steady-state setting was reached. By maintaining constant pressure while sampling for SF_6, stripping effects of gases could be minimised. The quantity of the water samples was 25 ml for stable isotope analysis, 1 l for tritium and 0.6–2.0 l for SF_6 measurement, respectively (Table 1).

The stable isotope ratios δD and δ^{18}O were determined with an equilibration unit coupled with a ThermoFinnigan Delta S isotope ratio mass spectrometer. Isotope ratios are conventionally expressed against the scale of the Vienna Standard Mean Ocean Water. The uncertainty for δ^{18}O and δD are ± 0.1 and ± 0.8 $\delta -$ ‰, respectively. After electrolytic enrichment, tritium analysis was performed by the liquid scintillation method (Perkin Elmer Quantulus 1220). The values are given in Tritium Units (TU) with one TU equalling one tritium atom in 10^{18} hydrogen atoms. The uncertainty for tritium measurement is $\pm 10\%$ with a detection limit of 0.3 TU. SF_6 sampling was carried out in special nitrogen prepared glass bottles. SF_6 analysis was executed by a headspace GC-ECD technique with reproducibility better than 1%. The procedure is described by von Rohden *et al.* [4].

4. Results and discussion

Electric conductivity profiles of both lakes (Figure 2) showed characteristic stratification pattern of the lake water column throughout the observation period. The winter profiles show stratified water columns in both lakes, due to enduring gradients in conductivity, which is the requirement for meromictic lakes even if the lakes were not covered by an ice sheet. For this reason, the permanent stratification of both lakes was confirmed. The chemocline in Lake Waldsee showed a seasonal variation of its depth from $\approx$0.8 to $\approx$1.6 m. In the surface water, κ_{25} values of 500 μS cm^{-1} contrasted values of 1000 μS cm^{-1} of the deep-water body. The chemocline position in Lake Moritzteich varied between 10.0 and 10.5 m and was caused by a difference in conductivity from 1000 μS cm^{-1} in the mixolimnion up to 1500 μS cm^{-1} in the monimolimnion. The second step of increasing conductivity between 13.5 and $\approx$ 16.0 m could be referred to groundwater inflow into Lake Moritzteich [5]. The variations of the chemocline had been discussed in [3,5].

The stable isotope profiles of both lakes (Figure 2) showed a significant gradient in isotope signatures occurring in the chemocline zone. Evaporative enrichment of the heavier oxygen isotopes

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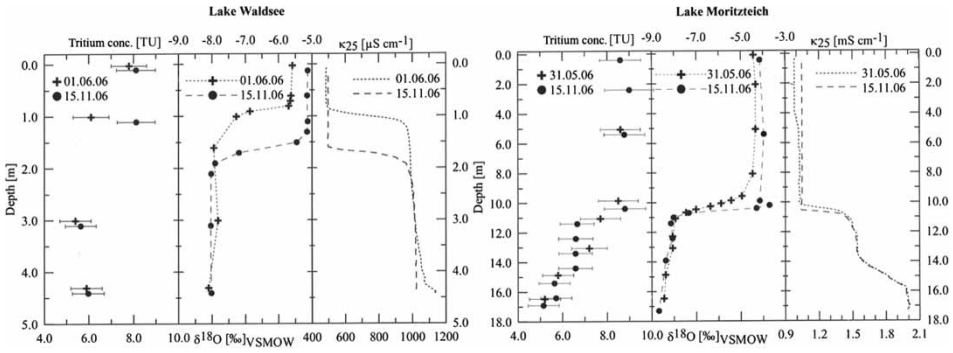

Figure 2. Lake water profiles of Lake Waldsee (left) and Lake Moritzteich (right). The chart shows the tritium activity, the δ^{18}O and κ_{25} profiles of each lake at two different points of time.

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in the surface water with values up to −3.72 ‰ in Lake Moritzteich contrasted isotopically with lighter water in the monimolimnion, which was separated from evaporation effects by the chemocline and which, in both lakes, was known as dominated by groundwater inflow. Deep-water samples provided δ^{18}O values between −6.36 and −8.67 ‰ for Lake Moritzteich and −6.86 to −8.10 ‰ for Lake Waldsee. Recent isotope signatures of regional aquifers average −9.30 and −65.9 ‰ for δ^{18}O and δD, respectively [6].

Concerning the measured tritium activities of the lake waters, there were certain differences between Lake Moritzteich and Lake Waldsee. The tritium profile of Lake Moritzteich was characterised by decreasing tritium content towards the lake bottom. The surface water body indicated tritium activities between 8.5 and 9.0 TU in both the measured data sets. Below the chemocline, the tritium content dropped down to values between 5.1 and 7.7 TU. On the first sight, Lake Waldsee did not demonstrate the separation between mixolimnion and monimolimnion in the tritium profiles as well as Lake Moritzteich. Nevertheless, on closer inspection, it was obvious that the tritium profiles were also separated into mixolimnion and monimolimnion. Due to a downward drift of the mixo-/monimolimnion transition from summer to winter time, in the June data set, the second tritium sample from above belonged to the deep-water and to the surface water in the winter samples, respectively. The surface values of tritium concentration averaged 8.0 TU and the tritium samples of the monimolimnion varied close to 5.8 TU.

Groundwater samples that were collected around Lake Moritzteich (MMT) and Lake Waldsee (MWS) scattered between −6.42 and −9.59 ‰ for δ^{18}O. Plotting stable isotope data of groundwater and lake water samples in a δD/δ^{18}O diagram (Figure 3) is a simple way to get an idea about lake water distribution in the aquifer and hence allows determination of the lake water proportion in the monitoring wells. Therefore, it was possible to roughly estimate groundwater flow directions by means of stable isotope signatures. Generally, wells with a high amount of lake water are not expected to be located within the upstream flow of the lake. The winter situation in Figure 3 shows that at Lake Waldsee, each well except MWS 3 was located on a mixing line between evaporation-affected surface water of the lake and the endmember ‘pure’ groundwater (recent isotope signatures of regional aquifers: −9.30 ‰ for δ^{18}O, −65.9 ‰ for δD). The summer situation offers a different pattern of isotope distribution in the monitoring wells. Only the water of well MWS 4 showed a significant evaporation effect. The isotope signatures of MWS 4 indicated values of −6.46 ‰ and −51.4 ‰ for δ^{18}O and δD, respectively. This can be explained by the fact that groundwater flow direction in the catchment area of Lake Waldsee is supposed to be oriented from south to north. Since there are many other small mining lakes located near Lake Waldsee, it can be assumed that well MWS 4 is dominated by groundwater, which has already undergone evaporation within a ‘precursor lake’. Considering preferred flow paths in the former

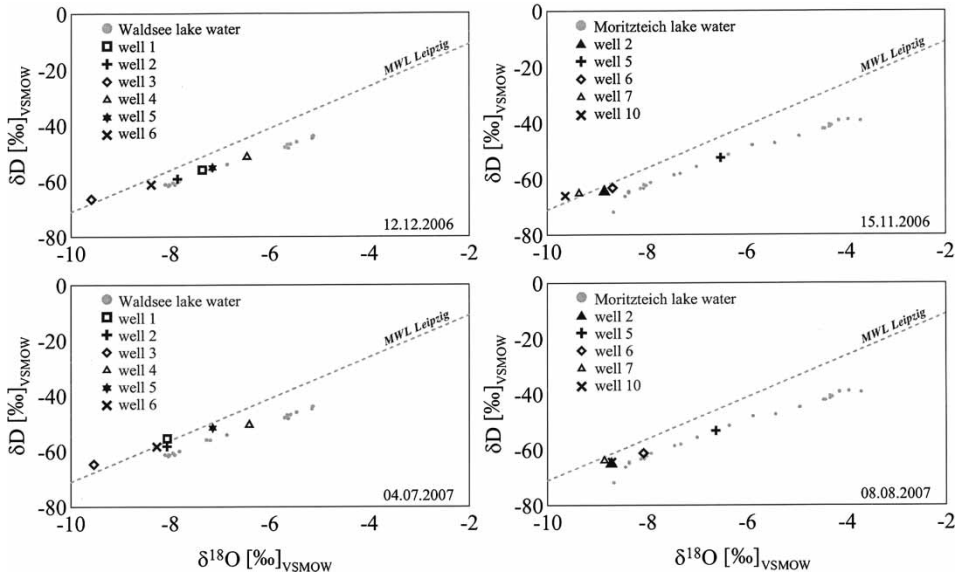

Figure 3. $\delta D/\delta^{18}O$ diagrams of water samples obtained at Lake Waldsee and Lake Moritzteich to detect evaporative enrichment in groundwater wells and lake water–groundwater interactions, respectively.

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lignite mining area, it is supposable that Lake Waldsee was recharged by subsurface inflow that was considerably enriched in heavy isotopes due to prior evaporation effects. Lake Moritzteich showed a distribution pattern in stable isotope signatures of groundwater similar to that of Lake Waldsee. The hydrodynamic regime is subject to seasonal changes, and only well MMT 5 showed a constant quantity of lake water infiltration. One cannot exclude groundwater recharge with water, which was already enriched in heavy isotopes. The results show that stable isotopes in order to track groundwater flow directions as an independent tracer cannot be applied for every study site.

The groundwater samples from the banks of both lakes showed large differences in tritium concentration (Figure 4). The groundwater wells on the northern shore of Lake Moritzteich (MMT 5 and 6) exhibited tritium values of 17.4 and 19.1 TU, respectively, and on the eastern shore 1.3 TU (MMT 2). The data, which could be obtained from groundwaters around Lake Waldsee, spread to a similar extent: between 5.4 TU at the western margin (MWS 6) and 17.7 TU southeast of the lake (MWS 3). Stable isotope data of the groundwater wells (Figure 3) in a $\delta D/\delta^{18}O$ diagram could show that several wells at both lakes were influenced by lake water infiltration.

In order to correct the measured tritium data from the lake water influenced wells to the original tritium content of the groundwater endmember, it was necessary to determine lake water proportions (X_{lake}) in the water of the groundwater wells. Based on a linear two-component mixing model [7] for the analysed groundwaters (Equation (1)) between an estimated pure groundwater endmember (δ_{gw}) and the lake water (δ_{lake}), Equation (2) allows the straightforward calculation of X_{lake}:

$$\delta_{\text{well}} = X_{\text{lake}}\delta_{\text{lake}} + X_{\text{gw}}\delta_{\text{gw}} \quad \text{with} \quad X_{\text{lake}} + X_{\text{gw}} = 1 \tag{1}$$

$$X_{\text{lake}}(\%) = 100\frac{\delta_{\text{well}} - \delta_{\text{gw}}}{\delta_{\text{lake}} - \delta_{\text{gw}}} \tag{2}$$

with X describing the amount of groundwater in the well (well), the lake water (lake) and the groundwater endmember (gw) as well as the respective isotope signatures (δ). Since seasonal data

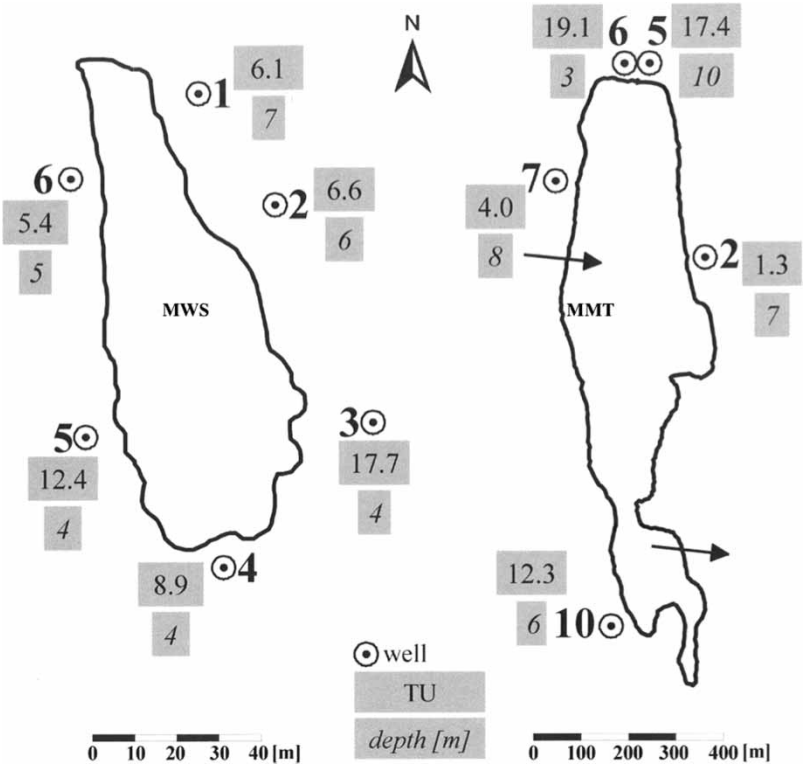

Figure 4. Location of the groundwater wells, their depth and the measured tritium concentration in the groundwater samples at Lake Waldsee (left) and Lake Moritzteich (right), respectively. The arrows indicate the surface in- and outflow of Lake Moritzteich.

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in stable isotopes of groundwater samples may show more negative results than mean average values, the background values (δ_{gw}) for both lakes were set to be the lightest δ_{well} value that was measured during both field campaigns (−9.59% for Lake Waldsee and −9.64% for Lake Moritzteich). Corrected tritium concentrations of the non-lake groundwater component were obtained analogously according to Equation (3):

$$T_{gw} = \frac{T_{well} - X_{lake} T_{lake}}{X_{gw}}. \quad (3)$$

As indicated by stable isotopes, groundwater in well MMT 5 at the northern shore of Lake Moritzteich had a lake water proportion of more than 60 %. Therefore, the extrapolated tritium content of the respective aquifer rose up to 30.9 TU compared with 17.4 TU measured in the sample. However, well MMT 2 should be supposed to be a mixture of tritium-free groundwater and lake surface water as well as MMT 7 whose corrected tritium data were little lower than the measured values. At Lake Waldsee, there are two wells, which showed significant changes in tritium activity after taking into account the mixing between lake and groundwater. Groundwater well MWS 5 showed a corrected value of nearly 40 TU, which could be attributed to the high lake water fraction in the well. The calculated negative tritium value of well MWS 4 (−55.8 TU) might be explained by the fact that the lake water proportion of 100 % did not have to be derived implicitly from Lake Waldsee as mentioned above. However, if the water in well MWS 4 is supposed to be lake water from Lake Waldsee or the 'precursor' lake, it is consequential that it was in equlilibrium with the atmosphere and therefore offers tritium concentrations of ∼9 TU.

Table 2. Lake water proportions (X_{lake}) for winter 2006, tritium concentrations (measured and reconstructed), SF_6 concentrations and approx. SF_6 age of groundwater samples.

Well	X_{lake} (%)	$TU_{measured}$	$TU_{corrected}$	SF_6 conc. (fmol/l)	SF_6 age (a)
Lake Waldsee					
MWS 1	74	6.1 ± 0.6	6.9	0.99 ± 0.01	15.9
MWS 2	57	6.6 ± 0.7	6.2	1.06 ± 0.01	14.9
MWS 3	0	17.7 ± 2.2	17.7	–	–
MWS 4	100	8.9 ± 0.9	*−55.8*	2.07 ± 0.02	3.7
MWS 5	80	12.4 ± 1.1	39.4	–	–
MWS 6	40	5.4 ± 0.6	5.1	0.85 ± 0.01	23.5
Lake Moritzteich					
MMT 2	15	1.3 ± 0.5	0.0	0.84 ± 0.01	17.9
MMT 5	61	17.4 ± 1.9	30.9	0.99 ± 0.01	15.9
MMT 6	19	19.1 ± 1.7	21.5	1.24 ± 0.01	12.9
MMT 7	5	4.0 ± 0.6	3.8	0.67 ± 0.01	20.3
MMT 10	0	12.3 ± 1.2	12.3	0.30 ± 0.01	27.7

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The tritium data of the groundwater wells simply allow the qualitative determination of mean residence times, which might be sufficient to get an idea about approximate groundwater ages in the study area [8]. Applying simple lumped-parameter models to these admixture-corrected tritium contents of the groundwaters from lake margins, we obtained residence times between 10 and 30 years except well MMT 2 and well MWS 4. Waters of MMT 2 were expected to have been recharged prior to 1952. For well MWS 4, it was not reasonable to correct the tritium data for lake water infiltration; hence, it was assumed that it was modern water originating from the upstream mining lake (Table 2).

SF_6 analysis was conducted in summer 2007 for all groundwater wells except MWS 3 and MWS 5. Due to poor capacity, both these wells were not adequate for SF_6 sampling. 'Piston flow' groundwater ages between 12.9 and 27.7 years could be calculated for every groundwater well except well MWS 4, which offered a residence time of 3.7 years. With the exception of MMT 2, the SF_6 ages supported the rough estimate of the tritium-based mean residence times, although SF_6 ages were not corrected to lake water infiltration. Nevertheless, even if a certain caution is required in handling SF_6 data of 'young' waters because of unknown excess air and recharge temperature [9], this data could be considered valid because of the local conditions as mentioned above. The data of MMT 2 did not show a good agreement between SF_6 and tritium values. A similar situation was described for the Bogucice Sands aquifer in southern Poland [10]. Apart from that, this disagreement could be explained by the lake water amount of 15 % in well MMT 2, which was bearing a SF_6 signal and concentrates the SF_6-free background of the groundwater. Furthermore, it could not be ruled out that neither some geogenic SF_6 is present in the study area nor the samples were contaminated during sampling or measurement procedure.

5. Conclusions

In summary, the tritium data of waters of both lakes were evident to represent modern water from precipitation and recent groundwater recharge. The monimolimnion of each lake showed a mixture of young as well as tritium-poor groundwater originating from times prior to 1952.

Distribution patterns of lake water infiltration in the monitoring wells at both lakes did not allow the determination of a particular groundwater flow direction at each site except for the southern part of the Waldsee catchment. The results of SF_6 and tritium analyses generally showed a good agreement. The spatial distribution and the large variations in lake water-corrected tritium data

confirmed the complicated hydrogeological settings in the study area. The attempt to reconstruct tritium concentrations of non-lake water-affected groundwater in the wells offers a large variation between piston flow ages. The water in the wells could originate from ancient times, the bomb-peak phase, as well as from very modern waters. As the geology and therefore the hydrogeology is very intricate, one cannot act on the assumption that a homogeneous aquifer is present in the study area. It was supposed that seasonal shifting in the hydrodynamics of the subsurface lake catchments forced changes in lake water–groundwater interactions (Figure 3), and therefore a continuous reshuffling of several water components in the monitoring wells: tritium-free groundwater, modern recharge as well as lake water, which was an admixture of different waters anyway.

However, the summation of different tracing tools, sparse data and an unmanageable study site may limit the reliable application of hydrological attempts but may offer a rough assessment of local conditions.

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Water Balance Studies in Remote Areas – Limits and Possibilities in Evaluating Groundwater – Lake Water Interactions by Stable Isotopes

Anne Seebach[a*], Kay Knöller[a]

[a]*Department of Isotope Hydrology, Helmholtz Centre for Environmental Research – UFZ, D-06120 Halle, Germany.*

Summary

The application of stable isotopes is a common tool for evaluating water balances of lakes. In this paper, we present a two-year isotope data set from three study sites in Lusatia, Eastern Germany, where pit lakes have been explored: Mine lake 107 (ML107), lake Waldsee and lake Moritzteich. ML107 is part of a pit lake area in the Lusatian Lignite District near Plessa. The latter two lakes are located in the *Muskau Arch Fold*, which is an ice-thrust ridge of a moraine formed by the Elsterian glaciation. Aim of the investigations was to quantify monthly groundwater recharge into pit lakes. Two of the three investigated lakes are meromictic, which makes it essential to adapt existing water balance models to permanently stratified lakes. For isotope analyses, high-resolution data sets of lake water, precipitation, and groundwater were collected in addition to comprehensive meteorological records of the three study sites. Evaporation pan experiments as well as the design of a special procedure for collecting monthly average samples of lake evaporate complete the survey of the isotope water balance concerning parameters. Unfortunately, unexpected problems arose during processing the work programme. Namely, the isotope signatures of the local groundwater bodies are not uniform in none of the investigated sites. Moreover, seasonal lake water level fluctuations accompany regular changes in the hydrodynamic system; surface water infiltration into the groundwater aquifer alternates with groundwater exfiltration into the lake water body. The results of this study show that the hydro-/geological setting at the study sites in the *Muskau Arch Fold* is far too complex for the application of well-established water balance models in terms of quantification of groundwater recharge. Furthermore, evaporation pan experiments carried out in the framework of these investigations do not bring any satisfactory results under local meteorological conditions. Regardless of problems mentioned above, the present work introduces a nearly complete high-resolution isotope data set from three mine lakes in the Lusatian Lignite District. So the first local meteoric water and evaporation line were generated for the study areas. The novel technique for sampling lake evaporate allowed obtaining more than thirty water samples which provided (discontinuous) records of isotope signatures of lake evaporate δ_E. For all evaluated components of the hydrological balance (precipitation, groundwater, lake water, lake evaporate) seasonal pattern were observed. Such as, the amplitude height of the temporal variations in surface lake water isotope composition was between 2.43 ‰ (Moritzteich) and 4.43 ‰ (ML107) for oxygen-18, respectively.

Keywords: Oxygen-18; Deuterium; Isotope Water Balance; Evaporation; Water Vapour; Pit Lake; Stratification of Lakes

1. Introduction

The stable isotopes of the water molecule have been applied extensively as environmental tracers in several hydrological studies. Oxygen-18 (^{18}O) as well as deuterium (^{2}H, D) are distinguished tools to follow the hydrological cycle at different scales (Gibson et al., 2005). Stable isotopes are advantageous tracers since they occur naturally in the environment; they show a conservative behaviour and chemical

*Corresponding author. anneseebach@gmx.net

44 inertness. Their relative and absolute abundances in the hydrosphere are mainly
45 controlled by evaporation and condensation (Welhan and Fritz, 1975), whereas
46 fractionation effects vary for oxygen-18 and for deuterium, respectively. Therefore,
47 water molecules undergoing phase changing processes are labelled by a
48 characteristic fingerprint, which allows tracing them for hydrological purposes. More
49 precisely, two independent fractionation processes drive the separation between light
50 and heavy isotope species. In the first place, temperature dependent equilibrium
51 fractionation plays an important role related to the global water cycle. In $\delta^{18}O$ versus
52 δD space, the relationship between oxygen-18 and deuterium in meteoric waters
53 (precipitation, atmospheric water vapour) is defined by Craig's Global Meteoric Water
54 Line (GMWL) (Craig, 1961):
55 $$\delta D[‰] = 8\delta^{18}O + 10 \quad (1)$$
56 On local or regional scale, Local Meteoric Water Lines (LMWL) may be found
57 depending on respective build-up conditions of water vapour and rainout (Araguás-
58 Araguás et al., 2000; Gat et al., 2001). Secondary, additional kinetic fractionation,
59 which is linked to evaporation processes, steps to the fore. Due to differing kinetic
60 fractionation factors for oxygen-18 and deuterium, respectively, evaporation affected
61 waters deviate from the meteoric water line along a line with a lower slope than that
62 of the MWL. The slope of the so called evaporation line (EL) mainly depends on the
63 local relative humidity and commonly varies between 4 and 6 (Clark and Fritz, 1997).
64 The intersection point of the local EL with the LMWL allows an estimate of the
65 weighted mean isotope signature of annual precipitation in the catchment (Gibson et
66 al., 2005). Thus, evaporative enrichment of heavy isotope species enables the simple
67 and quick classification between meteoric waters and evaporation affected waters
68 like e.g. lake waters.

Since the late 60s of the 20th century, numerous authors published many different studies with different beginnings and questions related to isotope water balance approaches. Dinçer used the isotope water balance technique to calculate groundwater inflow rates of two lakes in subhumid south-western Turkey (Dinçer, 1968). Zimmermann and Ehalt though attempted the approach to gain information about short-term evaporation rates at lake Neusiedl (Zimmermann and Ehalt, 1970). Gat however attempted this procedure to estimate kinetic fractionation factors for water stable isotopes undergoing evaporation supported by a given water balance of lake Tiberias, Israel (Gat, 1970). Zimmermann stood in the forefront of applying water balance calculations to artificial lakes which haven't reached steady state conditions (Zimmermann, 1978). Contrary to other authors, whose calculations based on oxygen-18, Zimmermann used the deuterium content of two quarry ponds to estimate groundwater inflow rates. Zuber reviewed a number of published data to derive empirical kinetic enrichment factors for deuterium by fitting calculated evaporation lines to the observed isotope content of several lake waters (Zuber, 1983). Aly et al. followed the Gat's studies and transferred the evaporation pan approach to the Aswan High Dam Lake in order to derive evaporation height as well as empirical relationships between isotopic enrichment and the water lost due to evaporation (Aly et al., 1993). For determining inflow/evaporation ratios in the Northwest Territories, Canada, Gibson et al. investigated two contrasting watersheds by means of evaporation pan experiments and detailed hydrologic studies (Gibson et al., 1993). During the following years, many other geoscientists tried to refine the water balance approach towards evaporation studies like Saxena, who worked with constant volume evaporation pans to evaluate molecular diffusion parameters (Saxena, 1996) or Gibson et al., who used different methods to validate isotope based evaporation rates (Gibson et al., 1996a; Gibson et al., 1996b). For evaluating the chemical mass

budget of two dredged lakes in temperate climatic Austria, Yehdegho and Probst revealed an isotope study in order to approximate the impact of the dredged lakes on the adjacent groundwater field (Yehdegho and Probst, 2001). The same approach was followed by Knöller and Strauch at a former mine site in Germany to assess yearly sulphate load into an acid pit lake (ML111) (Knöller and Strauch, 2002). Many other authors (Gibson et al., 1998), (Yehdegho et al., 1997), (Gibson et al., 1999), (Özaydin et al., 2001), (Sacks, 2002) (Gibson and Edwards, 2002), (Edwards et al., 2002), and (Vallet-Coulomb et al., 2006) worked with the isotope water balance models to investigate analogical problems.

More than 40 years after first attempts of calculating lake water budgets by means of stable isotopes, this paper presents a new study from Lusatia, Germany. Aim of the recent study was the transfer of established isotope balance equations to permanently stratified lakes in order to determine groundwater inflow rates. Additionally, results from the comprehensive investigations at different study sites were expected to bring forward the applicability of the Craig and Gordon term for determining isotope signatures of lake evaporate in central European climate.

Background of these investigations are environmental problems related to former extensive opencast lignite mining activities in Lusatia, Eastern Germany. Acidic mine drainage (AMD) is a common feature at post-mining tailing sites (Grünewald, 2001). Due to pyrite oxidation, aerated dump sediments release high quantities of ferrite and sulphate, which is charged into the lakes by groundwater inflow (Blodau, 2005). This process leads to an unremitting input of acidity into the lakes often resulting in pH values lower than 3.5 (Nixdorf et al., 1998). The total final volume of flooded open-cast mines in Lower Lusatia for instance will be about approximately 9 billion m^3 (Werner et al., 2001). Regarding the large number of residual lakes in Lusatia, the importance of knowledge about problems concerning AMD (Knoll et al., 1999) and

biogeochemical processes rises enormously (Friese et al., 1998). The understanding and assessment of processes taking place in these particular lakes require detailed information about groundwater – lake water interactions, mainly the amount of groundwater recharging the lakes.

2. Study Sites

For the investigation of groundwater – lake water interactions, three locations were chosen in the Lusatian lignite district, Eastern Germany (figure 1). Lake Waldsee and lake Moritzteich are situated in the *Muskau Arch Fold*, about 150 km southeast of Berlin. Both lakes show meromictic stratification pattern. Mine lake 107 (ML107) is located near Plessa, approximately 140 km in the south of Berlin and contrary to the previous lakes it shows a polymictic water body.

Figure 1: Map of the study area. Arrows indicate Waldsee (1), Moritzteich (2), and ML107 (3). The shaded signature shows the position of the *Muskau Arch Fold*.

2.1 Lake Waldsee and Lake Moritzteich

Both, lake Waldsee and lake Moritzteich are situated in a geological region called *Muskau Arch Fold*, an Elsterian ice-pushed ridge of a moraine. The geological and therewith hydrogeological setting is very complex due to folding and faulting caused by glacial and tectonic processes (Pechstein, 2008; Wolkersdorfer and Thiem, 1999). Tertiary, formerly horizontally deposited lignite, sand, silt, and clay layers were deformed and steeply uplifted (Kupetz, 1997) to a horseshoe-shaped belt of push moraines. The glaciotectonic distortion of the soft sediments attained ~270 m in depth beneath the 430-510 m thick glacier (Kupetz and Kessler, 1997). This process resulted in large-scale pattern of geologic structures such as faults, anticlines, synclines, and imbricated folds (Kupetz and Kessler, 1997).

148 The hydrogeological situation is complicated and as far unknown in detail. Kupetz
149 supposed that the aquifers in the synclines are separated from each other and show
150 only sparse or not any hydraulic connections (Kupetz, 1995). Previous investigations
151 showed that Waldsee is connected to the local groundwater body (Seebach et al.,
152 2008).
153 From geomorphological view, however the study area appears small-scale structured
154 with little hills and valleys following the lineament of the moraine loop. Various
155 abandoned underground and open pit excavations from lignite and clay mining shape
156 the landscape. Open cast as well as underground lignite mining in the study area
157 started in 1843 and lasted until 1959 (Kupetz, 1996). Recently, most of the
158 excavations are filled with acidified iron-rich water (Kasinski et al., 2004).
159 The climate in the study area is of temperate continental character with an average
160 temperature of 8.4°C by an annual temperature range between -1.3 and 17.9°C
161 (DWD, 2007). The mean humidity is nearby 80 % (Piehl, 1985) and long-term
162 precipitation records indicate an annual amount of 927 mm (Veit et al., 1987). Annual
163 potential evaporation rates for open surface water bodies with average depths of
164 2-4 m and 4-9 m amount to 762 mm and 751 mm, respectively (Richter, 1984).
165 Waldsee and Moritzteich are located in a densely wooded part of the *Muskau Arch*
166 *Fold*. The key data of the lakes are specified in table 1. Waldsee does not have a
167 surface inflow but a small outlet drains the lake if it reaches a certain lake level.
168 Moritzteich has a seasonal surface inflow on its west side and a surface discharge on
169 the southeast bank. After post-mining phreatic rise since 1960, Moritzteich attained
170 its recent lake level in the 1980s (Haselbach and Schoß, 1995). During the 1990s,
171 the surface water of Waldsee showed a lower pH (3.0-3.4) (Nixdorf et al., 1998) than
172 it recently does (~6.9). This might be probably a result of road constructions next to
173 the lake (Nixdorf et al., 2001). Both lakes are stratified and show meromictic

stratification pattern, which is they do not have a seasonal overturn of the whole water column. The reason for this characteristic is differences in water density, which are caused by temperature and/or solutes. In this case, stratification is maintained due to high content of iron and sulphate in the deep-water layers of Moritzteich and Waldsee (Dietz et al., 2009). The surface water parcels (mixolimnion) are separated from the deep-water body (monimolimnion) by a region of high vertical density gradients, called chemocline. The chemocline hampers the mass transport between these two lake sections, the turbulent component being far more important than the molecular diffusion for water transport.

Both lakes show a seasonal shift of the chemocline depth. During summertime, the chemocline is eroded by temperature induced mixing of the surface water, which reaches the chemocline region and which is stronger than the stability of the stratification. In winter however, enhanced groundwater inflow in the monimolimnion might be responsible for gradual chemocline rise by pushing it upward (Von Rohden et al., 2009).

2.2 Mine Lake 107 (ML107)

Mine lake 107 (ML107) is part of a pit lake area located near Plessa. In this open cast pit lignite mining lasted from 1897 until 1928 although mine drainage was ceased not until the 1960s (Nixdorf et al., 2001). The particular of ML107 is its low mean depth (1.1 m) accompanied with distinct excavation pattern (5.0 m) in the central part of the lake (LMBV, 2006). Because of its shallow character and the relative large surface area, the lake is polymictic (Lessmann et al., 1999). Due to pyrite and marcasite alteration, ML107 shows very low values in pH (~2.5). The lake is not linked to other lakes in its vicinity by surface in- and outflow and thereby is only controlled by precipitation, evaporation and its groundwater connection. The

seasonal lake level fluctuations amount to 0.43 m and result in volume variations of up to 35 %.

Table. 1: Key data of the investigated lakes.

Figure 2: Bathymetric maps of the investigated lakes Waldsee (1), Moritzteich (2), and ML107 (3).

3. Methods and Material

3.1 Theory

Following the general hydrologic water balance term:

$$\frac{\Delta V_L}{\Delta t} = P - E + I_S - O_S + I_{GW} - O_{GW} \quad (2)$$

the isotope water balance can be written as:

$$\frac{\Delta V_L \delta_L + \Delta \delta_L V_L}{\Delta t} = P\delta_P - E\delta_E + I_S\delta_S - O_S\delta_L + I_{GW}\delta_{GW} - O_{GW}\delta_L \quad (3)$$

where δ represents the mean isotope signature associated with the precipitation rate P, evaporation rate E, volumes of surface inflow I_S and surface outflow O_S as well as groundwater inflow I_{GW}, subsurface outflow O_{GW}, and V_L the mean volume of the lake during Δt. Goal of the water balance calculations was the evaluation of groundwater inflow rates to each lake described above. As two of the lakes show stratified water bodies, one has to calculate their water budgets disconnected from each other according to:

$$\frac{\Delta V_{MIX} \delta_{MIX} + \Delta \delta_{MIX} V_{MIX}}{\Delta t} = P\delta_P - E\delta_E + I_S\delta_S - O_S\delta_{MIX} + I_{GW}\delta_{GW} - O_{GW}\delta_{MIX} \quad (4a)$$

for the surface water body (mixolimnion) and:

$$\frac{\Delta V_{MON} \delta_{MON} + \Delta \delta_{MON} V_{MON}}{\Delta t} = I_{GW}\delta_{GW} - O_{GW}\delta_{MON} \quad (4b)$$

for the deep water layers (monimolimnion), which are separated from precipitation and evaporation impact. Taking into account the fluctuations of the chemocline depth as described earlier, the respective isotope water balance terms have to be modified concerning the water volume, which is removed from the monimolimnion by chemocline subsidence (Von Rohden et al., 2009). Following these assumptions, equations (4a), and (4b) can be completed to:

$$\frac{\Delta V_{MIX}\delta_{MIX} + \Delta\delta_{MIX}V_{MIX}}{\Delta t} = P\delta_P - E\delta_E + I_S\delta_S - O_S\delta_{MIX} + I_{GW}\delta_{GW} - O_{GW}\delta_{MIX} + \Delta V_C\delta_C \quad (5a)$$

and

$$\frac{\Delta V_{MON}\delta_{MON} + \Delta\delta_{MON}V_{MON}}{\Delta t} = I_{GW}\delta_{GW} - O_{GW}\delta_{MON} - \Delta V_C\delta_C \quad (5b)$$

where V_C is the volume of monimolimnion water, which is incorporated into the surface water layers due to chemocline erosion and δ_C the related weighted isotope signature. Accordingly, equation (2) has to be written as follows:

$$\frac{\Delta V_{MIX}}{\Delta t} = P - E + I_S - O_S + I_{GW} - O_{GW} + \Delta V_C \quad (6a)$$

$$\frac{\Delta V_{MON}}{\Delta t} = I_{GW} - O_{GW} - \Delta V_C \quad (6b)$$

Rearranging equations (5a), (5b), and substitution with (6a) and (6b), respectively allows the calculation of groundwater inflow rates for the mixolimnion and the monimolimnion in the following manner:

$$I_{GW_MIX} = \frac{\Delta\delta_{MIX}V_{MIX} + E\delta_E - P\delta_P - I_S\delta_S + \delta_{MIX}(P - E + I_S) + V_C(\delta_{MIX} - \delta_C)}{\delta_{GW} - \delta_{MIX}} \quad (7a)$$

and

$$I_{GW_MON} = \frac{\Delta\delta_{MON}V_{MON} - \Delta V_C(\delta_{MON} - \delta_C)}{\delta_{GW} - \delta_{MON}} \quad (7b)$$

Except the isotope signature of the lake evaporate δ_E, all water balance related parameters are easily to evaluate. To determine δ_E, Craig and Gordon found a

246 relationship between relative humidity h, fractionation factor α*, equilibrium (ε*) and
247 kinetic (ε_k) enrichment factors, isotope signatures of lake surface water as well as the
248 isotopic composition of atmospheric water vapour δ_A (Craig and Gordon, 1965):

249 $$\delta_E = \frac{\alpha^* \delta_L - h\delta_A - \varepsilon}{1 - h + 10^{-3}\varepsilon_k}$$ (Gibson and Edwards, 2002) (8)

250 The variables in equation (8) are described in the following. The fractionation factors
251 α* for deuterium as well as oxygen-18 were calculated according to Horita and
252 Wesolowski (Horita and Wesolowski, 1994). The total enrichment factor ε is defined
253 as:

254 $$\varepsilon = \varepsilon^* + \varepsilon_k$$ (9)

255 with the equilibrium enrichment factor ε*, derived from α* and the kinetic fractionation
256 factor ε_k, which is a function of relative humidity and may be determined simply by
257 the following equation (Gonfiantini, 1986):

258 $$\Delta\varepsilon_k{}^{18}O = 14.2(1-h)$$ for oxygen-18 (10a)

259 $$\Delta\varepsilon_k D = 12.5(1-h)$$ for deuterium (10b)

260 Assuming equilibrium conditions between local precipitation and atmospheric water
261 vapour, the isotope signature of the ambient water vapour δ_A can be determined by
262 (Froehlich, 2000):

263 $$\delta_A = \delta_P - \varepsilon^*$$ (11)

264 Several other authors worked with a modified form of equation (8):

265 $$\delta_E = \frac{1}{1 - h + \varepsilon_k}\left(\frac{\delta_L - \varepsilon^*}{\alpha} - h\delta_A - \varepsilon_k\right)$$ (Gonfiantini, 1986) (12)

266 $$\delta_E = \frac{\delta_L - h\delta_A - \varepsilon}{1 - h + \varepsilon_k}$$ (Gibson et al., 1999) (13)

267 $$\delta_E = \frac{\delta_L - h\delta_A - \varepsilon}{1 - h}$$ (Vallet-Coulomb et al., 2006) (14)

Nevertheless, all of the given terms harbour uncertainties concerning the impact of relative humidity. The application of these equations in temperate climates with typically high relative humidity (70-80 %) yields very uncertain values of δ_E (Zimmermann and Ehalt, 1970). This is caused by the moisture exchange between atmosphere and lake surface; at high ambient humidity the reflux becomes notably overstated (Gat, 1995).

3.2 Field Work, Sampling Procedures and Data Processing

Several groundwater monitoring wells were installed around each lake (see fig. 8). For isotope analyses, groundwater samples were taken with a peristaltic pump (Eijkelkamp) on a bimonthly basis after purging the wells until steady state conditions of electric conductivity (κ_{25}), pH, and temperature were achieved. Sampling of vertical lake water profiles was conducted every month with a 10 cm resolution in the chemocline regions of both Waldsee and Moritzteich. ML107 was sampled in a 50 cm resolution every second month. The data sets of the vertical profiles were interpolated to a 10 cm resolution for each lake. Precipitation was collected once a month at each lake for the entire sampling interval with a Hellmann device. The bottles in the samplers were filled with polystyrene pellets to protect the collected precipitation against evaporation. Additionally, the precipitation samplers were installed in the lake to prevent heating of the sampling system in the summer months. In-lake evaporation pans of 200 L volume were installed at each lake and sampled monthly. The evaporation pan related to Moritzteich was deployed on a nearby lake in order to protect it against vandalism. Purpose of the evaporation pans was the evaluation of evaporation height as well as the determination of isotopic reference numbers linked to the evaporation process. Depending on precipitation and evaporation rates, the pans were refilled with lake water or decanted to reobtain their

initial volume. In case of replenishment, the new water composition within the evaporation pan as well as the refilled water was sampled. The amount of the refilled or decanted water was documented to calculate evaporation rates. To complete the survey in all water balance related parameters, a new method for collecting lake evaporate was applied. Therefore, a construction had been designed that affords average samples on a monthly basis (Seebach et al., 2010b). The water vapour sampling unit was positioned on a floating platform and placed into a plastic rain barrel. A car battery was linked to a clock timer that operated a peristaltic pump, which drew adjacent air in 10 cm height above lake surface. The air was piped into gas sampling bags that were stored into the rain barrel. The pumps worked every four hours for 30 minutes. The sample bags contained 60 and/or 80 L. The evacuation of the sample bags took place by using cold traps connected to vacuum exhaust. Vertical profiles of electrical conductivity were determined monthly at Waldsee and Moritzteich with an automatic CTD probe (Idronaut, Brugherio, Italy) and a resolution of 2 cm. Additionally, meteorological data (air temperature, relative humidity, wind speed, and wind direction) were recorded in a 10-minute interval by weather stations on Waldsee and Moritzteich, respectively. At both lakes, a thermistor chain logged water temperatures in several depths every 10 minutes. For ML107 no local meteorological and surface water temperature data were available. Therefore, accordant data were transferred from a nearby mine lake (ML111). Boehrer et al. could demonstrate the validity of data migration from one lake to another (Boehrer et al., 2000).

3.3 Laboratory Measurements

All samples from lake water, groundwater, precipitation, and lake evaporate as well as from evaporation pans were analysed for stable isotope ratios of oxygen-18 and

deuterium. Isotope ratios are expressed in δ-notation as part per thousand deviation relative to Vienna Standard Mean Ocean Water (VSMOW). Except for lake evaporate, the measurements were carried out using a H_2/CO_2 equilibration unit coupled with a Thermo Finnigan Delta S isotope ratio mass spectrometer. The analytical uncertainty is 0.1 ‰ for $\delta^{18}O$ and 0.8 ‰ for D, respectively. Due to their little volume, lake evaporate samples were measured with pyrolysis method described by Gehre et al (Gehre et al., 2004). Therefore, only 0.5 to 1.0 µL of liquid sample are sufficient for the on-line high temperature analysis of deuterium and oxygen-18 with a measurement precision of 0.1 ‰ for $\delta^{18}O$ and <1 ‰ for δD, respectively.

4. Results and Discussion

4.1 Isotope distribution in Lake Water

The isotope variations in vertical lake water profiles followed the seasonal cycle to a greater or lesser extent, depending on the respective depth and volume of the well-mixed surface water parcels. For Waldsee and Moritzteich the interpolated oxygen-18 profiles scattered from -7.98 ‰ to -3.33 ‰ and -7.77 ‰ to -2.44 ‰ in the mixolimnia, respectively. In the monimolimnia, the values spread between -8.59 ‰ and -3.91 ‰ for Waldsee, and -8.83 ‰ and -3.51 ‰ for Moritzteich. The oxygen-18 profiles of ML107 showed a seasonal cycle as well. The extend of the ranging $\delta^{18}O$ values was 5.33 ‰ from a minimum of -4.16 ‰ in the deepest water samples to a maximum of +1.17 ‰ for summerly surface waters.

Figure 3: Vertical lake water profiles of the three study lakes Waldsee (1), Moritzteich (2), and ML107 (3). The figures show the interpolated range of the oxygen-18 values (left) and mean electric conductivity (κ25) profiles (right). For visualisation of the varying chemocline region, the dashed lines show measured profiles of sampling dates with the uppermost and lowest position of the density gradients.

4.2 Isotope compositions of local groundwater, precipitation and evaporation

Groundwater

Groundwater observation wells at the investigated sites showed distinct pattern of isotope signatures during the 2.5 years sampling period. At Waldsee, oxygen-18 values between -9.59 ‰ (MWS-3) and -5.53 ‰ (MWS-4) were found. Observation well MWS-6 (mean -8.28 ‰) and MWS-7 (mean -8.41 ‰) on the western shore of the lake showed the lightest isotope signatures in contrast to MWS-4 at the southern bank (mean -6.30 ‰). The isotope distribution at Moritzteich was characterised by a scatter from -9.73 ‰ (MMT-3) to -5.76 ‰ (MMT-9) for oxygen-18. The evaluation of isotope data of 17 observation wells showed that groundwaters could be clustered into three groups. One bunch defines groundwater wells, which were always isotopically lighter than the Moritzteich lake water, the second groundwater well group shows more enriched water than the lake water, and the third group was characterised by significant changes in isotopic composition of the respective groundwater. The classification of groundwater wells located at ML107 was less difficult: Values varied between -0.93 ‰ (M107-1) and -9.69 ‰ (M107-4). In average, wells M107-3, M107-4, and MPL-11 were characterised by isotopically light groundwater (-9.34 ‰ to -8.47 ‰ for $\delta^{18}O$). Wells M107-1, M107-2, and MPL-1 displayed enriched water samples between mean values of -5.38 ‰ and -1.85 ‰.

It should be pointed out that the isotopic composition of deep aquifers is generally homogeneous, and the seasonal changes are often small and sometimes disappear (Longinelli et al., 2008). In our case studies, seasonal variations in groundwater isotope signatures were significant and allowed the assumption, that the lake water regime is linked very closely to the groundwater system.

Table 2: Summary of the measured oxygen-18 and deuterium values in groundwater wells at Waldsee (1), Moritzteich (2), and ML107 (3).

376

377 **Precipitation and Evaporation**

378 Precipitation collected at each study site indicated similarity in isotope values and
379 annual distribution. The measured data were corrected according Richter (Richter,
380 1995) regarding errors induced by evaporation and the wind. Figure 4c/d compares
381 the three investigated sites concerning precipitation amount as well as its isotope
382 signatures. Isotope signatures in local precipitation vary between -1.32 ‰ and
383 -13.82 ‰ for Waldsee, -1.70 ‰ and -13.00 ‰ for Moritzteich and -3.69 ‰ and
384 -15.90 ‰ for ML107, respectively. Values for monthly precipitation heights in the
385 entire investigation period scattered from 7 mm for Moritzteich in May 2008 and
386 136 mm at Waldsee in May 2007.

387 Missing data of total evaporation (vandalism, overflow of the pans as a result of
388 strong rain events etc.) were substituted by values calculated according Linacre
389 (Linacre, 1977). Monthly evaporation heights at ML107 were found to spread among
390 3 mm in December 2007 and 124 mm in June 2008. At Waldsee, evaporation heights
391 were between 10 mm in December 2006 and 124 mm in July 2006. At Moritzteich,
392 the range of evaporation height values was between 7 mm for February 2007 and
393 111 mm in March 2007. At each study site, the seasonal meteorological conditions
394 correspond very well with the fluctuations of the lake levels (see figure 4e).

395 **Figure 4: Summary of water balance related parameters for each lake.**
396

397 **Isotope Signatures of Lake Evaporate**

398 At two study sites (Waldsee and Moritzteich) the isotope signature of the lake
399 evaporate could measured directly for several sampling periods.

400 For sampling intervals with lack of measured data, eqs. (8), (12), (13), and (14) were
401 used to find the best fit with the local evaporation line. Therefore, an evaporation line

related to the data of Waldsee was generated for each equation. Figure 5a demonstrates the different scatter plots for the current equations. The evaporation line related to equation (8) shows an impressive agreement with the local evaporation line. For this reason, missing data of lake evaporate were calculated according equation (8). Even if the sampling procedure described by Seebach et al. (Seebach et al., 2010b) might still retrieve uncertainties, the achieved values seem to be more reliable than data obtained by eq. (8) derived by the Craig & Gordon model (see figure 5b). Figure 6 gives an impression of the distinct magnitudes of directly obtained δ_E values versus calculated data in temporal dimension. It is obvious, that the deviation between the data sets gets large in periods with high relative humidity. However, since lake evaporate isotope signatures are demanded for the high resolution water balance, calculated ones according eq. (8) substitute missing measured data.

Figure 5a: Comparison of calculated δ_E values for Waldsee in D / O space. Equations (8), (12), (13), and (14) were used to demonstrate the scatter of calculated data using different terms for evaluating δ_E.

Figure 5b: Comparison between measured and calculated values of lake evaporates' isotope signatures in D / O space. Measured δ_E data are only available for several sampling intervals at Waldsee and Moritzteich. The dashed box shows a detailed capture of the region with measured data points.

Figure 6: Comparison between measured and calculated values of lake evaporates' isotope signatures in time series related to relative humidity. Measured δ_E data are only available for several sampling intervals at Waldsee and Moritzteich.

The use of evaporation pan experiments in order to gain knowledge about fractionation processes, the derivation of water balance related parameters to specify eqs. (8), (12), (13), and (14) was not successful. During processing the evaluated data it got looming, that the sampling intervals were too long concerning the poor weather conditions in the study area. Hence, the evaporation pans only offered evaporation height data.

4.3 Isotope Water Balance

The main focus on this study was to estimate groundwater inflow rates into the three lakes on a monthly basis. For each lake, the water balance model was calculated using different values for the input parameter δ_{GW}. At first, for lake Waldsee and lake Moritzteich, the model was run with the background value of local evaporation unaffected groundwaters, in this case, the lightest groundwater isotope value obtained at the study site ("background value approach"). Secondly, the lightest δ_{MIX} value was chosen to estimate subsurface inflow rates ("lightest mixolimnion sample approach"). As a third step, the lightest δ_{MON} value was considered in the model ("lightest monimolimnion sample approach"). The different values for δ_{GW} used for the water balance for each lake are summarised in tab. 3. The estimation of inflow rates into the mixolimnia followed eq. (7a), inflow rates into the monimolimnia were conducted according eq. (7b).

Table 3: Overview about used δ_{GW} values for estimating groundwater inflow rates.

Following eqs. (7a), (7b), and (8) using the required parameters presented in (fig. 4) as well as the respective δ_{GW} values exhibited in table 3, yielded results for subsurface inflow rates compiled in figure 7.

Lake Waldsee

With the "background value approach" the gained subsurface inflow rates I_{GW_MIX} for lake Waldsee mixolimnion varied between +63 m^3d^{-1} and -43 m^3d^{-1} respective +53 m^3d^{-1} and -51 m^3d^{-1} for oxygen-18 and deuterium. The "lightest mixolimnion sample approach" gained inflow rates I_{GW_MIX} between +94 m^3d^{-1} and -68 m^3d^{-1} respective +73 m^3d^{-1} and -76 m^3d^{-1} for oxygen-18 and deuterium. With the "lightest

460 monimolimnion sample approach", inflow rates from -56 m^3d^{-1} to +79 m^3d^{-1} and
461 -50 m^3d^{-1} to +52 m^3d^{-1} were maintained for oxygen-18 and deuterium, respectively.
462 Modelling groundwater inflow rates into lake Waldsee monimolimnion with the
463 "background value approach" yielded I_{GW_MON} values from -26 m^3d^{-1} to +25 m^3d^{-1} and
464 -84 m^3d^{-1} to +114 m^3d^{-1} for oxygen-18 and deuterium, respectively. Applying the
465 "lightest monimolimnion sample approach" surrendered groundwater inflow rates
466 I_{GW_MON} between +49 m^3d^{-1} and -48 m^3d^{-1} for oxygen-18 respective +114 m^3d^{-1} and
467 -84 m^3d^{-1} for deuterium.
468
469 **Lake Moritzteich**
470 For lake Moritzteich mixolimnion the "background value approach" reached inflow
471 rates I_{GW_MIX} of -4713 m^3d^{-1} to +4295 m^3d^{-1} and -2520 m^3d^{-1} to +2432 m^3d^{-1} for ^{18}O
472 and D, respectively. The "lightest mixolimnion sample approach" gained inflow rates
473 I_{GW_MIX} between +6934 m^3d^{-1} and -7450 m^3d^{-1} respective +3577 m^3d^{-1} and
474 -3659 m^3d^{-1} for oxygen-18 and deuterium. With the "lightest monimolimnion sample
475 approach", inflow rates from -5574 m^3d^{-1} to +5113 m^3d^{-1} and -2133 m^3d^{-1} to
476 +2048 m^3d^{-1} were calculated for oxygen-18 and deuterium, respectively.
477 Assessing inflowing groundwater I_{GW_MON} into the lake Moritzteich monimolimnion by
478 means of the "background value approach" varied from -1658 m^3d^{-1} to +853 m^3d^{-1} for
479 oxygen-18 and from -9155 m^3d^{-1} to +4209 m^3d^{-1} for deuterium, respectively. The
480 application of the "lightest monimolimnion sample approach" for quoting groundwater
481 recharge into the monimolimnion provides values between +1533 m^3d^{-1} and
482 -3512 m^3d^{-1} respective +2144 m^3d^{-1} and -3518 m^3d^{-1} for oxygen-18 and deuterium.
483
484 **ML107**

For ML107 only two models were calculated; namely, the "background value approach" and the "lightest mixolimnion sample approach" which worked in this case with the lightest isotope signature ever evaluated in ML107 lake water.

Determining subsurface inflow rates I_{GW_MIX} into Mine lake 107 with the "background value approach" yielded dimensions between +620 m^3d^{-1} and -489 m^3d^{-1} respective +358 m^3d^{-1} and -1366 m^3d^{-1} for oxygen-18 and deuterium. Using the "lightest mixolimnion sample approach" inflow rates from -6297 m^3d^{-1} to +5229 m^3d^{-1} and -10019 m^3d^{-1} to +961 m^3d^{-1} were calculated for oxygen-18 and deuterium, respectively.

Figure 7: Calculated groundwater inflow rates into the investigated lakes using distinct values for δ_{GW}. For lake Waldsee and lake Moritzteich, inflow rates are presented separately for mixolimnion and monimolimnion, respectively. For ML107 the values rate for the entire water body.

4.4 Problems associated with Isotope Water Balance Models

It is obvious, that the calculated subsurface inflow rates for all of the three lakes are questionable regarding the negative values obtained applying any of the input data for δ_{GW}. In respect to lake Waldsee and lake Moritzteich, the stratified water column could explain the inapplicability of the foregone equations. On the other hand, for ML107 even eq. (7a) had to be valid to evaluate confident groundwater inflow rates in the well mixed water body. Even eq. (7) was not applicable to determine inflow rates into ML107. For this reason, another common term was approached to approximate inflow rates. According to

$$X = E/I \tag{15}$$

where X is the fraction of inflowing water lost by evaporation described by Gonfiantini (Gonfiantini, 1986) in the following manner

$$X = \frac{(\delta_{MIX} - \delta_P)(1 - h + \Delta\varepsilon)}{(\delta_{MIX} + 1)(\Delta\varepsilon + \varepsilon/\alpha^*) + h(\delta_A - \delta_{MIX})} \tag{16}$$

513 and rearranging eqs. (15) and (16) arises to

514 $$I = E\frac{(\delta_{MIX}+1)(\Delta\varepsilon+\varepsilon/\alpha^*)+h(\delta_A-\delta_{MIX})}{(\delta_{MIX}-\delta_P)(1-h+\Delta\varepsilon)} \quad (17)$$

515 The calculated values for X are presented in tab. 4. The resulting groundwater inflow
516 rates are summarised in tab. 5.

517 However, even if this equation only works for ML107 and the well mixed mixolimnion
518 water parcels in lake Waldsee and lake Moritzteich, respectively, the received inflow
519 rates did not convince at all. Again, negative values for inflow rates were obtained.
520 This fact might be due to the calculated values for X (see tab. 4). Theoretically, the
521 fraction X has to evince values between 0 and 1. Concerning the data presented in
522 tab. 4, which were calculated for oxygen-18 and deuterium, respectively, negative
523 values as well as values >1 occurred for each lake. It follows, that also the equation
524 (17) is inapplicable because of unreliable values of X.

525 Regarding an integrative approach for modelling inflow rates on a yearly basis, using
526 equations (7a) and (7b), yielded data that were more reliable. According to that, for
527 the first investigation year at lake Waldsee we could determine daily inflow rates
528 I_{GW_MIX} between 15 … 22 m^3d^{-1} and 20 … 29 m^3d^{-1} for oxygen-18 and deuterium,
529 respectively. For the second year at lake Waldsee, the inflow rates I_{GW_MIX} amount
530 from 8 … 11 m^3d^{-1} and 11 … 15 m^3d^{-1} for oxygen-18 and deuterium, respectively.
531 This was round about half of daily inflow rates during the first year. Inflow rates
532 I_{GW_MON} into lake Waldsee accounted for the first investigation year only negative
533 inflow rates, which is unacceptable. For the second year, daily inflow rates I_{GW_MON}
534 from 1 … 2 m^3d^{-1} and 7 m^3d^{-1} were deduced for oxygen-18 and deuterium,
535 respectively.

536 For lake Moritzteich daily inflow rates I_{GW_MIX} between 432 m^3d^{-1} and 688 m^3d^{-1} for
537 ^{18}O and 326 m^3d^{-1} to 562 m^3d^{-1} for ^2H were calculated. The estimation of I_{GW_MON}

yielded results from 52 m^3d^{-1} to 57 m^3d^{-1} for oxygen-18 and 56 m^3d^{-1} to 60 m^3d^{-1} for deuterium. Even if the gained results for inflow rates into the monimolimnion of appear significantly more consistent than the respective mixolimnion inflow rates, it is obvious, than the model did not work again with the ascertained input parameters. The relation between mixolimnion and monimolimnion water body is approx. 4:1, according to I_{GW_MON} of ~50 … 60 m^3d^{-1}, the inflow rates in the upper water body must be 4 times higher than that value to maintain constant stratification conditions in the water column. The calculated data for I_{GW_MIX} might be overestimated.

The assessment of daily inflow rates into ML107 was completely unsuccessful – feeding the model with ^{18}O data yielded negative inflow rates (-391 … -103 m^3d^{-1}). At least calculating the model with deuterium data, reaches inflow rates between 256 m^3d^{-1} and 949 m^3d^{-1}.

Table 4: Summary of the obtained values of X (fraction of inflowing water lost by evaporation) for the investigated lakes Waldsee, Moritzteich, and ML107 for each sampling interval and average values for entire years during the investigation period.

Table 5: Summary of the obtained groundwater inflow rates based on different values of δ_{GW} for the investigated lakes Waldsee, Moritzteich, and ML107 for each sampling interval and average values for entire years during the investigation period.

As we could show, none of the models provided reliable data under the existing conditions at each study site. This fact constrains the following reflections:

Hydrogeological frame conditions

The isotope signature of the local groundwater is an essential input parameter for isotope water balance models. As described in the previous, the distribution pattern of isotope signatures in groundwaters occurring in the study area *Muskau Arch Fold* is very heterogeneous. Furthermore, the study sites might not be explored sufficient enough regarding the hydrogeological setting. The selection of the lakes was

568 primarily directed towards their stratification pattern. Only the research of Pechstein
569 (Pechstein, 2008) produced a small scale assessment of the (hydro-) geological
570 structure of the study site. During these investigations he tried to refer the sampled
571 groundwater wells at lake Waldsee and lake Moritzteich to the different aquifers. The
572 linkage of isotope data with hydrodynamic aspects (survey of the groundwater
573 heights in the wells relative to lake levels) showed that at lake Waldsee only three
574 well locations could potentially feed the lake from hydrodynamic perspective. Namely
575 the wells MWS-2, MWS-3, and MWS-4. From isotope hydrological view groundwater
576 recharge is only possible from ESE located MWS-3. But it is of vital importance, that
577 there is another small mine lake ca. 100 m south of lake Waldsee. Since the
578 hydrodynamic gradient between the “precursor lake” towards lake Waldsee was
579 constant throughout the investigation period, one could assume that MWS-4
580 contained already evaporative enriched water which flowed into lake Waldsee.
581
582 **Figure 8: Theoretical groundwater flow directions concerning hydrodynamics and isotope**
583 **hydrology for lake Waldsee and lake Moritzteich.**
584
585 The situation at lake Moritzteich looks different. The hydrodynamic mapping looks
586 like the groundwater recharge happens nearly circular around the lake. From isotope
587 hydrological view the question arises in what sense MMT-5 governs the isotope
588 hydrology of the lake water. The groundwater containing in that well is in average
589 1.26 ‰ heavier than the monimolimnion water and 2.70 ‰ lighter than the
590 mixolimnion water. Likewise questionable is MMT-9 at the western shore of lake
591 Moritzteich. At this site, already isotopically enriched water could be infiltrated from
592 the nearby ditch system.
593 Another important uncertainty concerning the balance model is induced by the
594 considerable fluctuations of the lake levels (lake Waldsee 0.29 m, lake Moritzteich

595 0.41 m). Therefore, the hydrological gradients shift in minimum twice a year. During
596 the season of groundwater recharge, the lakes are mainly controlled by groundwater
597 inflow conditions. During the vegetation period, lake water probably infiltrates into the
598 aquifer. The interplay between infiltrating and exfiltrating conditions between aquifer
599 and lake water body makes it impossible to catch the “right” fingerprint of recharging
600 groundwater because of mixing up both water components in the aquifer nearby the
601 lake. Detailed aspects of investigated lake water groundwater interactions at lake
602 Waldsee and lake Moritzteich are described in Seebach et al (Seebach et al, 2010a).
603 Summarizing the groundwater related “problems” one can say, that water balance
604 models only work with one single groundwater component of known isotope
605 signature, which is not given at lake Waldsee and lake Moritzteich, respectively.
606
607 **Meteorological frame conditions**
608 Published research, where isotope water balance models have been applied, were
609 mostly conducted in areas with relative low ambient humidity and or for evaluating
610 evaporation heights. Concerning this, the work of Gibson et al., Dinçer and Gat
611 should be mentioned. Each of the investigated study areas was blessed with
612 advantageous local frame conditions that allowed neglecting important parameters.
613 Reviewing the cited literature regarding their focus and the applied methods allows
614 demonstrating the limitation of commonly applied isotope water balance models to
615 very special questions as we presented here. The respective frame conditions of the
616 study sites are of immense importance for the results. However, in the following we
617 itemize some selected isotope water balance attempts, which were undertaken
618 during the last decades, their frame conditions, and the respective objective target:

- while Dinçer (Dinçer, 1968) evaluated the groundwater inflow rates into the two lakes in Turkey, he worked with a third lake of known water balance to adapt the critical parameters such as all evaporation related isotope signatures
- when Zimmermann and Ehalt (Zimmermann and Ehalt, 1970) attempted to determine the evaporation rate of lake Neusiedl in Austria, they eliminated the total inflow into the lake in the water balance term and solved the equation towards gaining results for total E
- the studies of Gat (Gat, 1970) describe the deduction of separation factors operating evaporation pans at lake Tiberias and the lakes known water balance
- Welhan and Fritz (Welhan and Fritz, 1977) used class-A evaporation pans to determine flux weighted δ_E for longer periods of time and other evaporation related isotope parameters whereas they excluded the inflow component
- the many studies of Gibson et al. (Gibson et al., 1996a) were conducted in Canada and most of them focussed on the determination of evaporation heights during the thaw season – because of the presence of thick permafrost deep groundwater interaction was negligible for their approaches
- also other authors' investigations failed due to the uncertainties originating from isotope water balance approaches, such as Longinelli (Longinelli et al., 2008), who struggled against adverse frame conditions, investigating the Garda lake water budget or Welhan and Fritz (Welhan and Fritz, 1975) who gained negative groundwater inflow rates into Perch lake

Technical frame conditions

The successful performance of long-term field experiments depends operatively on the duration of sampling intervals and the distance to the study area (in our case roundabout 300 km). For that reason, it was impossible to respond short-dated on

technical problems concerning the water vapour sampling unit. Oftentimes unpredictable complications arose and lead to losses of the actual data set as well as of the data set of the following sampling interval. Furthermore, the data processing indicated that the temporal sampling resolution was not adequate for the evaporation pan experiments. For the detailed assessment of evaporation experiments, daily sampling intervals during long precipitation free periods are sufficient. Realising these high requirements would not have been possible within these studies.

5 Conclusions

The stable isotope study of the three investigated lakes is of interest because it covers several features related to isotope water balance models. There are four primary conclusions of this work.

- In explicit detail, we could show that common approaches to balance water budgets of lakes are not suitable for stratified lakes even in the case of modified water balance equations.
- It was found that the successful application of isotope water balance model depends on many factors [local (hydro-) geological, meteorological and technical frame conditions], which are often not predictable.
- Despite of the raised problems related to isotope water balance models, a nearly complete two-year data set of meteorological and isotope data was ascertained for two stratified and a well-mixed lake in a former open cast pit area.
- Furthermore, the operation of a specifically designed water vapour sampling unit can be regarded to be successful.

Acknowledgements

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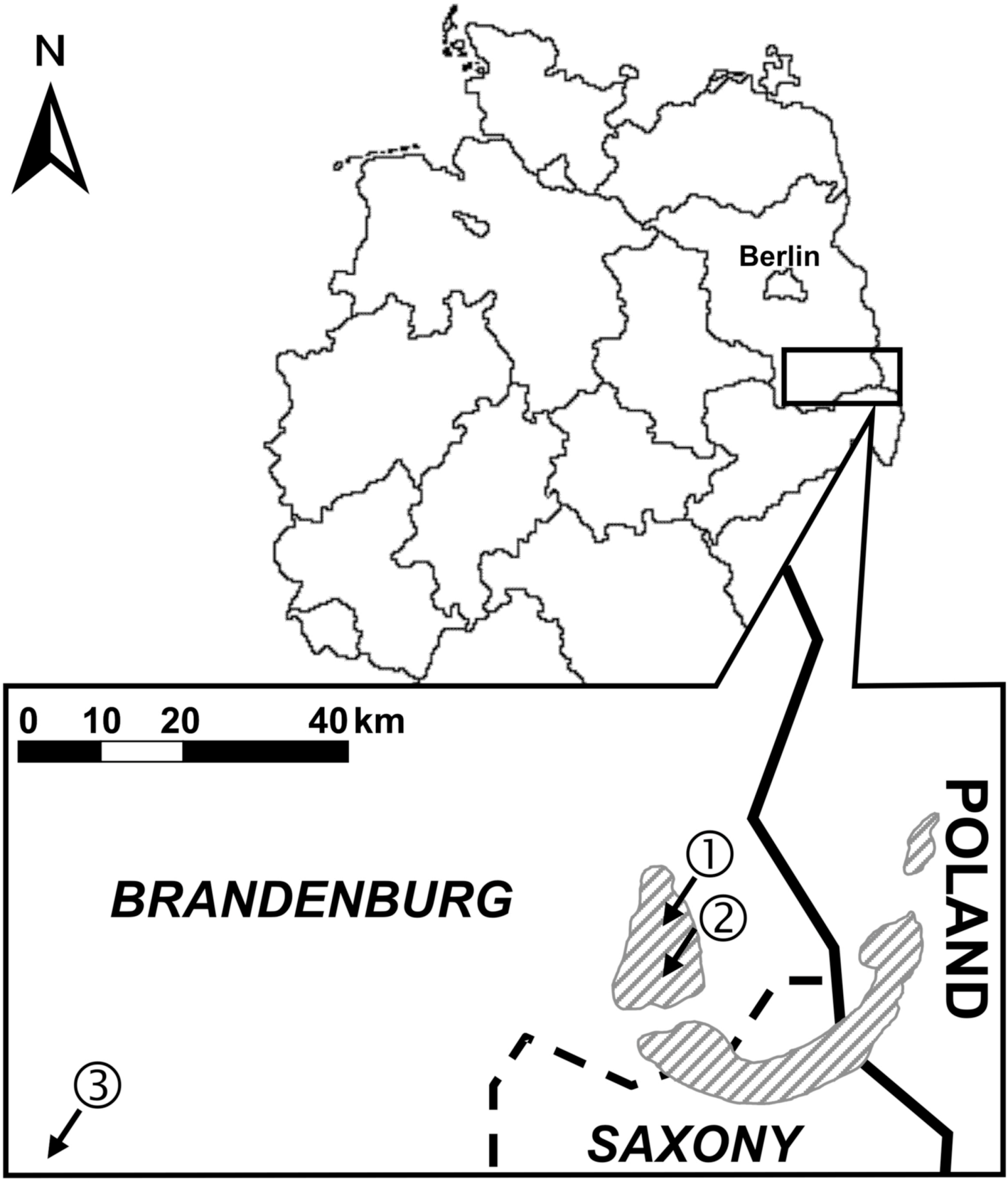
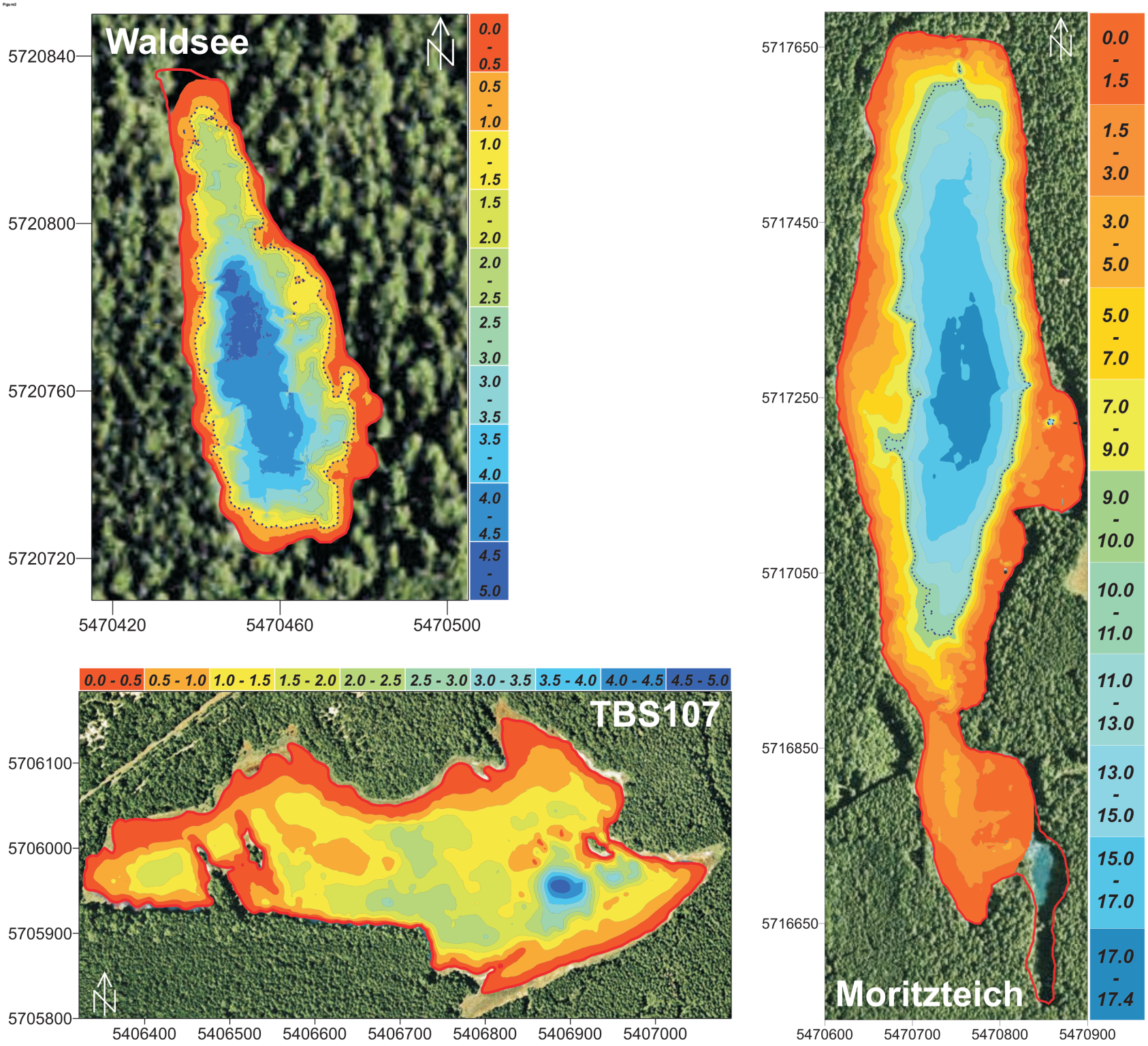
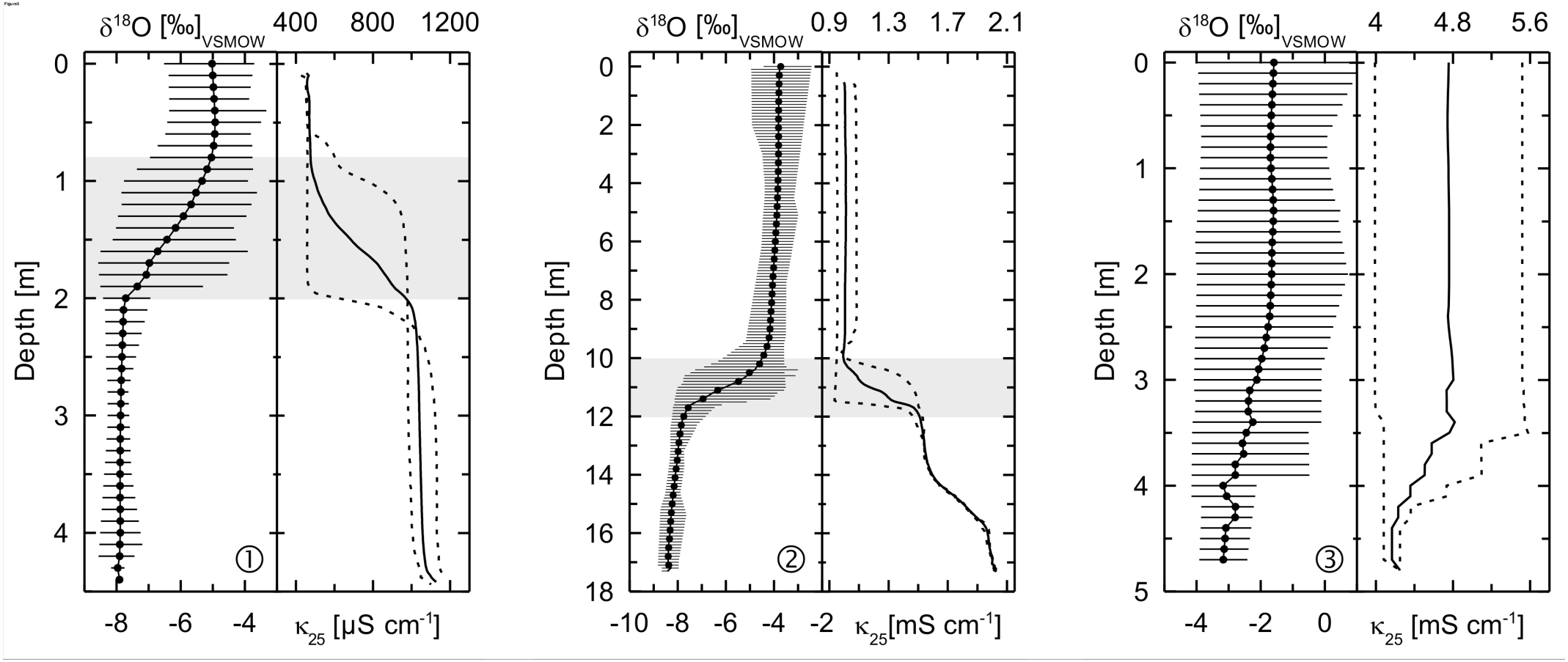
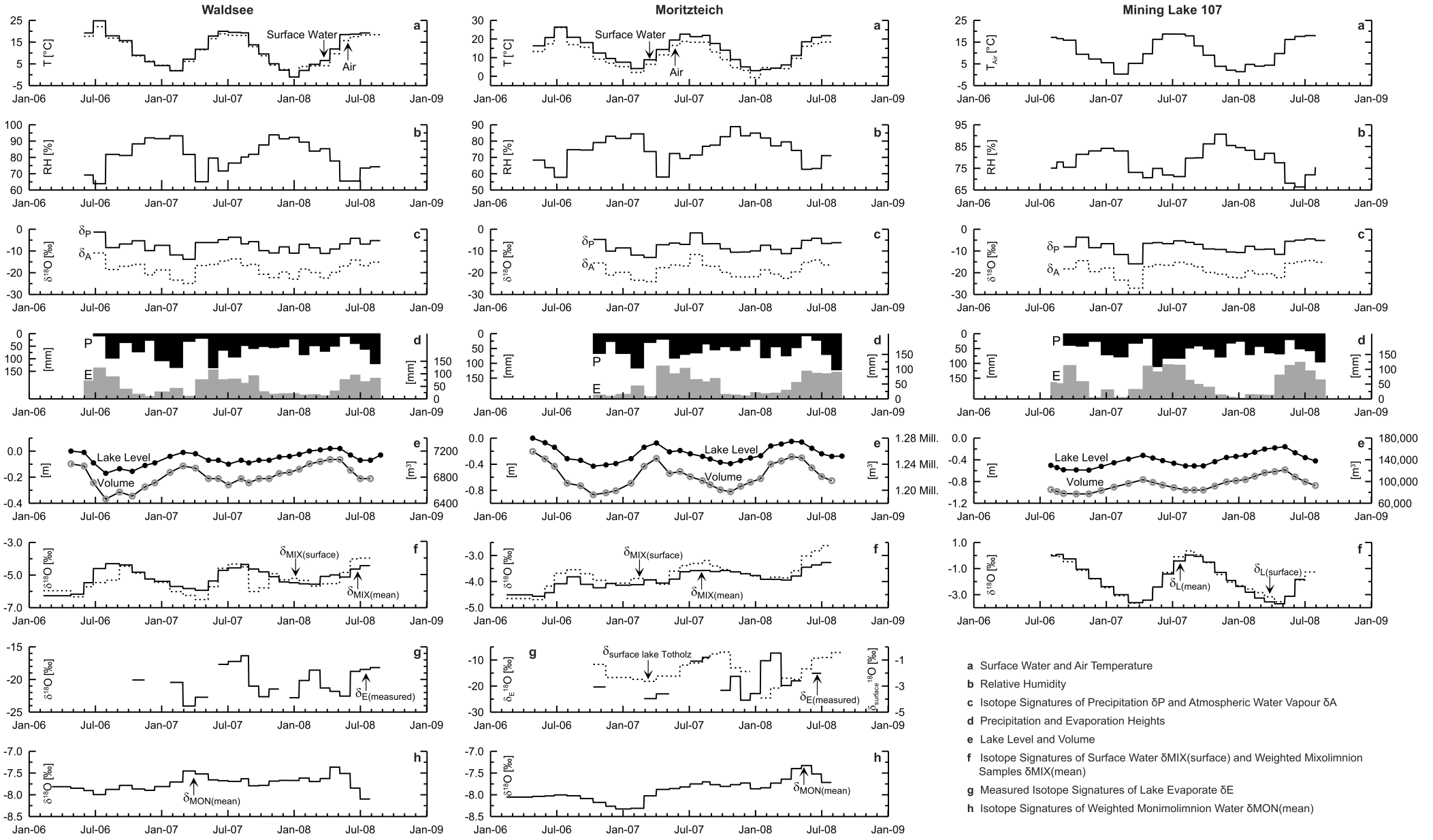
Figure1




Waldsee
5720840
5720800
5720760
5720720
5470420
5470460
5470500
0.0 - 0.5
0.5 - 1.0
1.0 - 1.5
1.5 - 2.0
2.0 - 2.5
2.5 - 3.0
3.0 - 3.5
3.5 - 4.0
4.0 - 4.5
4.5 - 5.0
0.0 - 0.5 0.5 - 1.0 1.0 - 1.5 1.5 - 2.0 2.0 - 2.5 2.5 - 3.0 3.0 - 3.5 3.5 - 4.0 4.0 - 4.5 4.5 - 5.0
TBS107
5706100
5706000
5705900
5705800
5406400
5406500
5406600
5406700
5406800
5406900
5407000
5717650
5717450
5717250
5717050
5716850
5716650
0.0 - 1.5
1.5 - 3.0
3.0 - 5.0
5.0 - 7.0
7.0 - 9.0
9.0 - 10.0
10.0 - 11.0
11.0 - 13.0
13.0 - 15.0
15.0 - 17.0
17.0 - 17.4
Moritzteich
5470600
5470700
5470800
5470900


δ18O [‰]VSMOW
400
800
1200
Depth [m]
0
1
2
3
4
①
-8
-6
-4
κ25 [μS cm-1]
δ18O [‰]VSMOW
0.9
1.3
1.7
2.1
0
2
4
6
8
10
12
14
16
18
②
-10
-8
-6
-4
-2
κ25[mS cm-1]
δ18O [‰]VSMOW
4
4.8
5.6
0
1
2
3
4
5
③
-4
-2
0
κ25 [mS cm-1]



a Surface Water and Air Temperature

b Relative Humidity

c Isotope Signatures of Precipitation δP and Atmospheric Water Vapour δA

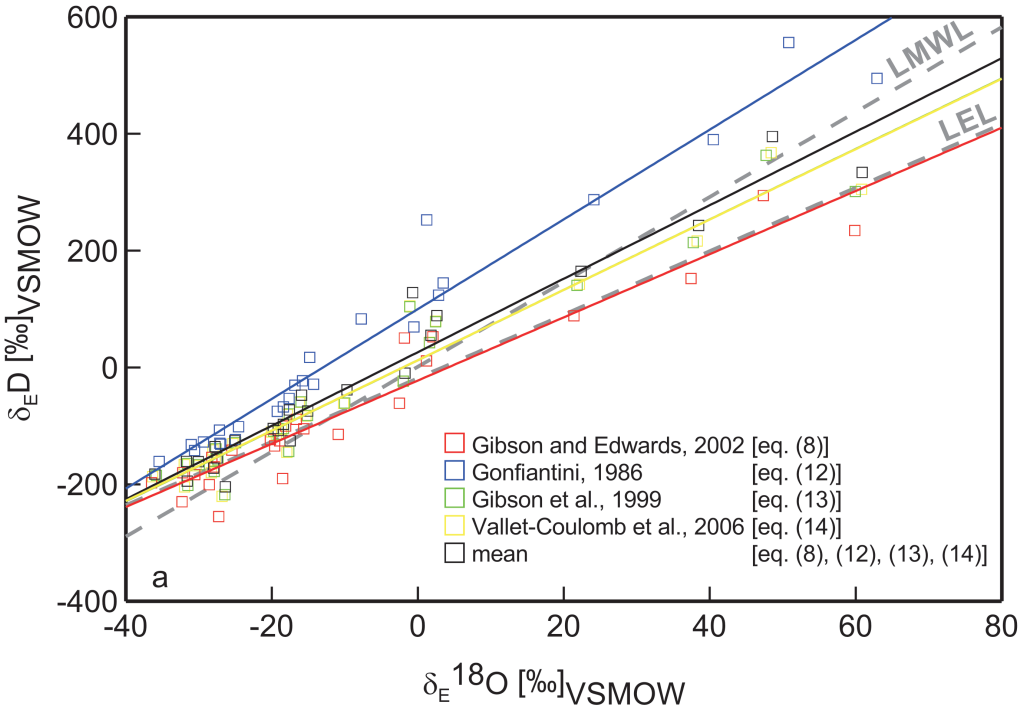
d Precipitation and Evaporation Heights

e Lake Level and Volume

f Isotope Signatures of Surface Water δMIX(surface) and Weighted Mixolimnion Samples δMIX(mean)

g Measured Isotope Signatures of Lake Evaporate δE

h Isotope Signatures of Weighted Monimolimnion Water δMON(mean)


600
400
200
0
-200
-400
-40
-20
0
20
40
60
80
δE D [‰]VSMOW
δE 18O [‰]VSMOW
LMWL
LEL
Gibson and Edwards, 2002 [eq. (8)]
Gonfiantini, 1986 [eq. (12)]
Gibson et al., 1999 [eq. (13)]
Vallet-Coulomb et al., 2006 [eq. (14)]
mean [eq. (8), (12), (13), (14)]
a

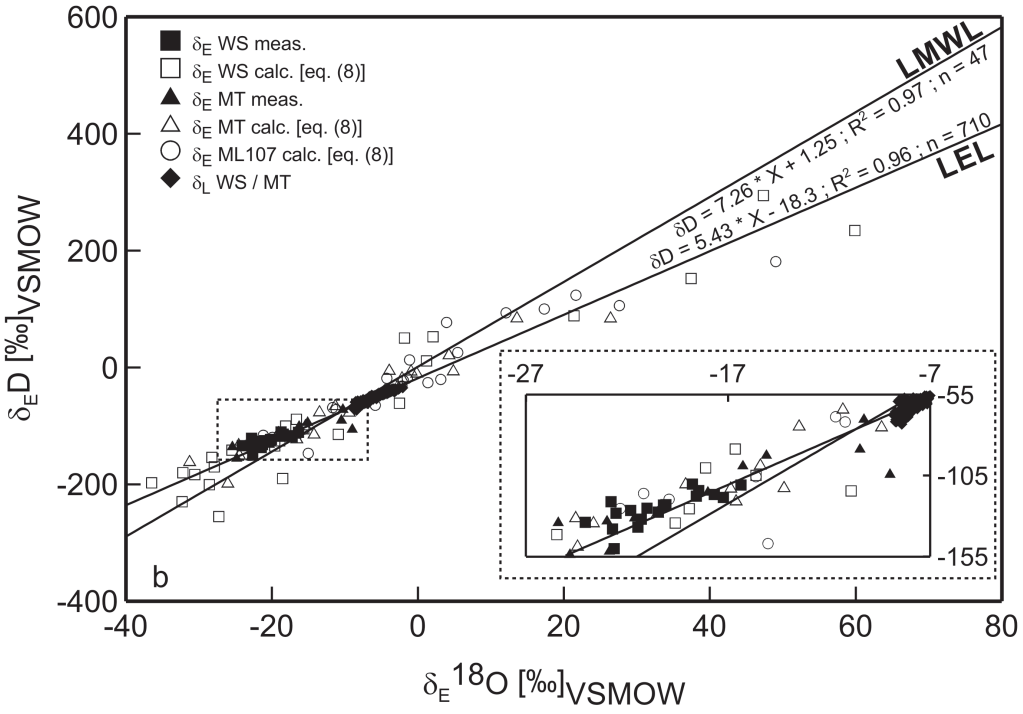
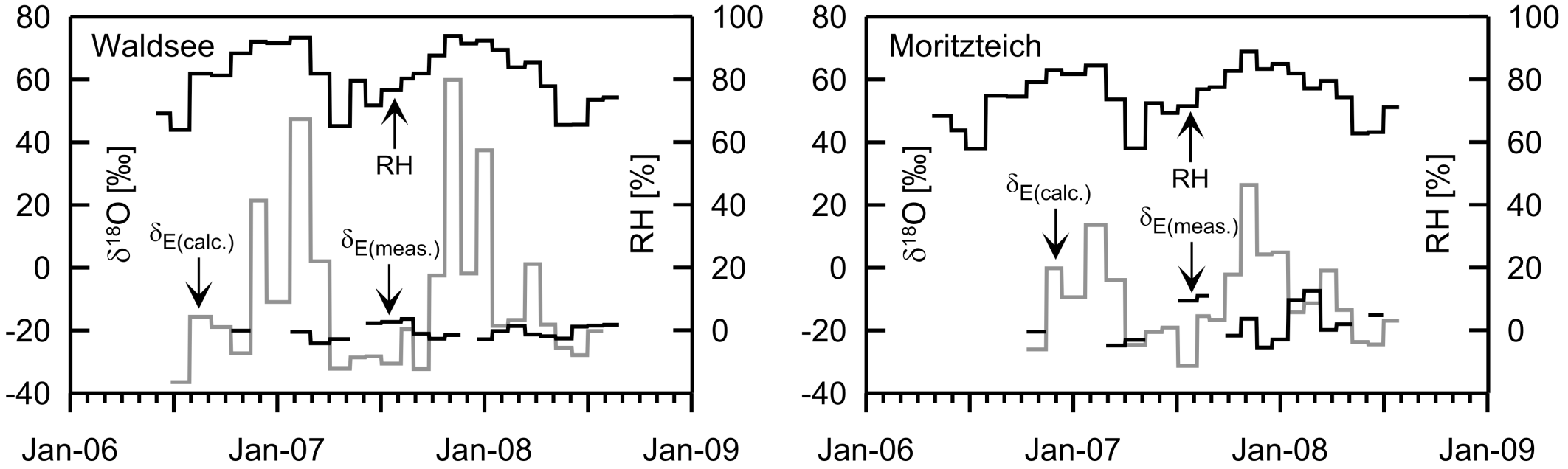
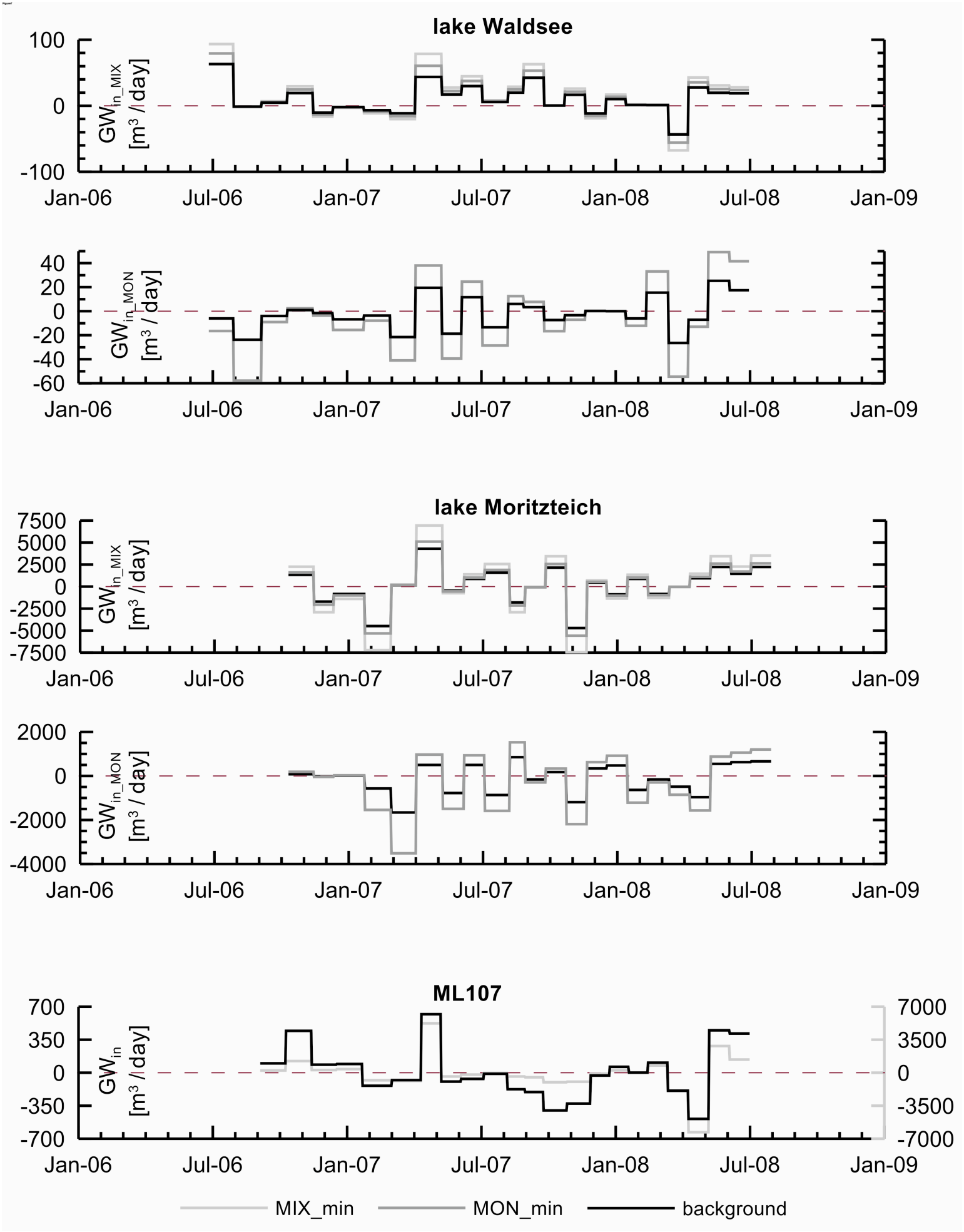

600
400
200
0
-200
-400
-40
-20
0
20
40
60
80
δE D [‰]VSMOW
δE 18O [‰]VSMOW
δE WS meas.
δE WS calc. [eq. (8)]
δE MT meas.
δE MT calc. [eq. (8)]
δE ML107 calc. [eq. (8)]
δL WS / MT
LMWL
LEL
δD = 7.26 * X + 1.25 ; R² = 0.97 ; n = 47
δD = 5.43 * X - 18.3 ; R² = 0.96 ; n = 710
-27
-17
-7
-55
-105
-155
b

Figure6


Waldsee
Moritzteich
δ18O [‰]
RH [%]
δE(calc.)
δE(meas.)
RH
80
60
40
20
0
-20
-40
100
80
60
40
20
0
Jan-06
Jan-07
Jan-08
Jan-09


lake Waldsee
100
0
-100
GW_in_MIX [m³ /day]
Jan-06
Jul-06
Jan-07
Jul-07
Jan-08
Jul-08
Jan-09
40
20
0
-20
-40
-60
GW_in_MON [m³ / day]
lake Moritzteich
7500
5000
2500
0
-2500
-5000
-7500
2000
-2000
-4000
ML107
700
350
-350
-700
GW_in [m³ /day]
7000
3500
-3500
-7000
MIX_min
MON_min
background

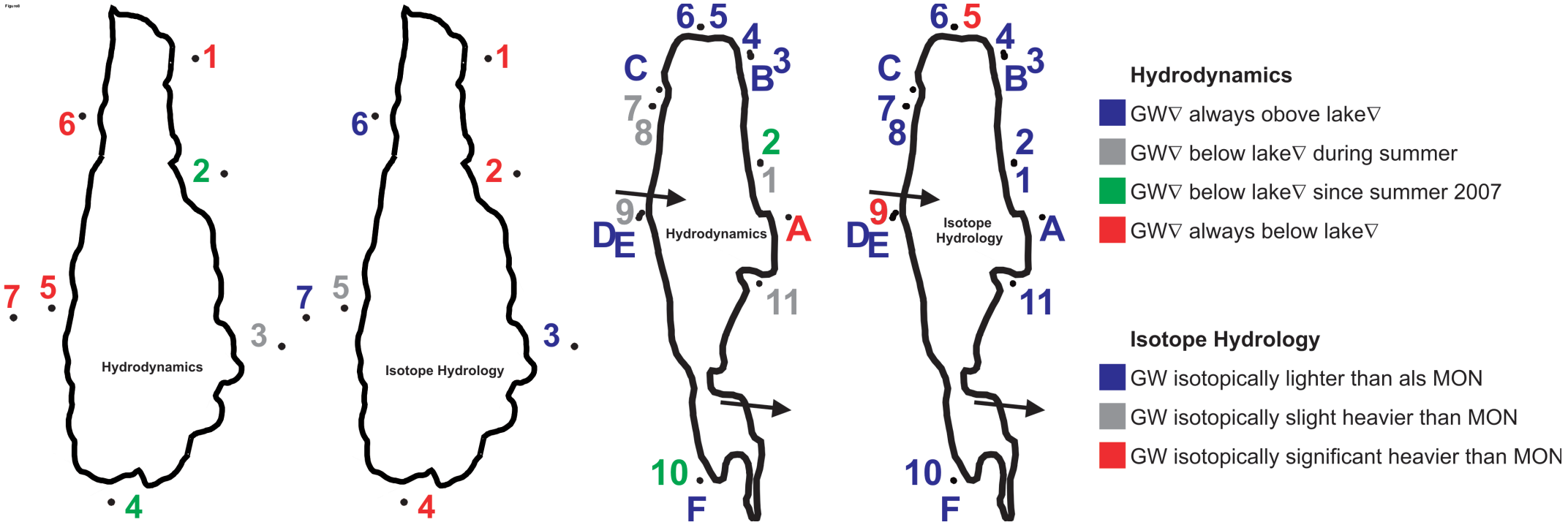

Hydrodynamics
Isotope Hydrology
Hydrodynamics
Isotope Hydrology
Hydrodynamics
GWV always above lakeV
GWV below lakeV during summer
GWV below lakeV since summer 2007
GWV always below lakeV
Isotope Hydrology
GW isotopically lighter than als MON
GW isotopically slight heavier than MON
GW isotopically significant heavier than MON

Table1
Click here to download Table: Tab.01_keydata_lakes.pdf

	lake Waldsee	lake Moritzteich	ML107
position	51°37'14.00'' N 14°34'16.88'' E	51°35'21.03'' N 14°34'33.24'' E	51°28'50.32'' N 13°39'18.44'' E
A [m^2]	3,318	165,779	113,866
V [m^3]	7,004	1.26 Mio.	128,245
V_{MIX} [%]	32 - 66	78 - 85	-
V_{MON} [%]	34 - 68	15 - 22	-
Z_{max} [m]	4.9	17.8	5.0
Z_{mean} [m]	2.1	7.6	1.1
κ_{25_MIX} [mS cm^{-1}]	0.49	1.00	4.64
κ_{25_MON} [mS cm^{-1}]	1.01	1.67	-
$pH_{_MIX}$	~6.9	~3.0	~2.5
$pH_{_MON}$	~6.7	~7.0	-

tab02
Click here to download Table: Tab.02_keydata_wells.pdf

	Depth [m]	Oxygen-18 [‰]$_{VSMOW}$			Deuterium [‰]$_{VSMOW}$		
		mean	max	min	**mean**	max	min
Waldsee							
MWS-1	7	**-7.22**	-6.57	-8.06	**-55.6**	-54.6	-57.1
MWS-2	6	**-7.88**	-7.24	-8.14	**-58.7**	-57.5	-60.2
MWS-3	4	**-9.10**	-8.16	-9.59	**-64.1**	-60.0	-66.6
MWS-4	4	**-6.30**	-5.53	-7.43	**-51.6**	-49.5	-57.9
MWS-5	4	**-7.34**	-6.92	-8.74	**-55.5**	-51.6	-59.5
MWS-6	5	**-8.28**	-7.64	-8.94	**-60.7**	-58.3	-62.8
MWS-7	2	**-8.41**	-7.21	-8.81	**-59.7**	-56.1	-61.6
Moritzteich							
MMT-1	3	**-8.83**	-7.47	-9.26	**-63.9**	-60.4	-65.1
MMT-2	7	**-9.17**	-8.72	-9.38	**-65.3**	-63.9	-65.9
MMT-3	7	**-9.59**	-9.43	-9.73	**-67.4**	-66.6	-67.9
MMT-4	3	**-9.56**	-9.40	-9.71	**-67.7**	-66.5	-68.2
MMT-5	10	**-6.62**	-5.87	-6.90	**-53.5**	-50.4	-55.6
MMT-6	3	**-8.46**	-7.84	-9.06	**-62.4**	-60.6	-63.6
MMT-7	8	**-9.07**	-8.30	-9.36	**-64.4**	-62.2	-65.5
MMT-8	4	**-9.11**	-8.43	-9.49	**-64.7**	-62.5	-67.3
MMT-9	3	**-6.32**	-5.76	-6.88	**-49.9**	-48.3	-52.6
MMT-10	6	**-9.26**	-8.10	-9.64	**-66.0**	-62.5	-67.3
MMT-11	5	**-9.02**	-8.13	-9.54	**-64.7**	.62.4	-66.6
MMT-A	18	**-9.04**	-8.87	-9.17	**-65.4**	-64.6	-67.1
MMT-B	17	**-8.95**	-8.51	-9.20	**-64.9**	-63.8	-66.0
MMT-C	15	**-8.89**	-7.44	-9.46	**-65.5**	-61.4	-67.2
MMT-D	8	**-8.69**	-7.71	-9.03	**-62.8**	-60.2	-63.6
MMT-E	21	**-9.24**	-7.93	-9.54	**-66.5**	-62.9	-67.4
MMT-F	17	**-9.00**	-8.01	-9.24	**-64.8**	-61.8	-65.8
ML107							
M107-1	6	**-1.85**	-0.93	-3.90	**-29.2**	-24.2	-39.1
M107-2	6	**-3.19**	-2.47	-5.85	**-38.0**	-33.3	-57.3
M107-3	6	**-9.24**	-8.76	-9.58	**-65.1**	-63.9	-66.6
M107-4	6	**-9.34**	-9.15	-9.69	**-66.7**	-65.8	-68.2
MPL-1	3.2	**-5.38**	-4.37	-6.29	**-46.2**	-43.2	-50.9
MPL-11	5	**-8.47**	-7.63	-8.78	**-62.0**	-59.4	-64.0

Tab.03
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relevant δ_{GW} for water balance	lake Waldsee		lake Moritzteich		ML107	
	^{18}O	D	^{18}O	D	^{18}O	D
background value of non evaporation affected groundwater	-9.59	-66.6	-9.73	-68.2	-9.69	-68.2
lightest isotope value obtained in mixolimnion water	-7.97	-60.0	-7.59	-59.5	-4.16	-40.5
lightest isotope value obtained in monimolimnion water	-8.58	-66.9	-8.83	-73.3		

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fraction of inflowing water lost by evaporation	lake Waldsee		lake Moritzteich		ML107	
	^{18}O	D	^{18}O	D	^{18}O	D
27.06.06 - 31.07.06	-0.0758	-0.1618				
31.07.06 - 07.09.06	0.2093	0.2964				
07.09.06 - 12.10.06	0.0866	0.0625			0.5268	1.1683
12.10.06 - 16.11.06	0.0134	-0.0434	0.0123	-0.0676	0.0942	-0.0151
16.11.06 - 13.12.06	-0.3710	-0.2831	0.5664	3.0705	0.9333	11.2456
13.12.06 - 24.01.07	0.1080	0.0729	0.2232	0.2441	0.2471	0.1699
24.01.07 - 01.03.07	-0.1420	-0.1265	-4.9938	-0.6412	-7.5967	-0.7954
01.03.07 - 04.04.07	2.0730	-0.6597	0.6310	-32.9257	1.8855	-1.1360
04.04.07 - 10.05.07	0.0050	-0.0490	0.0889	0.0817	0.0971	0.1100
10.05.07 - 06.06.07	0.0217	-0.0534	0.0770	0.0315	0.1364	0.0896
06.06.07 - 04.07.07	0.0015	-0.0321	0.1103	0.1009	0.2105	0.2918
04.07.07 - 08.08.07	-0.0213	-0.0747	-0.0441	-0.0699	0.1882	0.2127
08.08.07 - 29.08.07	0.0840	0.0573	0.1132	0.1071	0.4094	0.4556
29.08.07 - 26.09.07	0.0320	-0.0291	0.1063	0.0445	0.5689	0.5320
26.09.07 - 24.10.07	1.2647	59.9832	0.4000	1.3781	-0.6484	-0.6054
24.10.07 - 21.11.07	-0.1237	-0.1367	-0.4710	-0.4722	-0.2204	-0.2907
21.11.07 - 19.12.07	0.2291	-0.5123	1.0381	-2.4906	-0.8917	-0.5516
19.12.07 - 15.01.08	-0.2217	-0.2287	1.0317	1.2136	-1.7802	-0.7297
15.01.08 - 12.02.08	0.0489	-0.0014	0.1437	0.1019	0.3331	0.4078
12.02.08 - 13.03.08	0.1252	0.2349	0.2480	0.4481	0.3527	0.8732
13.03.08 - 09.04.08	1.2157	-1.2166	0.6196	7.0004	0.3793	0.6914
09.04.08 - 06.05.08	0.1651	0.2624	0.2046	0.3870	0.6382	-3.4756
06.05.08 - 04.06.08	0.0459	0.0131	0.0471	0.0022	0.0732	0.0441
04.06.08 - 01.07.08	-0.0118	-0.1119	0.0221	-0.0755	0.0995	0.0329
01.07.08 - 29.07.08	0.0764	0.0468	0.1086	0.0585		
27.06.06 - 04.07.07	**0.1161**	**0.2306**				
04.07.07 - 01.07.08	**0.1372**	**0.4446**				
04.04.07 - 09.04.08			**0.1547**	**0.3721**		
05.06.07 - 04.06.08					**0.4327**	**-3.0150**

groundwater inflow rates I_{GW_MIX} and I_{GW_MION} applying different models and distinct isotope values for δ_{GW} [m^3/d]	lake Waldsee												lake Moritzteich												ML107					
	"background value approach"				"lightest mixol. sample approach"		"lightest monimol. sample approach"				I = (E/X) - P		"background value approach"				"lightest mixol. sample approach"		"lightest monimol. sample approach"				I = (E/X) - P		"background value approach"		"lightest mixol. sample approach"		I = (E/X) - P	
	^{18}O		D		^{18}O	D	^{18}O		D		^{18}O	D	^{18}O		D		^{18}O	D	^{18}O		D		^{18}O	D	^{18}O	D	^{18}O	D	^{18}O	D
	MIX	MON	MIX	MON	MIX		MIX	MON	MIX	MON	MIX		MIX	MON	MIX	MON	MIX		MIX	MON	MIX	MON	MIX		MIX		MIX		MIX	
27.06.06 - 31.07.06	63	-6	53	28	94	73	79	-16	52	27	-161	-76																		
31.07.06 - 07.09.06	-1	-24	-17	-53	-2	-23	-2	-58	-17	-51	23	12																		
07.09.06 - 12.10.06	5	-4	-6	39	7	-8	6	-9	-6	37	37	54													99	-358	240	-911	558	317
12.10.06 - 16.11.06	19	1	21	-37	29	29	25	2	20	-35	125	-53	1329	81	-2513	308	2252	-3659	1606	192	-2124	131	4790	-1307	445	358	1236	961	1881	-12645
16.11.06 - 13.12.06	-10	-1	-24	-10	-16	-35	-13	-4	-24	-10	-9	-10	-1703	-15	-641	850	-2906	-915	-2062	-40	-545	310	-92	-174	85	41	283	121	-79	3
13.12.06 - 24.01.07	-2	-7	-15	6	-3	-21	-3	-16	-14	6	6	16	-840	11	146	3718	-1407	210	-1011	30	124	929	-32	-56	92	-39	377	-134	161	610
24.01.07 - 01.03.07	-7	-4	-7	-33	-11	-11	-9	-8	-7	-31	-48	-51	-4475	-565	-2520	-9155	-7205	-3649	-5324	-1545	-2133	-3518	-666	-941	-138	-216	-793	-954	-247	-16
01.03.07 - 04.04.07	-11	-22	-5	-10	-20	-8	-16	-41	-5	-9	-5	-7	150	-1658	333	-515	236	485	177	-3512	282	-275	-114	-173	-78	-450	-865	-3110	-52	-78
04.04.07 - 10.05.07	44	19	38	-3	79	64	60	38	38	-3	1410	-147	4295	499	2432	606	6934	3577	5113	970	2048	322	5653	6164	620	71	5229	459	4756	4266
10.05.07 - 06.06.07	17	-19	36	17	28	54	22	-39	35	17	628	-294	-453	-777	-1282	-1012	-742	-1872	-542	-1496	-1083	-542	5760	14852	-94	-183	-392	-738	2320	4312
06.06.07 - 04.07.07	30	12	30	29	45	43	38	25	30	28	5899	-297	863	497	131	876	1388	188	1026	940	111	472	5073	5590	-65	-373	-197	-1146	1666	1439
04.07.07 - 08.08.07	6	-13	18	-73	8	26	7	-29	18	-71	-369	-115	1582	-864	-649	-2032	2562	-920	1885	-1583	-553	-1150	-7616	-4942	-11	-200	-28	-515	1864	1942
08.08.07 - 29.08.07	20	6	22	114	29	30	25	13	22	111	105	160	-1801	853	1404	1444	-2910	1977	-2145	1533	1200	845	3712	3941	-175	-387	-405	-920	470	646
29.08.07 - 26.09.07	42	3	47	-2	63	66	53	8	47	-2	332	-389	-62	-160	-218	-2977	-98	-306	-73	-296	-186	-1837	3485	8855	-205	-373	-481	-869	196	436
26.09.07 - 24.10.07	0	-7	-1	-40	1	-1	0	-17	-1	-39	-7	-10	2149	179	177	396	3451	249	2554	337	152	256	-76	-241	-400	-573	-1005	-1395	-424	-234
24.10.07 - 21.11.07	17	-3	17	-4	26	24	21	-7	17	-4	-29	-27	-4713	-1187	-147	-368	-7450	-207	-5574	-2191	-125	-233	-565	-565	-328	-407	-959	-1130	-541	-174
21.11.07 - 19.12.07	-12	0	-51	-1	-19	-76	-15	0	-50	-1	1	-15	459	344	51	-35	706	73	538	626	44	-22	-250	-339	-30	-156	-104	-495	-181	-45
19.12.07 - 15.01.08	10	0	15	17	17	23	14	0	15	16	-16	-16	-889	477	655	102	-1364	932	-1042	921	558	64	-15	-33	62	-27	254	-98	-130	-15
15.01.08 - 12.02.08	1	-6	4	-13	2	6	2	-12	4	-12	23	-1389	875	-635	222	1240	1341	316	1024	-1212	188	748	153	447	1	-143	6	-591	-139	164
12.02.08 - 13.03.08	1	15	1	86	2	2	1	33	1	82	7	-1	-831	-160	-248	4209	-1279	-354	-975	-292	-211	2144	160	-44	107	-98	763	-490	-30	73
13.03.08 - 09.04.08	-43	-26	-28	-84	-68	-42	-56	-55	-27	-79	-13	-16	-23	-485	13	-2236	-36	19	-27	-852	11	-1013	-236	-493	-191	-249	-1936	-1511	-148	78
09.04.08 - 06.05.08	28	-7	18	5	43	26	36	-13	17	5	13	5	952	-963	-2439	-2070	1467	-3504	1117	-1568	-2070	-1103	1291	527	-489	-1366	-6297	-10019	211	-92
06.05.08 - 04.06.08	20	25	24	57	31	35	25	49	23	54	190	671	2221	548	567	387	3443	802	2611	877	484	217	11784	252814	450	130	2838	681	6208	10449
04.06.08 - 01.07.08	19	18	22	74	28	32	24	42	22	68	-999	-113	1452	628	829	432	2265	1164	1710	1059	709	236	23939	-7216	415	284	1400	947	4841	15329
01.07.08 - 29.07.08													2228	665	-97	445	3525	-135	2636	1203	-83	236	4150	8128						
27.06.06 - 04.07.07	**15**	**-6**	**20**	**-5**	**22**	**29**	**18**	**-16**	**20**	**-5**	**38**	**14**																		
04.07.07 - 01.07.08	**8**	**1**	**11**	**7**	**12**	**15**	**10**	**2**	**11**	**7**	**28**	**1**																		
04.04.07 - 09.04.08													**432**	**52**	**386**	**60**	**688**	**562**	**512**	**57**	**326**	**56**	**1427**	**382**						
05.06.07 - 04.06.08																									**-103**	**256**	**-391**	**949**	**221**	**-66**

Table5
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Lebenslauf

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