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ORIGINAL ARTICLE

Relationship between ecological indicators (Arcellacea), total mercury concentrations and grain size in lakes within the Athabasca oil sands region, Alberta

L. A. Neville · R. T. Patterson · P. Gammon · A. L. Macumber

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Abstract Industrial mercury (Hg) sources associated with the processing of Athabasca oil sands (AOS), Alberta, Canada, may pose an environmental risk to nearby water bodies via either waterborne or airborne transport. Using a dataset derived from 63 lakes in the area, this study investigates the relationships between total-Hg (THg), organic matter, grain size, and lake ecology as measured by environmentally sensitive arcellacean (testate lobose amoebae) communities. The lakes studied include 59 lakes within a 75 km radius of the operations, plus four distal lakes ~150 km from the main industrial operations. Hg transport to the lakes is primarily through airborne pathways. The four distal lakes in the Peace–Athabasca Delta (~150 km downstream of the AOS operations) were examined to determine if the operation is emitting potential waterborne inputs, in addition to airborne inputs, and to identify any associated impact to those ecosystems. Total mercury in lakes close to the AOS were similar to values recorded in lakes farthest away. THg was most closely linked to the silt fraction, suggesting much of the Hg in these lakes is minerogenic in origin, either adsorbed and/or lattice-bound. THg is not statistically related to organic matter as has been observed in other Canadian lakes. The ecologic response to THg levels was investigated via the distribution of key indicator species and, or species diversity (Shannon diversity index). The spatial extent of arcellacean ecosystem stress in the study lakes did not correlate with THg concentrations. This is perhaps due to the generally low THg levels found in these lakes, all except one had THg concentrations lower than current CCME guidelines. While these findings may rule out any direct link between THg concentrations in the lakes and observed Arcellacea faunas, ecosystem stress unrelated to THg was observed northeast of the AOS, which warrants further examination. The results of this research suggest that the natural lake arcellacean faunas in the region are not being significantly impacted by current THg concentrations.

Keywords Athabasca · Oil sands · Mercury · Contamination · Arcellacea · Testate lobose amoebae · Grain size

L. A. Neville (✉) · R. T. Patterson · A. L. Macumber
Ottawa-Carleton Geoscience Centre and Department of Earth Sciences, Carleton University, Ottawa, ON K1S 5B6, Canada
e-mail: lisaneville@cmail.carleton.ca

P. Gammon
Geological Survey of Canada, Ottawa, ON K1A 0E4, Canada

Introduction

The Athabasca oil sands (AOS) represent one of Canada's most economically important natural resources (Alberta Energy 2008). The process of upgrading bitumen to synthetic crude oil involves coking, coke combustion, and production of wastes and fly ash (Jang and Etsell 2006; Allen 2008). Environment Canada's National Pollutant Release Inventory (NPRI 2010) suggests that upgrading is a substantial and increasing source of priority air pollutants. Consequently, the transport of priority pollutants such as Hg from oil sands industrial processes and their ecological impacts have been extensively researched (Hazewinkel et al. 2008; Kelly et al. 2009, 2010; Curtis et al. 2010; Kurek et al. 2013), and with regulatory focus [e.g., Regional Aquatics Monitoring Program (RAMP 2012); Cumulative Environmental Monitoring Association (CEMA 2012)].

Mercury (Hg) is a trace element that can be toxic to aquatic biota at elevated concentrations (CEPA 1985). Airborne Hg emissions from oil sands industrial sources in 2010 were approximately 120 kg, which represents a six-fold increase from 2002 (NPRI 2010). This is consistent with the findings of Kelly et al. (2010) who reported elevated concentrations of Hg in the snow packs and waters surrounding, and downstream of the oil sands operations. However, results of a recent lake sediment core study in the region concluded that current Hg levels are above background levels but have been decreasing due to improved industrial practices (Wiklund et al. 2012).

This study investigates THg concentrations in the surface sediments of 63 lakes encompassing a broad geographic radius around the oil sands operations. Hg commonly enters a lake via surface run-off, and then generally is quickly transported to and deposited with the lake bottom sediments. Lake sediments are an important component of aquatic ecosystems, providing habitat for a wide range of benthic and epibenthic organisms. Exposure to deleterious contaminants in sediments may represent a potential hazard to the health of these organisms, and this harm may be propagated throughout the ecosystem (Luoma and Carter 1993; Mason et al. 1995; CCME 2001). Effective assessment of this hazard requires that a causal link be established between the distribution of substances of environmental concern in the sediments and the occurrence of adverse biological effects (Environment Canada 1997). The objective of this research is to delineate the spatial distribution of THg in sediment–water interface sediments of lakes to determine if they are: (1) primarily influenced by atmospheric deposition, and/or (2) receiving input from snowmelt, streams or rivers and related tributaries downstream of the oil sands operations; and (3) make an assessment of any THg-induced ecological pressure on lacustrine benthic fauna using environmentally sensitive Arcellacea as a proxy.

Ecological indicators

Arcellacea, informally known as thecamoebians (Patterson and Kumar 2002) or testate lobose amoebae (Mitchell et al. 2008), are protists that constitute an important component of the benthic community within the microbial trophic level in lakes and wetlands (Patterson and Kumar 2000; Beyens and Meisterfeld 2001; Elliott et al. 2012). Species and strains are characterized by a simple sac-like, decay-resistant organic test of pseudo-chitinous material that is variably agglutinated in different species (Scott et al. 2001; Patterson and Kumar 2002). Arcellaceans are useful in environmental research as they are characterized by rapid generation times and sensitivity to environmental conditions at the sediment/water interface and epibenthic zone (Collins et al. 1990; Neville et al. 2010a). Their abundant fossilized remains preserve a record of pollutant responses and changing environmental conditions over time (Patterson 1987; McCarthy et al. 1995; Asioli et al. 1996; Patterson et al. 2002; Boudreau et al. 2005; Patterson et al. 2012a; Watchorn et al. 2012). Unlike most microfossil groups, arcellaceans are resistant to dissolution in lower pH environments (Swindles et al. 2007) and are one of the few microfossils that can be used to investigate depositional conditions at the sediment/water interface of lacustrine freshwater environments (Patterson et al. 1985; Roe et al. 2010).

Variations in arcellacean community assemblages have been successfully used to investigate the impact of mine tailing deposition (Patterson et al. 1996; Reinhardt et al. 1998; Kauppila et al. 2006; Kihlmanm and Kauppila 2009, Kihlman and Kaupila 2010, 2012), remediation in oil sands tailings ponds (Neville et al. 2011), and identification of lake pH (Reinhardt et al. 1998; Kumar and Patterson 2000; Escobar et al. 2008; Patterson et al. 2012b). Patterson et al. (1996) and Reinhardt et al. (1998) have demonstrated that arcellacean assemblages and species diversity are sensitive to, and can be significantly impacted by, heavy metal contamination, specifically Hg and arsenic.

Study area

Sixty-three freshwater lakes from two major drainage basins (the Athabasca and the Peace/Slave River Basins) in northern Alberta were sampled for this study (Fig. 1). The majority of these lakes are situated within a 75 km radius of Fort Mackay, the center of major AOS surface mining and upgrading (Kelly et al. 2010). Fifty-seven lakes were selected to potentially capture the spatial distribution of atmospheric borne emissions. These lakes are not directly connected to any drainage network that passes through the industrial areas, and consequently they are considered to only receive industrial emissions via atmospheric transport. Six additional lakes were selected along a 150 km transect on the Athabasca river, located directly upstream ($n = 1$), within the AOS mining operation ($n = 1$) and downstream ($n = 4$) of the mining operation as far as the Peace–Athabasca Delta (PAD). These lakes are partially linked to the oil sands through permanent or intermittent tributaries, or potentially influenced by flooding events. These lakes may therefore contain both waterborne and airborne emissions derived from oil sands mining operations (Fig. 1).

The sampled lakes comprise sites that are distal and proximal; upwind and downwind; upstream and downstream from the AOS mining operations (Fig. 1). The prevailing wind direction in the region is variable and may come from

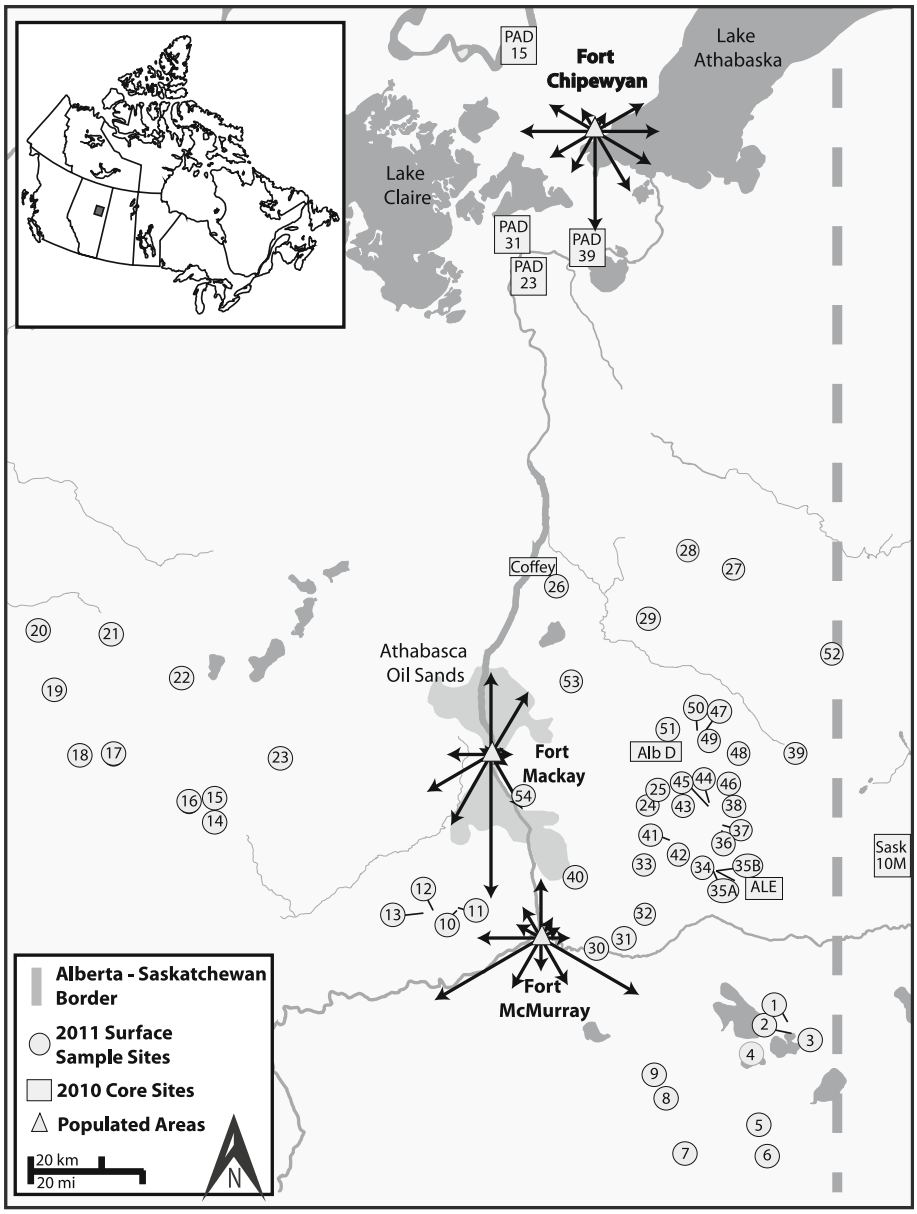

Fig. 1 Map of study area showing study lakes in relation to the Alberta's Athabasca oil sands operations and *arrows* indicating prevailing wind directions at different times of the year

the north, west, northeast and northwest depending on the time of the year (Environment Canada 2003) (Fig. 1). Bedrock in Alberta is primarily sedimentary and consists of sandstone, siltstone, shale, coal, and limestone, some of which is bituminous. The till veneer across much of the area is mainly comprised the same components (Andriashek et al. 2002). Most of the studied lakes were formed by retreating glaciers and till piles, where holes or kettles were left in the landscape due to melting ice (Mitchell and Prepas 1990). Notable exceptions are the northern lakes located in the PAD, which are part of a large sandy braided delta system (Hugenholtz et al. 2009). In all lakes in the PAD region there is potential for oil sands derived material to have been introduced by natural means, i.e., via local erosion and/or dissolution of tills and/or bedrock.

Methodology

Sediment and water collection

Selection criterion for the lakes included: accessibility, size, depth and substrate type. Ideal lakes had water depths of less than 8 m to reduce the potential for sampling sediments exposed to bottom water anoxia during summer or winter water column stratification, but greater than 2 m so as to avoid disruption to the sediment bottom as a result of freezing during the winter. Bottom water anoxia precludes arcellacean colonization because these benthic organisms require oxygenated conditions. Sites further than 3 m away from the shoreline were targeted to reduce shore effects (e.g., erosion) on arcellacean assemblages.

Table 1 Site names and coordinates of lakes investigated, water quality data measured during sample collection, sediment total bulk concentrations of Hg (dry weight, mg kg^{-1}), Rock–Eval® data, sediment grain size fractions, and species diversity index (SDI)

Site names	Latitude (°N)	Longitude (°W)	Temperature (°C)	Conductivity (ms/cm)	THg (ppb)	S1 (mg HC/g[a])	S2 (mg HC/g[a])	TOC[b] (%)	RC[c] (%)	MINC[d] (%)	Sand (%)	Silt (%)	Clay (%)	SDI
Sask 10 M	56 55 15.1	109 43 53.5	9.5	94.6	68	52.80	104.62	28.44	13.91	1.0	6.3	86.6	7.2	2.17
Alb D	57 04 00.1	110 50 44.8	10.4	94.1	98	21.84	92.07	24.03	13.04	1.5	4.6	90.8	4.7	2.53
ALE	56 52 42.0	110 34 5.3	8.6	111.5	210	22.16	85.21	24.48	14.13	1.5	6.0	84.6	9.4	2.04
Coffey	57 38 37.2	111 25 20.3	9.5	195	117	2.22	16.82	11.02	8.38	1.93	0.5	88.8	10.7	1.74
PAD 31	58 29 43.8	111 31 19.2	7.4	171	62	1.56	14.11	5.25	3.61	1.2	0.6	81.7	17.6	2.53
PAD 39	58 27 56.8	111 11 02.7	7.6	146.5	74	6.11	36.72	10.20	6.02	2.8	3.4	82.4	14.2	2.10
PAD 23	58 23 28.3	111 26 37.7	7.9	92	55	19.24	75.73	24.33	15.03	1.79	0.0	79.1	20.9	1.61
PAD 15	58 57 00.0	111 29 35.2	8.5	159	75	0.33	2.39	2.92	2.37	1.13	0.0	74.3	25.7	1.76
1	56 29 09.08	110 14 01.0	15.4	160	76	29.54	106.86	25.20	12.45	4.1	51.9	43.4	4.7	1.91
2	56 26 51.01	110 12 12.02	16.3	213.5	41	43.68	152.66	33.77	15.86	2.2	18.3	71.8	9.9	1.78
3	56 26 01.05	110 07 24.09	15.9	168.6	67	33.55	144.46	36.40	19.64	2.0	20.9	70.5	8.5	2.06
4	56 23 14.5	110 24 05.09	16.2	229.1	64	24.14	126.14	24.14	10.56	1.3	23.7	65.2	11.2	2.01
5	56 12 12.07	110 21 50.04	16.3	134.7	108	18.34	153.42	33.21	17.30	1.8	4.5	78.8	16.7	1.31
6	56 07 56.00	110 19 50.09	14.9	32.6	142	21.56	126.71	39.48	24.92	2.4	6.9	80.9	12.2	1.58
7	56 08 06.08	110 42 35.02	15.9	121.6	106	29.30	222.50	44.27	21.32	2.0	25.6	70.4	4.0	1.52
8	56 16 42.09	110 47 53.01	17.2	269.3	23	0.59	3.59	1.60	1.11	0.5	92.1	6.0	1.9	2.04
9	56 18 33. 05	110 51 21.08	17.8	201.1	45	36.87	159.83	36.44	18.36	1.8	10.2	78.9	10.9	1.83
10	56 46 32.08	111 47 24.04	16	134	113	29.82	159.66	38.50	20.84	1.8	19.7	74.0	6.3	1.73
11	56 46 58.04	111 47 21.05	16.4	75	95	28.98	170.19	41.33	22.78	1.8	16.7	79.0	4.3	1.41
12	56 46 30.09	111 54 51.07	16.4	99.2	109	32.37	138.07	34.75	18.79	1.5	7.1	81.3	11.6	2.04
13	56 46 14.09	111 56 41.09	15.8	69.7	67	34.94	163.22	41.06	22.56	1.8	11.6	72.5	15.8	1.60
14	57 00 16.00	112 56 23.04	15	95	94	20.57	122.47	33.75	20.07	1.9	10.0	76.2	13.8	2.00
15	57 01 57.07	112 57 06.05	16.1	253.5	75	22.30	153.69	27.84	12.02	1.5	16.3	70.6	13.1	2.31
16	57 03 09.00	113 03 22.01	15.9	84.3	110	26.99	232.86	40.77	17.54	1.3	6.4	84.1	9.5	2.05
17	57 10 30.06	113 24 54.04	14.4	57.9	109	8.82	54.12	13.66	7.70	0.9	83.5	12.8	3.7	2.15
18	57 11 18.07	113 34 20.02	14.5	53	102	16.49	77.42	19.33	10.46	1.1	8.6	78.6	12.8	2.22
19	57 20 40.04	113 41 35.07	14.6	97	105	17.14	105.27	27.90	16.25	1.6	6.6	78.4	15.1	1.82
20	57 29 19.04	113 45 58.01	14	112	103	27.28	111.02	29.23	16.20	1.5	12.0	77.4	10.6	1.64
21	57 28 53.02	113 25 26.07	14.4	141.8	119	18.29	72.34	23.97	15.10	1.2	12.4	80.2	7.4	1.46
22	57 22 05.02	113 05 28.04	15.1	48.5	135	15.18	97.95	24.74	14.03	1.4	11.6	74.1	14.3	2.18
23	57 10 09.07	112 37 38.01	15.8	119.1	82	25.98	163.30	34.01	16.87	1.2	5.2	78.8	16.0	0.89
24	57 02 39.08	110 53 20.00	16.1	55.7	127	26.95	149.61	32.02	15.77	1.4	18.5	76.0	5.5	1.44
25	57 04 47.00	110 49 45.00	16.2	163.6	108	22.71	115.01	26.87	14.05	1.3	17.6	75.8	6.6	1.56
26	57 36 05.05	110.39 21.03	16.6	227.8	92	48.25	128.27	32.70	16.29	1.3	23.6	72.0	4.4	1.32
27	57 38 53.2	110 28 18.0	16.7	124.1	77	54.46	118.12	30.42	14.38	1.3	25.6	70.3	4.1	1.17
28	57 41 21.08	110 42 06.04	17.1	256.8	61	46.43	167.10	36.55	17.13	1.2	21.5	73.5	5.0	0.95

Table 1 continued

Site names	Latitude (°N)	Longitude (°W)	Temperature (°C)	Conductivity (ms/cm)	THg (ppb)	S1 (mg HC/g[a])	S2 (mg HC/g[a])	TOC[b] (%)	RC[c] (%)	MINC[d] (%)	Sand (%)	Silt (%)	Clay (%)	SDI
29	57 31 09.09	110 53 08.00	17.1	118	76	39.00	174.85	40.70	21.00	1.5	11.7	85.5	2.8	1.16
30	56 40 14.05	111 08 17.03	13.5	404.7	29	19.11	68.14	15.47	7.44	7.2	42.8	51.5	5.7	1.73
31	56 41 39.05	111 00 08.08	17.8	34.2	63	30.34	131.36	28.52	13.80	1.1	12.8	71.5	15.8	1.51
32	56 45 57.07	110 54 42.08	16.6	116	76	34.64	169.86	40.28	21.39	1.5	14.8	74.9	10.3	1.86
33	56 53 35.01	110 54 05.05	16.6	42.6	120	34.13	170.83	38.76	19.83	1.4	11.7	80.9	7.4	1.64
34	56 52 53.04	110 37 28.02	17.2	192.1	87	26.42	123.05	34.32	20.14	1.7	8.2	77.0	14.8	1.70
35A	56 52 42.09	110 34 24.06	16.4	137	93	19.39	93.49	26.05	15.28	1.4	20.2	65.8	14.0	1.48
35B	56 52 49.06	110 33 14.07	16.9	138.6	12	1.54	5.20	2.17	1.44	0.3	95.1	3.8	1.1	1.45
36	56 58 57.01	110 32 37.01	15.6	60.9	134	5.28	31.91	11.39	7.59	0.9	38.4	54.2	7.4	1.32
37	56 59 43.03	110 31 51.07	15.8	158.3	96	18.72	106.49	35.51	23.04	2.3	13.7	77.6	8.6	0.80
38	57 02 24.03	110 29 09.02	15.9	74.2	101	11.64	68.36	20.07	12.25	1.3	13.6	74.2	12.2	2.18
39	57 10 30.08	110 11 15.05	17	136.9	58	54.07	158.39	35.00	15.81	1.2	36.2	60.1	3.7	1.28
40	56 51 14.03	111 13 55.08	17.6	103.1	76	35.76	172.01	43.17	23.98	1.5	72.6	26.0	1.3	1.75
41	56 57 30.04	110 46 39.05	15.6	75.5	58	33.87	189.45	43.70	23.23	1.5	41.6	54.5	3.8	2.10
42	56 57 07.04	110 45 12.06	15.9	77.7	84	30.68	159.06	40.71	23.03	1.7	15.7	75.3	9.1	1.85
43	57 01 59.01	110 43 03.03	15.8	70.4	157	12.69	64.39	23.40	15.52	1.6	15.2	76.3	8.5	2.21
44	57 03 07.06	110 36 12.02	14.8	31.4	130	23.41	146.57	32.13	16.46	1.4	12.6	77.5	9.9	2.16
45	57 02 44.05	110 35 40.01	14.7	37.1	127	26.22	159.18	34.89	17.88	1.6	16.2	76.5	7.3	0.97
46	57 04 44.05	110 30 37.05	15.1	43.1	151	13.53	72.20	21.46	13.10	1.3	8.4	76.3	15.3	2.14
47	57 09 07.01	110 31 04.01	14.9	158.1	62	27.07	117.61	27.39	13.99	3.2	17.7	74.0	8.3	1.56
48	57 10 18.08	110 27 23.00	15.3	124.9	120	20.00	136.56	31.44	16.85	1.7	10.8	77.5	11.8	1.88
49	57 12 48.02	110 35 33.08	15.1	102.4	93	16.99	117.56	30.67	17.73	2.1	8.6	82.6	8.8	2.01
50	57 14 19.02	110 39 02.08	15	63.9	113	18.02	103.84	33.34	21.02	2.3	8.9	78.5	12.5	1.98
51	57 14 08.02	110 47 42.05	14.1	133.3	86	10.23	70.20	25.03	16.55	1.9	4.5	78.2	17.3	1.42
52	57 25 46.03	110 00 35.02	16	248.1	68	31.32	142.97	38.25	21.90	1.9	21.5	71.1	7.5	1.81
53	57 21 39.02	111 14 48.01	16.5	314.2	30	19.65	94.20	20.65	10.18	6.6	36.1	57.5	6.4	0.70
54	57 04 16.04	111 28 33.01	17.4	321.3	18	15.47	43.41	9.83	4.32	8.6	40.9	51.2	7.9	1.65
Median			15.8	119.1	92	22.71	118.12	30.42	15.86	1.51	13.6	76.0	9.1	1.75
Mean			15.0	133.0	89.4	24.1	114.5	28.2	15.3	1.9	20.0	70.3	9.7	1.7

[a] mg hydrocarbons/g of dry sediment

[b] Total organic carbon

[c] Residual carbon

[d] Mineral content

Sediment–water interface samples were collected from eight lakes in August 2010 and 55 lakes in August 2011 (Table 1; Fig. 1). Sample collection took place in two parts to meet the needs of the multidisciplinary CORES program. The 2010 sample set was collected from a raft using a Glew gravity corer (Glew et al. 2001). These cores were subsampled at 0.5 cm intervals using an extraction device (Glew et al. 2001) and stored in a cool room at Carleton University at 4 °C. The top 0.5 cm of the Glew core was sampled for analysis in this study. The 2011 sample set was collected from a float helicopter using an Ekman grab sampler, and again only the top ~ 0.5 cm was collected for analysis. Site 35 was comprised of two large bodies of water connected by a stream. Both bodies of water were sampled and labeled 35A and 35B. Only grab and core samples that presented a well-preserved sediment–water interface were sampled.

Surface water samples were also collected at each site. Water property data was recorded at 50 cm water depth increments (Table 1) at each core location using a YSI multimeter equipped with thermodynamic probes.

Mercury analysis

The total bulk concentration of Hg (THg) in sediments was determined using a Leco AMA254 Advanced Hg Analyzer at the Geological Survey of Canada, Halifax (Leco Corporation 2012). Sample preparation included freeze-drying followed by powdering using an agate mortar and pestle. Prior to sediment analysis a clean sample and five blanks were run to condition the instrument. Analysis of two reference samples was subsequently carried out to derive a calibration curve against which the lake sediment analyses were compared. The average reproducibility was better than 2 % using $n = 12$ duplicate analyses.

Analysis of organic matter (Rock–Eval® Analysis)

The bulk quantity of total organic carbon (TOC) in the sediment was determined using Rock–Eval® 6 Analyses (Vinci Technologies, Rueil- Malmaison, France) at the Geological Survey of Canada, Calgary. The standard reference materials used for this method included IFP 160000, Institut Français du Pétrole and internal 9107 Shale standard. The analytical reproducibility based on measurements of series duplicates was generally better than 5 %.

Grain size analysis

Sediment samples were digested in a heated bath (~50 °C) with 10 % H_2O_2 to remove organics (van Hengstum et al. 2007; Murray 2002; Donato 2009). Digested samples were then analyzed using a Beckman Coulter LS 13 320 laser diffraction analyzer fitted with a universal liquid medium (ULM) sample chamber over a measurement range between 0.4 and 2,000 µm. The samples were loaded into the machine until an obscuration level of 10 ± 3 % was attained. The Fraunhofer diffraction model was used to estimate grain sizes.

Arcellacea analysis

Arcellaceans were separated from 2 cc of sediment from each sample by seiving to remove the >250 and <37 µm fractions. The 37–250 µm fraction was subdivided into aliquots for quantitative analysis using a wet splitter (Scott and Hermelin 1993). The wet aliquots were subsequently examined under an Olympus SZH10 dissecting binocular microscope (40–80× magnification) until a statistically significant number of specimens were quantified (Patterson and Fishbein 1989). In most cases >150 ($n = 22$), and more where possible >300 ($n = 35$) arcellaceans were counted per sample. Identification of arcellaceans followed standard reference keys (e.g., Medioli and Scott 1983; Kumar and Dalby 1998). Scanning electron micrograph images of common species and strains were obtained using a Tescan Vega-II XMU VP scanning electron microscope at the Carleton University SEM facility (Fig. 2).

Species diversity was calculated using the Shannon diversity index (SDI) (Shannon 1948). SDI is a measure of faunal diversity and is useful for determining the relative health of the community from which the sample was taken (Patterson and Kumar 2002). The SDI is calculated using the following formula:

$$\mathrm{SDI} = -\sum_{I}^{S}\left(\frac{X_i}{N_i}\right)\times\ln\left(\frac{X_i}{N_i}\right)$$

where X_i is the abundance of each taxon in a sample, N_i is the total abundance of the sample, and S is equal to the species richness of the sample. Generally harsh environmental conditions are normally characterized with an SDI between 0.3 and 1.5, transitional/intermediate conditions range from 1.5 to 2.5 and favorable stable conditions have an SDI >2.5 (Magurran 1988; Patterson et al. 2002).

Correlation analysis

Correlation analysis was conducted on temperature, conductivity, THg, organic matter parameters [S1, S2, total organic carbon (TOC), residual carbon (RC)], % mineral concentration, % water, % sand, % silt, % clay and SDI. The Pearson product-moment correlation coefficient (Pearson's r) is a measure of the linear dependence (correlation) between two variables (Rodgers and Nicewander 1988).

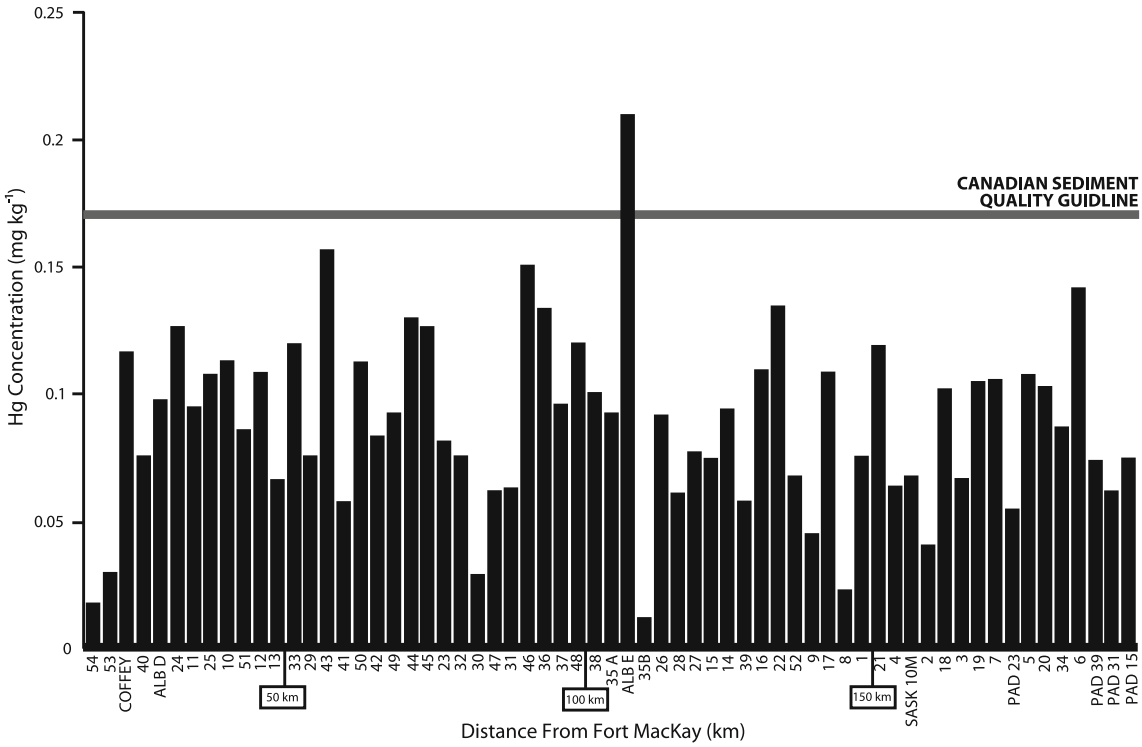

Fig. 2 Histogram depicting the total concentrations of Hg (mg kg^{-1}) in study sediments versus distance of sites from Fort McKay relative to the Canadian sediment quality guidelines (CCME 2002) and concentrations in sediment from Central Alberta

Spatial interpolation

Spatial interpolation was depicted using a conventional inverse distance weighting technique incorporated in the geostatistical analysis component of the ArcGIS software (Zimmerman et al. 1999).

Results and discussion

Total mercury concentrations in study lakes

Total mercury concentrations in surface sediments from the 63 lakes studied ranged from 0.018 to 0.210 mg kg^{-1} (average 0.089 mg kg^{-1}) (Table 1). Only lake "Alb E" with a THg concentration of 0.21 mg kg^{-1} (Table 1; Fig. 2) had a concentration higher than the Canadian Sediment Quality Guidelines for the Protection of Aquatic Life (CCME 2002) of 0.17 mg kg^{-1}, albeit by a methodology that can report higher Hg concentrations than the methodology within the CCME guidelines (Table 2). The relative distance of this sample from the oil sands facilities cannot explain the observed higher concentrations of THg in Alb E. Generally, atmospheric deposition should generate higher concentrations in close proximity to a pollutant point source followed by declining concentrations with increasing distance from the emission source (a "bull's-eye" spatial pattern; Kelepertsis et al. 2006; Fältmarsch et al. 2007). An expanded geochemical study of these lakes may produce chemical data for normalizing THg concentrations, which could reduce the influence of limnological and/or geogenic variability between lakes and provide a more critical interpretation of

Table 2 Comparison of values produced using different mercury analysis methodologies

Study	Method used	Comparison to CCME guidelines
CCME Guidelines (2002)	Strong acid digestion	–
This study	Thermal decomposition	Can overestimate (Hg) in comparison to CCME
Sanei et al. (2010)	Aqua regia	Similar to guidelines
Friske and Hornbrook (1991)	Aqua regia	Similar to guidelines
Environment Canada (1997)	Aqua regia	Similar to guidelines
Outridge et al. (2011)	Thermal decomposition	Can overestimate (Hg) in comparison to CCME

All values are compared to the values produced using the Canadian Sediment Quality Guidelines for the Protection of Aquatic Life (CCME) methodology. Bulk Hg concentration analytical methods produce estimates equivalent to or higher than those recommended by the CCME guidelines (López-Antón et al. 2012)

the spatial Hg distribution in relation to the oil sands operation.

Correlation analysis using Pearson's r was conducted on the available data in order to minimize the signal to noise ratio associated with lake variability. The strongest correlation occurred between THg and silt ($r = 0.44$; $n = 63$, Pearson) fractions in sediments suggesting that THg concentrations are at least partially influenced by grain size. The correlation may imply that the processes impacting THg in the regions' lakes are: (1) adsorption on to silt-sized particulates, and/or; (2) the silt-sized fraction contains minerals with high Hg concentrations, and/or; (3) the sedimentary flux of these particulates varies across the region. The correlation analysis ruled out clays as the driver of THg sequestration in these lakes since the clay-sized fraction has a very weak correlation of ($r = 0.13$; $n = 63$, Pearson). In order to partially remove the influence of mineral matrix and grain size variability, THg concentrations were normalized to the silt fraction. Such a normalization procedure will minimize the bias due to silt concentration variability between lakes (cf. Covelli and Fountolan 1997; Kersten and Smedes 2002).

Previous research suggests that organic matter (OM) is important in controlling the distribution of mercury and other trace elements in soil and suspended or bottom aquatic sediment (Gibbs 1973; Orem et al. 1986; Chin and Gschwend 1991; Johansson and Iverfeldt 1994; Kainz et al. 2003; Mirlean et al. 2003). Typically it is understood that the enrichment of Hg in some sediment may be linked to the degree of a lake's OM production/input (e.g., OM scavenging of the atmospheric Hg input, Outridge et al. 2007). However, in this study the correlation between OM and Hg is low ($r = 0.19$; $n = 63$; Pearson) suggesting that OM is not controlling Hg distribution and does not significantly explain the observed Hg distribution pattern. Further research is required to explain this result.

A recent winter snow pack and summer water Hg survey in the region classified sites greater than 50 km away from the operation as being characterized by background Hg levels and showed that values increased with proximity to the operation (Kelly et al. 2010). In comparison to the study by Kelly et al. (2010), this study encompasses a larger geographic radius and includes a variety of sites up to 150 km away. Sites for the Kelly et al. (2010) study were identified as "background" or "near development" based upon the shape of the concentration-distance from development curve. Kelly et al. (2010) noted that none of the THg values downstream of the operation were significantly greater than those upstream but they also concluded that Hg values in melted snow, tributaries and in the Athabasca River exceeded the CCME (2002) guidelines. However, comparison of their filtered versus unfiltered Hg analyses suggest particulate material of unidentified form

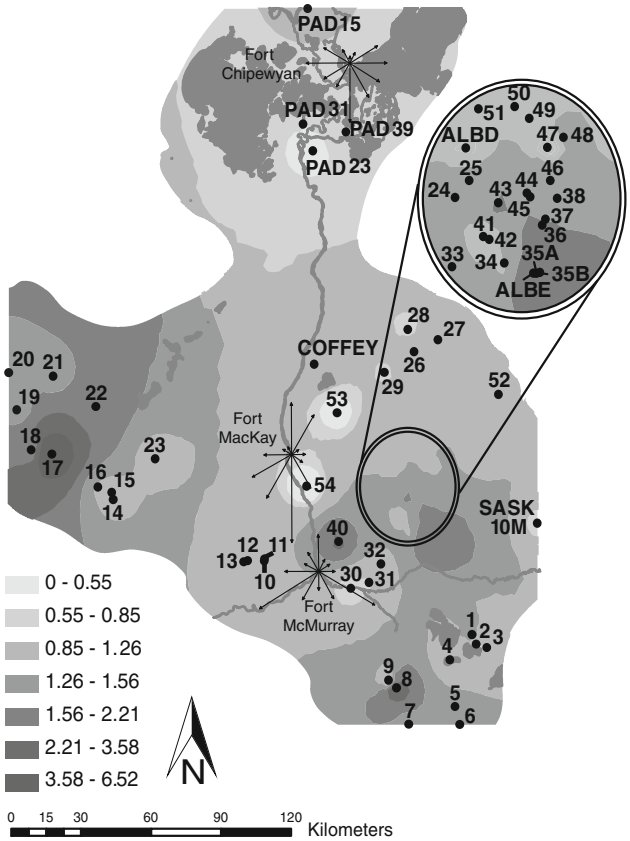

Fig. 3 Spatial interpolation (ArcGIS inverse distance weighting) of mercury concentrations normalized to silt in lakes surrounding the Athabasca oil sands operation

carries the Hg that produces a "bull's-eye" pattern around the oil sands operations. The dissolved Hg within the snowpack and water samples are below CCME (2002) guidelines and do not form a "bull's-eye" pattern and is interpreted by Kelly et al. (2010) to be mostly of local origin. Our samples produce a similar pattern (Fig. 3) to that of the dissolved Hg data of Kelly et al. (2010). The ecological importance or otherwise of the particulate-bound Hg within the snowpack has not been evaluated to date, which will be determined by the type of particulate material and how well it binds the Hg. Nevertheless, the thermal decomposition Hg analytical method used herein would dissolve all of this particulate material were it to enter the lake system. The lack of evidence for a similar Hg pattern to that reported by Kelly et al. (2010) suggests the particulate material is not yet a significant contributor to the lake Hg budgets. Unfortunately the unavailability of mass accumulation rates for the lakes studied hinders any further comparison.

The thermal decomposition THg analytical method used herein can produce higher Hg concentrations than obtained with a strong-acid digestion procedure (Table 2 and contained references). Despite using the thermal

decomposition THg analytical method the Hg values obtained here for the lake sediments surrounding the oil sands operations are within CCME (2002) guidelines and comparable to acid digested samples observed in the vicinity of coal-fired power plants in central Alberta (0.058–0.110 mg kg^{-1}, average 0.083 mg kg^{-1}; Sanei et al. 2010). In the National Geochemical Reconnaissance (NGR) program database of the Geological Survey of Canada (GSC), the mean background concentrations in Canadian lake and stream sediments are essentially identical at 0.074 and 0.075 mg kg^{-1}, respectively (measured using an aqua regia digestion; Friske and Hornbrook 1991). However, Hg concentrations as high as 15.03 mg kg^{-1} have been reported in the Great Lakes and of 0.987 mg kg^{-1} in the Toronto Harbor, Ontario (Environment Canada 1997).

Lake ecological health associated with THg distribution

Thirty arcellacean species and strains were identified from 63 sediment–water interface samples. The arcellacean SDI range was 0.70–2.53 (average 1.73; Table 1). These values are typical of lacustrine transitional/intermediate environmental conditions, indicating a relatively healthy lake environment (Patterson and Kumar 2000; Kumar and Patterson 2000). A study by Neville et al. (2010b) of arcellacean communities in lakes across vegetation zones across Alberta found SDI values from 1.35 to 2.17 (average 1.65); with the average SDI value for lakes in the boreal vegetation zone being 1.83, a value similar to the average SDI value of this study. The compositions of the arcellacean faunas observed are variably composed of *Centropyxis aculeata* (Ehrenberg 1832) strain "aculeata", *Centropyxis aculeata* strain "discoides", *Centropyxis constricta* (Ehrenberg 1843) strain "aerophila", *Difflugia oblonga* (Ehrenberg 1832) strain "oblonga", *Difflugia oblonga* strain "tenuis", and *Difflugia protaeiformis* (Ehrenberg 1830) strain "acuminata". A few of the lower SDI sites such as lake 53 are dominated by Centropyxid Arcellacea, but the majority of lakes contain mixed faunas of the tolerant and the sensitive species.

Inverse distance weighting of the arcellacean assemblages recovered from the lakes demonstrates that the highest diversity sites are located to the north in the Peace–Athabasca Delta, and to the west and southeast of the oil sands operation. Six sites characterized by lower SDI values (0.70–1.32; average 1.03) were observed to the northeast and west of the oil sands operations (Fig. 4), and warrant further investigation in order to determine the cause of the observed ecological stress. For the most part though the SDI versus THg values found in this study showed no significant correlation ($r = 0.00161$; $n = 63$, Pearson; Fig. 4), and the spatial distribution of increased

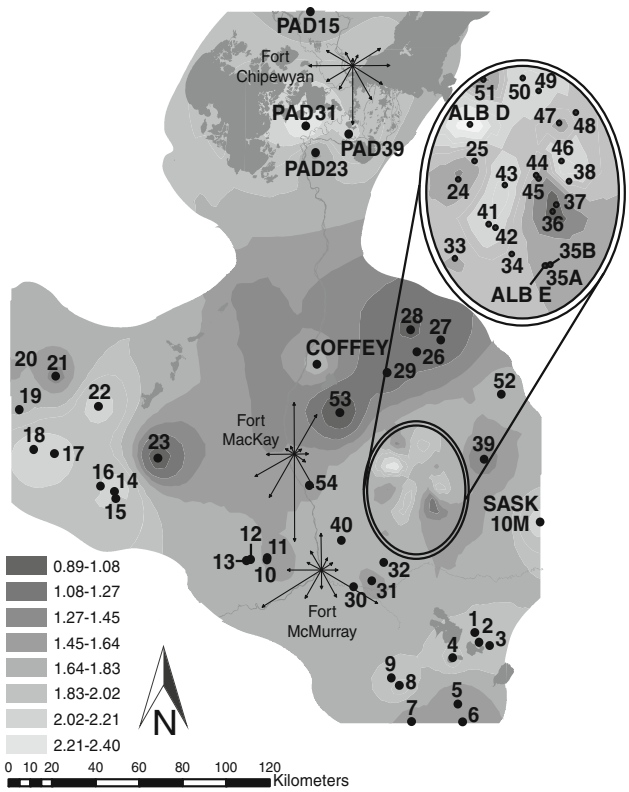

Fig. 4 Spatial interpolation (ArcGIS inverse distance weighting) of the Arcellacean species diversity index in lakes surrounding the Athabasca oil sands operation

THg values did not correlate with the centropyxid/difflugiid-type arcellacean ratio, indicating that THg concentrations in the studied lakes are not controlling species diversity and therefore are not acting as a benthic arcellacean ecological stressor.

Factors controlling lake ecology in the oil sands development area are clearly complex and influenced by a number of variables other than THg concentrations at the sediment–water interface. Previous studies utilizing arcellaceans as indicators of Hg contamination have found that SDI values are a good indicator of an ecosystem's response to Hg concentrations when Hg values reach a concentration threshold and become an environmental stressor (Reinhardt et al. 1998). In addition, studies by Patterson et al. (1996) and Reinhardt et al. (1998) found that centropyxid-type arcellaceans can tolerate higher levels of Hg in comparison to the difflugiid-type arcellaceans. Similarly, Neville et al. (2011) found that centropyxids could also be used to indicate ecosystem health and remediation efforts in oil sands tailings ponds. A correlation between arcellacean assemblage makeup (centropyxid/difflugiid-type arcellacean ratio) and SDI would therefore be expected in the current study if Hg were an environmental stressor in these lakes.

Conclusions

This study investigated THg concentrations and Hg input pathways both airborne and waterborne in lakes surrounding the Athabasca Oil Sands in northeastern Alberta, Canada. Additionally, the relationships between THg, organic matter, grain size, and lake ecology were also assessed.

Total mercury concentrations in surface sediments from the lakes studied were generally low, THg levels found in all but one of these lakes were below current CCME guidelines and similar to concentration in lakes in the vicinity of coal-fired plants in central Alberta. THg concentrations in lakes did not show an airborne or waterborne pattern of deposition, as values in lakes close to the operation were similar to values recorded in lakes farthest from the operation. THg was not statistically associated with organic matter as has been observed in other Canadian lakes but was most closely linked to the silt fraction within the grain size population.

Lake ecology was measured by environmentally sensitive Arcellacea (testate lobose amoebae) communities and suggested no correlation between THg concentrations, arcellacean SDI and assemblage makeup. A few sites characterized by stressed faunas were observed but they did not overlap with areas of enriched THg. Average SDI values observed in the vicinity of the oil sands operation are similar to values from lakes across Alberta. While these findings indicate THg is not an arcellacean ecological stressor in this region, stressed arcellacean communities were observed to the northeast of the Fort MacKay area and warrant further examination.

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