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Seabird Transfer of Nutrients and Trace Elements from the North Water Polynya to Land during the Mid-Holocene Warm Period, Carey Islands, Northwest Greenland

Peter M. Outridge,[1] Michael E. Goodsite,[2] Ole Bennike,[3] Nicole Rausch[4] and William Shotyk[5]

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ABSTRACT. Seabird guano from large nesting colonies is known to increase trace metal levels in adjacent terrestrial environments today, when global oceans are contaminated with Hg, Cd, and other metals. But the effect of seabird guano in the pre-industrial period has rarely been studied. We used stable C and N isotopic and trace element analyses of a peat core that represents ca. 2000 years of organic matter accumulation to examine the effect on trace elements and nutrients of a seabird colony that existed in northern Baffin Bay during the Holocene Thermal Maximum (ca. 8000–5000 yr BP). Although C and N concentrations were typical of those in other peats, isotopic data identified marine organic matter as the main source of N and a minor source of C in the peat and showed that the unknown seabird was a fish-eating species that was summer-resident for the 2000 yr period. Concentrations of Cd, Br, Sr, and Zn in peat were up to an order of magnitude higher than in ombrotrophic (air-fed) bogs elsewhere, whereas Hg and Cu concentrations were similar to those in other peats, suggesting relatively low levels of Hg and Cu in the guano. This surprising result for Hg contrasts with studies on modern seabirds, in which guano markedly increased environmental Hg concentrations. It could be a consequence of Hg concentrations in Arctic marine food webs in the pre-industrial period that were an order of magnitude lower than those of today.

Key words: mercury; cadmium; seabirds; guano; peat; North Water polynya; Holocene; Thermal Maximum

RÉSUMÉ. On sait aujourd’hui que le guano d’oiseaux marins provenant des grandes colonies nicheuses fait augmenter les teneurs en métaux-traces dans les milieux terrestres adjacents et que tous les océans sont contaminés au Hg, au Cd et autres métaux. Toutefois, l’effet du guano d’oiseaux marins dans la période préindustrielle a rarement fait l’objet d’études. Nous avons utilisé des isotopes stables de carbone d’azote et des analyses d’oligoéléments d’une carotte de tourbe, qui représente environ 2000 ans d’accumulation de matières organiques, pour examiner l’effet de la présence d’une colonie d’oiseaux marins qui a existé dans le nord de la baie de Baffin pendant le maximum thermique de l’Holocène (environ 8000 à 5000 ans BP) sur les oligoéléments et les nutriments. Même si les concentrations de C et de N étaient semblables à celles se trouvant dans d’autres tourbes, les données isotopiques ont démontré que la principale source de N, et une source mineure de C, provenaient de matières organiques marines, indiquant ainsi que les oiseaux marins inconnus étaient des espèces piscivores qui y ont résidé l’été pendant 2000 ans. Les concentrations de Cd, Br, Sr et Zn dans la tourbe étaient d’un ordre de grandeur supérieur à celles des tourbières ombrotrophes (alimentées par l’air) d’ailleurs, tandis que les concentrations de Hg et de Cu étaient semblables à celles d’autres tourbes, ce qui laisse sous-entendre des niveaux relativement bas de Hg et Cu dans le guano. Ce résultat concernant le Hg est surprenant, car il est contraire aux études modernes sur les oiseaux marins démontrant que le guano a considérablement augmenté les concentrations de Hg dans l’environnement. Cette augmentation pourrait être attribuable aux concentrations de Hg dans les réseaux alimentaires marins de l’Arctique lors de la période préindustrielle, d’un ordre de grandeur inférieur aux concentrations d’aujourd’hui.

Mots clés : mercure; cadmium; oiseaux marins; guano; tourbe; polynie des eaux du Nord; Holocène; maximum thermique

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INTRODUCTION

Understanding the processes controlling the distribution of potentially toxic trace metals such as mercury (Hg) and cadmium (Cd) in the Arctic is an important step in evaluating the risk they pose to humans and wildlife. Currently, these metals occur in sufficiently high concentrations in some areas and in some animal species to pose a potential

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[1] Corresponding author: Centre for Earth Observation Science, University of Manitoba, 125 Dysart Road, Winnipeg, Manitoba R3T 2N2, Canada; Geological Survey of Canada, Natural Resources Canada, 601 Booth Street, Ottawa, Ontario K1A 0E8, Canada; peter.outridge@canada.ca

[2] Institute of Technology and Innovation, University of Southern Denmark, Campusvej 55, 5230 Odense M, Denmark

[3] Geological Survey of Denmark and Greenland (GEUS), Øster Voldgade 10, DK-1350 Copenhagen K, Denmark

[4] European Commission Joint Research Centre, Institute for Transuranium Elements, 76125 Karlsruhe, Germany

[5] Department of Renewable Resources, University of Alberta, Edmonton, Alberta T6G 2H1, Canada

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risk to wildlife or to human consumers of wild animals (AMAP, 2005, 2011).

The role that biological vectors such as seabirds and anadromous fishes play in metal cycling in the Arctic environment has received increased attention recently, following the seminal work on the subject by Blais et al. (2005). It has been known for some time that the substantial quantities of marine-derived organic matter and nutrients deposited on shore in the guano of large populations of nesting seabirds can greatly increase productivity in receiving polar ecosystems (Cocks et al., 1998; Wainwright et al., 1998). The guano is also rich in certain metals and organic contaminants acquired from marine food chains, and studies have shown that the runoff from colonies significantly elevates the levels of a number of metals (Godzik, 1991; Brimble et al., 2009a; Choy et al., 2010) and organic contaminants (Blais et al., 2005; Foster et al., 2011) in nearby terrestrial and freshwater areas.

Most published studies on seabirds as biovectors have concerned environments of modern times, when industrial pollution has increased the concentrations of Hg and Cd in the world's oceans (Sunderland and Mason, 2007; Cullen and Maldonado, 2013). For Hg, these increases in seawater have been mirrored in food chains. For example, a synthesis by Dietz et al. (2009) showed that current levels of Hg in a variety of Arctic marine mammal and seabird populations are consistently ca. 10 times those of the pre-1850 period. There is no evidence, however, that Cd or other trace metals are generally elevated in modern Arctic animals (Outridge et al., 1997, 2005; AMAP, 2005). Given these historical changes, or the lack thereof, would seabird biovectors in the pre-industrial period have had significant effects on concentrations of metals and nutrients in terrestrial and freshwater environments, as they do in modern times? Also, because the abundance of different polar seabird populations is strongly but variably influenced by climatic shifts (Stempniewicz et al., 2007), did the delivery of nutrients and metals from seabird biovectors vary as climate changed during the Holocene? These questions have rarely been investigated. Nie et al. (2012) reported that variations in the deposition of penguin and seal feces into an Antarctic pond, caused by climate-related population fluctuations, have had significant effects on background sedimentary Hg concentrations and fluxes over the past seven centuries.

The goal of this paper is to explore these questions in an Arctic setting. We studied a mid-Holocene peat profile that is known to have been associated with a colony of seabirds on the Carey Islands, northern Baffin Bay, for more than 2000 years (Bennike et al., 2008) to assess the impact of the birds on Hg, Cd, and other elements, including nutrients nitrogen (N) and carbon (C). Stable isotopes of C and N were used to reconstruct variations in seabird guano inputs, because these isotopes have proven to be informative about organic matter and nutrient sources. Stable N isotope ratios (represented as $\delta^{15}N$) in animal tissues increase by approximately 2‰–4‰ with each trophic level, providing a means to infer the trophic level at which the birds were feeding (Peterson and Fry, 1987; Hobson et al., 2002). $\delta^{13}C$ values often exhibit lower trophic fractionation (0.8‰–1.0‰ per level), but may be indicative of the sources of carbon within a food web. Other elements examined included bromine (Br) and strontium (Sr), two additional possible indicators of marine inputs, as well as copper (Cu) and zinc (Zn), which are biologically essential metals and exhibit the highest concentrations of any metal in seabird tissues in the North Water (NOW) polynya of Baffin Bay (Campbell et al., 2005). Marked climate shifts are known to have occurred in this region during the mid-Holocene (Vinther et al., 2009), so we examine possible relationships between reconstructed temperature histories, seabird-derived organic matter, and metal concentrations.

MATERIALS AND METHODS

Environmental Setting and Peat Growth History

The study site (76˚44.3′ N, 73˚12.5′ W) is located on Nordvestø, the largest island in the Carey Islands group 60 km off northwest Greenland (Fig. 1). The island (ca. 16 km^2) consists mostly of a rolling elevated plateau (140–230 m a.s.l.) above steep gneissic cliffs. The Carey Islands lie within the NOW polynya, one of the largest Arctic polynyas, which supports a highly productive marine ecosystem in northern Baffin Bay (Tremblay et al., 2006). The present climate is High Arctic, with mean monthly temperatures at the Thule Air Base 115 km east of the Carey Islands ranging from −24.6˚C in February to 4.5˚C in July, and an annual average of −11.2˚C (ClimaTemps, 2015). Vegetation consists of a thin cover of bryophytes, dwarf shrubs, and perennial herbs and is classified as High Arctic tundra.

A series of peat moss hummocks up to 2.5 m high and up to ~8 m in diameter (M. Goodsite, pers. observ.) are found on rocky ground at an elevation of 140 m above sea level, close to the northern rim of the island below a north-sloping ridge (see photographs in Brassard and Blake, 1978). The mostly frozen hummocks consist of well-preserved remains of the coprophilous moss *Aplodon wormskioldii*, with an active layer ~10 cm deep in July (M. Goodsite, pers. observ.). This species is exclusively associated with dung, guano, or other N-enriched substrates (Steere, 1973). Radiocarbon dating of one hummock by Brassard and Blake (1978) gave a basal peat date of 6300 yr BP (conventional ^{14}C years) at 253–258 cm depth, while another age at the top of the hummock indicated that peat growth ceased at some point between 4500 and 4000 yr BP. Later, more extensive dating of another hummock nearby gave median ages of 7110 to 5135 cal. yr BP from 205 cm to the top of the frozen peat (Bennike et al.; 2008; see online Appendix 1: Fig. S1). Both sets of results place the period of peat growth during the mid-Holocene Thermal Maximum, which is thought to have occurred between 8000 and 5000 yr BP in the eastern Canadian Arctic and North Greenland (Gajewski, 2015).

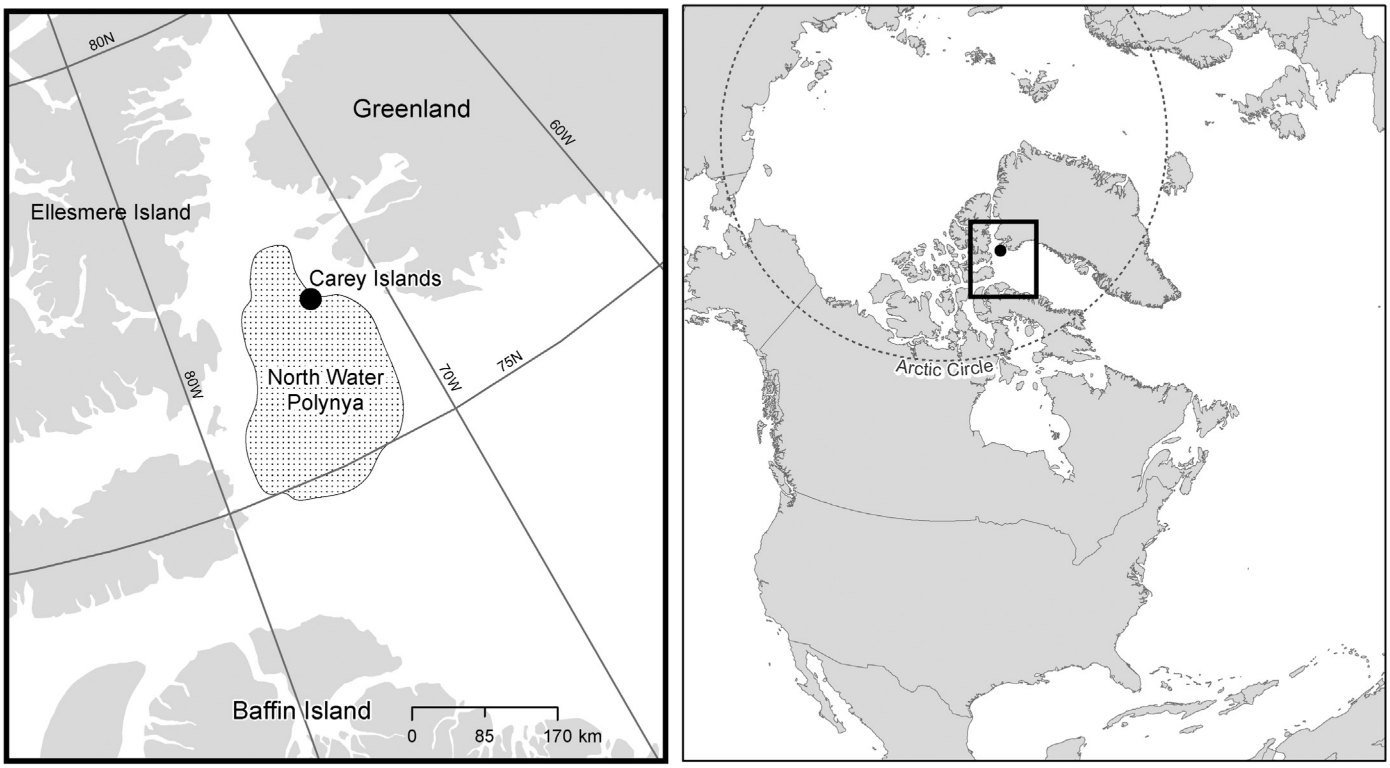

FIG. 1. Map showing location of the Carey Islands study site, with the approximate summer position of the North Water Polynya indicated by cross-hatching.

After this warm period, plant growth was negligible until recent decades owing to a colder climate and snow accumulation over the hummocks, which lasted into summer (Brassard and Blake, 1978).

The *A. wormskioldii* peat hummocks are associated with a former large nesting colony of seabirds, as indicated by fragments of feathers throughout the peat profiles, although no seabirds currently nest on this site (Brassard and Blake, 1978; Bennike et al., 2008). We attempted to identify the bird species from feather remains or by extracting bird DNA from the peat, but without success. The question of which species was involved is discussed later.

Field Work and Sample Preparation

The peat core described by Bennike et al. (2008) was used for the present study. Briefly, one of the larger peat hummocks at the site was cored to a depth of 207 cm in July 2001. A motorized peat corer (Nørnberg et al., 2004) was used because all but the upper few centimetres of peat is permanently frozen. The thawed active layer was discarded before coring, so that the samples represent only frozen peat. The core diameter was 9.7 cm, and core recovery was 100%. The core sections were frozen and returned to Denmark, where they were sectioned into 1 cm increments while in a frozen state. Part of each slice was set aside for biological analyses and part for chemical analysis. The latter samples were freeze-dried, and obvious fragments of feathers were removed. The edges of the peat slices were trimmed away with an acid-rinsed ceramic knife, and the remaining sample was homogenized in a centrifugal mill equipped with a titanium rotor and 0.25 mm Ti sieve (Ultracentrifugal Mill ZM 1-T, F. K. Retsch GmbH and Co., Haan, Germany).

Analytical Procedures and Statistics

Powdered subsamples of dried peat (~100 mg) were analyzed for total Hg by solid sample thermal desorption analyzer (LECO AMA-254), calibrated with Hg solutions of known concentrations prepared daily. Standard reference materials (SRMs) included with the analyses (BCR-281 Rye Grass [*N* = 7] and NIST 1515 Apple Leaves [*N* = 3]) gave measured mean ± SD values of 19.5 ± 1.5 and 43.3 ± 0.9 ng/g DW compared with certified values of 21 ± 2 and 44 ± 4 ng/g DW, respectively. The calculated lower limit of detection of the method was 4.4 ng/g DW.

Cadmium was determined by acid digestion (HF/HNO_3) of ~100 mg samples followed by graphite furnace absorption spectrometry (GF-AAS) on digest solutions diluted as appropriate to remain within the working range. A subset of the digest solutions was reanalyzed with flame AAS (which has a higher working range) because of the high concentrations. The median difference between methods was 5.8% (GF-AAS data were higher on average). Concurrent analysis of SRM BCR-62 Olive Leaves (*N* = 6) gave a mean ± SD concentration of 0.095 ± 0.020 μg/g DW, which compared well with the certified value of 0.10 ± 0.02 μg/g DW. Repeated analysis of two samples throughout the analytical work gave procedural relative standard deviations of 13.4%

and 12.8% ($N = 20$ each). The lower limit of detection was 0.03 μg/g DW.

The other trace elements (Br, Sr, Cu, and Zn) were determined in powdered subsamples by x-ray fluorescence (Energy-dispersive Miniprobe Multielement Analyzer [EMMA]) as described by Cherbukin and Shotyk (1996). The instrument was calibrated with a variety of international plant and soil SRMs, and the analytical accuracy and precision values were reported by Shotyk et al. (2000). The lower limits of detection (in μg/g DW) of the elements reported here are 0.5 Br, 1.0 Sr, 2.0 Cu, and 1.4 Zn.

This study employed stable isotope ratios of C and N, as well as total organic carbon (TOC) and total nitrogen (TN) concentrations, to better understand and quantify the impact of seabird guano on metal profiles, as recommended by Hobson (2005). Stable C and N isotopes, TOC, and TN were determined through combustion conversion of sample material to N_2 and CO_2 gases, which were carried in helium through a Costech Instruments Elemental Analyzer (CHNS-O ECS 4010, Milan) coupled to a Deltaplus XL Continuous Flow Stable Isotope Ratio Mass Spectrometer (Thermo Finnigan, Bremen). Samples were pre-rinsed with dilute hydrochloric acid to remove carbonates. We weighed a 0.5 mg subsample for TOC and $^{13}C/^{12}C$ determinations and a 1.5–2 mg subsample for TN and $^{15}N/^{14}N$ determinations. Sample and standard gases for N measurements were first passed through a Carbopure trap to remove CO_2. Carbon isotope ratio data are expressed in standard notation ($\delta^{13}C$) relative to Vienna PeeDee Belemnite, and N isotope ratios, as $\delta^{15}N$ relative to N_2 in air. Data quality was monitored and corrections made using an array of international reference material and in-house standards that are calibrated using certified SRMs: IAEA-N1 + IAEA-N2 (Ammonium Sulphate), IAEA-CH3 (cellulose) + IAEA-CH6 (sugar), USGS-40 + USGS-41 (L-Glutamic Acid). These SRM measurements were used in data normalization and to ensure daily mass spectrometry precision and accuracy. With these quality control checks, the expected accuracies for reportable data are +0.2‰ for $\delta^{13}C$ and +0.3‰ for $\delta^{15}N$. Duplicate measurements were made at the end of the sample batch on three samples interspersed throughout the run, giving mean differences between duplicates of 0.52% DW for TOC, 0.04% for TN, 0.15‰ for $\delta^{13}C$, and 0.09‰ for $\delta^{15}N$.

Data on several animal macrofossils (number of bird feathers, chironomid midge head capsules, and *Cyclorrapha* fly head capsules) reported by Bennike et al. (2008) are included in the present paper because their abundances were found to be significantly correlated with one or more chemical variables. Briefly, the frozen section of each slice set aside for biological analysis was thawed and wet-sieved on 0.4 and 0.2 mm sieves, and the residues were examined microscopically.

Relationships between elements, stable isotope ratios, and biological variables were initially tested statistically using non-parametric Spearman Rank Order correlation, whereas differences in metal concentrations between time periods were tested with Kruskal-Wallis One-Way ANOVA on Ranks (SigmaPlot v. 12.5, Systat® Software). These tests are not affected by deviations from data normality and homoscedasticity. Subsequently, the relative importance of organic matter concentration (TOC and TN) and source indicators ($\delta^{15}N$ and $\delta^{13}C$) in explaining metal concentration variability were tested with forward stepwise linear regression modeling (SigmaPlot v. 12.5). Concentration and isotope ratio data were $\log_n$ transformed prior to analysis (Sokal and Rohlf, 1981), which resulted in normally distributed data in all cases. Significant relationships noted in the text are at $p < 0.05$ unless otherwise stated.

RESULTS AND DISCUSSION

Carbon and Nitrogen as Seabird Input Indicators

Total organic carbon concentrations and $\delta^{13}C$ in bulk peat were not correlated (Table 1). Concentrations of TOC (mean + SD: 45.4 + 0.8% dry wt; Fig. 2a) were similar to values in pre-industrial ombrotrophic (precipitation- and air-fed) peats that were remote from marine influence (Disnar et al., 2008; Outridge and Sanei, 2010; Esmeijer-Liu et al., 2012). However, $\delta^{13}C$ values in *A. wormskioldii* suggested that marine inputs of C were a minor yet significant fraction of total C throughout the 2000 yr study period, mixed with terrestrial (soil/atmospheric) C. Stable C isotopic ratios in the peat reflect the uptake of C derived from two possible sources: marine organic matter (OM) and terrestrial C. These sources, or "isotopic end-members," currently exhibit distinctly different ranges of isotope ratios (see below). By comparing peat isotopic values with end-member values, we can infer the relative importance of the different sources by proportion (Hobson et al., 1994; Robinson, 2001). However, the $\delta^{13}C$ end-member values must be corrected for the effects of anthropogenic CO_2 emissions that exhibit depleted ^{13}C and have lowered average marine and terrestrial OM $\delta^{13}C$ since the beginning of the Industrial Revolution (the "Suess Effect"; Keeling, 1979). These adjustments should be regarded as approximate, particularly for terrestrial plant OM, because other variables that may change over long periods of time, such as air temperature, precipitation, and soil moisture, can also significantly affect plant $\delta^{13}C$ values (Tillman et al., 2010). Regional variations in the impact of the Suess Effect in oceans can also be expected (Bauch et al., 2000).

In the NOW polynya at present (samples taken in the 1990s), $\delta^{13}C$ in marine OM ranged from −22.5‰ to −18‰ (Hobson et al., 2002; Campbell et al., 2005), which is markedly higher than the range of −32.1‰ to −25.8‰ in terrestrial plants of the Carey Islands region (Blake, 1991). Average $\delta^{13}C$ of −27‰ in terrestrial OM in the Arctic Ocean (Schubert and Calvert, 2001) is consistent with the Carey Islands terrestrial values. This $\delta^{13}C$ value is typical of modern terrestrial C_3 plants in general, including mosses, which ranged from −35‰ to −22‰ with a mean of −27‰ (Meyers,

TABLE 1. Results of non-parametric Spearman Rank correlation analyses of elemental, isotopic, and biological variables in the peat core over two millennia. ("C. hir" – chironomid (midge) abundance; "C. ych" – *Cyclorropha* sp. (fly) abundance; "Feathers" – abundance of bird feathers. Values shown are correlation "r" coefficients; significance levels indicated as: * – $p < 0.05$, ** – $p < 0.01$, ** with bold font – $p < 0.001$. N varied between pair comparisons: N = 189–205 for inter-element pairs, N = 37 for element-isotope and element-TOC and -TN pairs, N = 97–103 for element-biological pairs, and N = 37 for isotopic-, TOC-, and TN-biological pairs).

	Cd	Cu	Zn	Br	Sr	TOC	TN	$\delta^{13}C$	$\delta^{15}N$	C. hir	C. ych	Feathers
Hg	**0.465****	−0.097	0.141*	**0.659****	**0.647****	0.085	**0.550****	0.083	0.237	**0.416****	**0.353****	0.080
Cd		−0.200**	**0.240****	**0.571****	**0.560****	−0.024	0.492**	0.003	0.418*	0.309**	0.173	0.205*
Cu			0.104	0.083	−0.007	−0.496**	**0.538****	−0.071	**0.661****	−0.123	−0.052	0.004
Zn				**0.295****	0.198**	−0.153	**0.598****	−0.325	**0.497****	−0.117	−0.080	0.153
Br					**0.736****	−0.102	**0.712****	−0.155	0.443**	0.306**	0.181	−0.004
Sr						−0.170	**0.652****	0.076	0.429**	**0.413****	0.278**	0.141
TOC							0.0868	−0.010	−0.335*	0.296	0.168	−0.174
TN								0.107	**0.781****	0.270	0.288	0.168
$\delta^{13}C$									0.122	−0.028	0.020	−0.278
$\delta^{15}N$										0.052	0.068	0.196
C. hir											**0.535****	-0.078
C. ych												−0.076

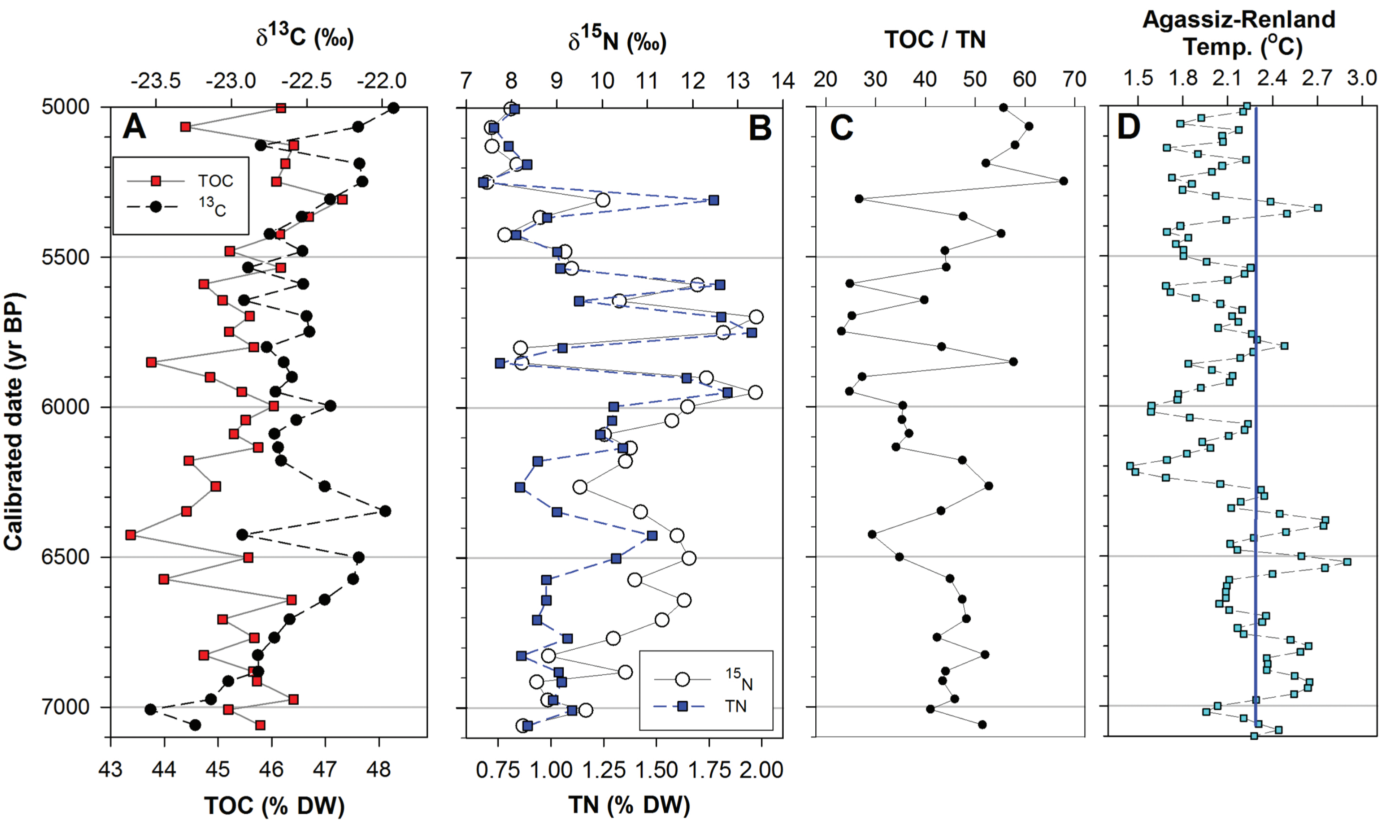

FIG. 2. Carbon (A) and nitrogen (B) stable isotope ratio and concentration profiles in the peat core, (C) TOC/TN in the peat core, and (D) reconstructed temperature histories for the region from 7100 to 5000 yr BP. In panel D, temperature data are reconstructed average air temperatures based on $\delta^{18}O$ in Agassiz-Renland glacier ice (Vinther et al., 2009). Each ice core data point is a three-point running average integrating temperatures over a 40-year span on average. The vertical line in (D) is the mean temperature for the period 10 200 to 5000 yr BP.

1994; Koch, 1998). Terrestrial plant $\delta^{13}C$ has exhibited an average decline of ~2.7‰ since the industrial era began (Zhao et al., 2001), while Arctic marine food chains may have experienced a decrease of ~2‰ (Bauch et al., 2000, 2002). Thus, if the modern terrestrial and marine $\delta^{13}C$ end-members are assumed to be −27‰ and −20‰, respectively, the Suess Effect-corrected Holocene end-member values would be −24.3‰ and −18‰. All of the peat samples (mean −22.6 ± 0.3‰, range −23.5 to −21.9‰; Fig. 2a) fell between the terrestrial and marine $\delta^{13}C$ end-members, which indicates a mixture of sources. A 50:50 mix of terrestrial and marine C in peat should exhibit $\delta^{13}C$ of −21.2‰; the mean peat $\delta^{13}C$ of −22.6‰ suggests an average 73:27 ratio of terrestrial to marine sources.

Total N in *A. wormskioldii* averaged 1.1 ± 0.3% DW, which is comparable to that in pre-industrial ombrotrophic peats elsewhere (Esmeijer-Liu et al., 2012). However, $\delta^{15}N$ results suggested that the marine source (guano) made more difference to the total amount of N in peat than to the total amount of C. Unlike $\delta^{13}C$ end-members, $\delta^{15}N$ end-member values were not adjusted for anthropogenic N effects in the 20th century because of a lack of evidence for significant changes in baseline $\delta^{15}N$ in Arctic marine food-chains since the pre-industrial period (Outridge et al., 2005, 2009) and conflicting evidence for a shift in northern terrestrial plants. Although Esmeijer-Liu et al. (2012) reported a substantial decrease in peat $\delta^{15}N$ over the past few centuries, they attributed this change partly to diagenesis of the older OM. Values of $\delta^{15}N$ in the teeth and bones of a range of northern terrestrial herbivore species from the Pleistocene and Holocene were similar to those of their modern counterparts (Bocherens et al., 1997, 2015), suggesting that the $\delta^{15}N$ values of their forage plants were also similar. In the absence of a clearly substantiated correction factor, the modern $\delta^{15}N$ end-member values are applied here to the mid-Holocene peat data.

$\delta^{15}N$ in Arctic terrestrial plant OM unaffected by marine N ranges from ca. −5‰ to +2‰ (Schubert and Calvert, 2001; Kristensen et al., 2011), which is clearly well below the values in *A. wormskioldii* (range: 7.5‰ to 13.2‰, mean ± SD: 10.0‰ ± 1.7‰; Fig. 2b). The peat $\delta^{15}N$ data are thus indicative of predominantly marine N inputs. However, the selection of a marine end-member value to exactly quantify the marine input is problematic. Since the identity of the mid-Holocene seabird species is not known with certainty, its likely trophic position and guano $\delta^{15}N$ value cannot be ascertained directly. In the NOW ecosystem, $\delta^{15}N$ values exhibit a range of 5‰ to 7‰ in marine particulate and algal OM and increase by 3.8‰ on average at each trophic level above this baseline (Hobson et al., 2002; Campell et al., 2005), which is typical of $\delta^{15}N$ patterns in food-chains. Seabirds currently inhabiting the NOW polynya exhibit average muscle $\delta^{15}N$ values ranging from 10.9‰ in the zooplantivorous Dovekie (*Alle alle*) to 16.5‰ – 17.2‰ in two species of piscivorous gulls (*L. hyperboreus* and *L. thayeri*), and these values reflect their different trophic niches (Campbell et al., 2005).

A possible trophic position for the seabird can be calculated from the peat $\delta^{15}N$ data. Total N concentrations and $\delta^{15}N$ in peat had a high positive correlation ($p < 0.001$, Table 1), which indicates that peat $\delta^{15}N$ was tightly controlled by the amount of TN inputs from guano. Coincident peaks of high TN, and high $\delta^{15}N$ values up to 13.2‰, occurred several times between 6000 and 5500 yr BP. Peat $\delta^{15}N$ represents a mixture of marine and terrestrial sources ranging from 0% to 100%. Since marine N cannot have contributed more than 100% of peat TN, the maximum peat $\delta^{15}N$ value of 13.2‰ means that the average guano $\delta^{15}N$ value must have been 13.2‰ or greater. In addition, some degree of isotopic fractionation can be expected between the guano and seabird tissues because of N metabolism. Wainwright et al. (1998) reported that guano in several Arctic seabird species was depleted in ^{15}N relative to blood (representing their tissues): guano $\delta^{15}N$ was on average 2.5‰ below blood values. If this average can be applied to the peat (guano) values in this study as a correction factor, then the unknown seabird species may have had an average tissue $\delta^{15}N$ value of at least 15.7‰ (minimum guano $\delta^{15}N$ of 13.2‰ + 2.5‰ fractionation correction). On the basis of the NOW basal food web $\delta^{15}N$ values of 5‰ – 7‰ (average of 6‰) and an increase of 3.8‰ at each trophic level (Hobson et al., 2002), it may be inferred that the mid-Holocene seabirds were feeding at least 2.6 trophic levels ((15.7−6)/3.8 = 2.6) above the marine algal baseline, on prey items such as zooplanktivorous and piscivorous fish species (cf. Bradstreet and Brown, 1985; Campbell et al., 2005). This inference should be treated cautiously, because the guano $\delta^{15}N$ values may have been altered after deposition by ammonification of urea-N, which has the potential to significantly increase the $\delta^{15}N$ signature of TN (Mizutani et al., 1986). Conversely, there may have been some contribution to peat TN from prey (fish) debris dropped near the colony, which would likely have had a lower average $\delta^{15}N$ value than guano, being a trophic level lower than the seabird tissues. This process would have tended to counteract the ammonification effect on peat $\delta^{15}N$.

The finding of a dominant marine source of N, and lesser marine C, in the peat is unlikely to be affected by OM decomposition and its effects on C and N isotopic ratios. TOC/TN, $\delta^{13}C$ and $\delta^{15}N$ values in peat bogs may be altered by decomposition to produce characteristic patterns (Agren et al., 1996; Kuhry and Vitt, 1996; Zaccone et al., 2011). The magnitude of $\delta^{13}C$ and $\delta^{15}N$ alteration due to long-term OM diagenesis in a variety of environments, including peat bogs, is less than ~2‰ for both isotope systems (Meyers, 1994; Sharma et al., 2005; Zaccone et al., 2011). This effect is minor compared to the difference between the average terrestrial and marine $\delta^{15}N$ end-members in the Baffin Bay region. Decomposition also tends to produce smooth trends in TOC, TN, and isotope ratios downcore, unlike the rapid 4‰ – 5‰ fluctuations of $\delta^{15}N$ in *A. wormskioldii*. We conclude therefore that any decomposition effects on peat OM stable isotopic values were negligible and will not alter the findings of the study. This conclusion is consistent with Brassard and Blake's (1978) report of remarkably well-preserved cells and macroscopic structures of *A. wormskioldii* leaves.

To explore possible relationships between changing Holocene climate and seabird abundance, we investigated whether changes in peat $\delta^{15}N$ and TN concentrations may be semi-quantitative indicators of seabird abundance. Peat TN increased or decreased by up to two-fold from one data point to the next several times between 6000 and 5300 yr BP; simultaneously, $\delta^{15}N$ fluctuated rapidly, by up to 5‰ within 100 years (Fig. 2b). If it is assumed that TN is proportional to guano deposition onto the peat surface and that the deposition value was related to seabird abundance at the time, then the isotopic evidence suggests that seabird abundance fluctuated dramatically in the latter part of the mid-Holocene Thermal Maximum. The suggested mechanism underlying a possible climate – bird abundance relationship is the link between air temperature and the summer ice-free area and productivity of the NOW polynya, where northern Baffin Bay seabirds feed during summer (Hobson et al., 2002). A warmer climate would favour a larger ice-free opening of the polynya, with the improved light climate supporting a more productive marine ecosystem (Hamel et al., 2002). In turn, this ecosystem could potentially support a larger population of nesting seabirds. The mid-Holocene Thermal Maximum was associated with relatively open summer waters in Baffin Bay, as evidenced by driftwood delivery to ancient beaches (Dyke et al., 1997) and diatom microfossils in marine sediments (Levac et al., 2001), which also suggest a larger and longer-lasting ice-free polynya than currently. Organic indicators in marine sediment cores show that the productivity of marginal areas of the polynya, especially the northern margin near the Carey Islands, is particularly sensitive to climate-driven changes in sea-ice cover and nutrient supply, whereas the interior core area is relatively stable (Hamel et al., 2002; Bailey et al., 2013).

A reconstruction of air temperatures during the mid-Holocene based on two glacier ice cores from the region showed several points of coincidence between inferred peaks in air temperature and peat TN and δ^{15}N, for example, at 5300 yr BP and 6600 – 5900 yr BP (Fig. 2d cf. 2b). But over the 2000-year study period as a whole, there were no significant correlations between air temperatures (interpolating ice core dates to match peat core dates) and the peat C and N isotopic and concentration profiles ($p > 0.10$ in all cases). One possible explanation for the lack of an association is that the relationship between seabird abundance and summer air temperature, if there is one, is not linear. Although polynya area and productivity may be important factors in seabird abundance generally, other possible limiting factors, such as limited nesting areas, predators, or inter-specific competition, may be influential (Stempniewicz et al., 2007). The area of the NOW that is free of sea ice may also be affected by variations in Greenland glacier calving and wind and ocean circulation patterns that are not directly related to air temperature (Bailey et al., 2013).

Despite the absence of evidence for close control of seabird abundance by the relatively warm climate of the mid-Holocene Thermal Maximum, the isotopic evidence indicates that fish-eating seabirds nested at the study site during summer continuously throughout the 2000-year study period. The decline in peat TN and δ^{15}N after 5300 yr BP could indicate a declining seabird population, a trend that accords well with climate reconstructions showing a transition to cooler temperatures commencing at ~5200 yr BP in North Greenland (Vinther et al., 2009; Gajewski, 2015). The continuous summer occupation when temperatures in North Greenland were on average ~1° – 1.5°C higher than today (Gajewski, 2015), followed by population decline and colony abandonment after the mid-Holocene Thermal Maximum until the present day, points to a primary relationship between climate and the presence or absence of the seabird species at the site. Although the date of complete abandonment of the nest site by the birds cannot be ascertained, it may have coincided with the cessation of peat growth soon after ~5135 yr BP (see online Fig. S1). With the recent warmer climate in northern Baffin Bay, summer populations of species such as the Atlantic Puffin (*Fratercula arctica*), which historically were locally rare, have been increasing, which could indicate that the species is recolonizing the area as summer temperatures increase (Burnham and Burnham, 2010).

Seabirds as Metal Biovectors in the Mid-Holocene

The patterns of isotopic ratios, as well as TOC and TN concentrations, may be compared to changes in peat metal concentrations (Fig. 3) to judge the impact of seabird guano on peat metal levels. A common feature of all of the metal profiles is significantly higher concentrations during the period from 6100 to 5500 yr BP than before or after that period. Correlation analyses showed that TN and δ^{15}N were of greater importance in terms of peat element concentrations than TOC and δ^{13}C, which in most cases were not significantly influential variables (Table 1). Biological variables (invertebrate abundance in particular) were highly correlated with some elements, especially Hg and Sr. Cadmium concentrations were correlated with chironomid abundance ($p < 0.01$) and number of bird feathers.

To judge the relative influence on metal levels of the changes in C and N isotopes and concentrations, each element was regressed against the OM variables using forward stepwise multiple linear regression. This analysis produced a ranked order of variables cumulatively explaining the most variance in metal concentrations (Table 2). Total N, which is assumed to represent variations in the amounts of deposited guano, was the most significant variable for all elements except Cu, and the only one of significance for Cd, Br, and Sr. Concentrations of peat Hg and Cu were also significantly ($p < 0.001$) affected by variations in δ^{15}N. Mercury levels in different species in the NOW ecosystem today are highly correlated with trophic position as inferred from δ^{15}N, and Cu is weakly correlated (Campbell et al., 2005). The peat results could be interpreted, therefore, to mean that variations in seabird trophic position during the mid-Holocene may have influenced the rates of Hg and Cu deposition onto the peat, separately from the effect of the guano deposition rate. However, the correlation between peat TN and δ^{15}N makes this conclusion problematic because the effect of isotopic variations may be confounded by changes in N input.

How do the metal concentrations in *A. wormskioldii* compare with those in other peat deposits from the pre-industrial period? We have not found other literature reports of metals in guanogenic peat from this era. From the modern period, surface moss data from Spitsbergen showed that the ranges of Cd (1.1 – 8.0 µg/g DW), Cu (2.6 – 7.3 µg/g

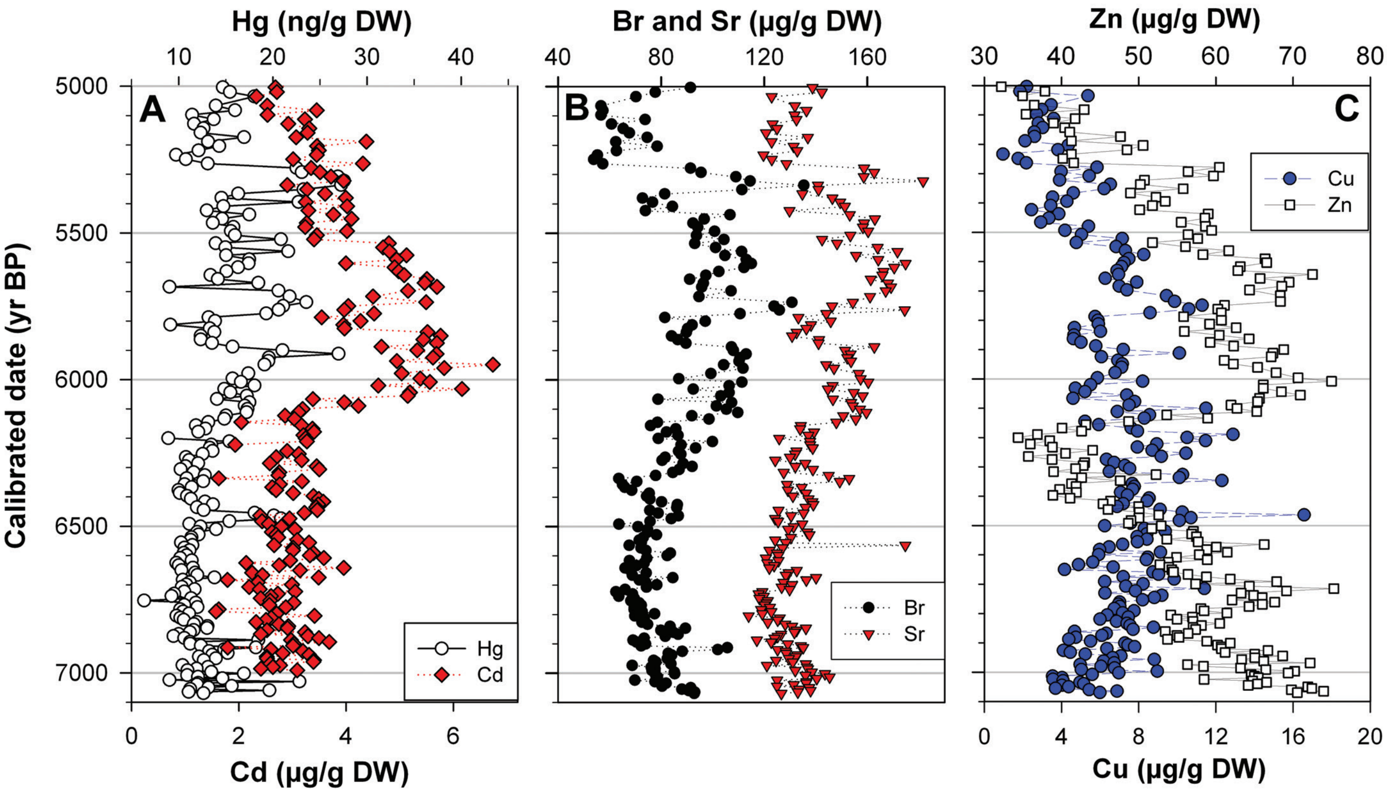

FIG. 3. Trace metal and other element concentrations in the Carey Islands peat core from 7100 to 5000 yr BP (A) Hg and Cd, (B) Br and Sr, and (C) Cu and Zn.

TABLE 2. The significant organic matter concentration and source indicators that explain the most variance in peat trace element concentrations. Results of forward stepwise linear regressions are shown. Degrees of freedom = 36 in all cases. Only the variables that contributed significantly to explaining element concentrations were included in the table.

Element	Model step	Variable entered into model	Cumulative model r^2	Probability of entry into model
Hg	1	TN	0.45	< 0.001
	2	$\delta^{15}N$	0.64	< 0.001
Cd	1	TN	0.24	0.003
Br	1	TN	0.56	< 0.001
Sr	1	TN	0.43	< 0.001
Cu	1	$\delta^{15}N$	0.49	< 0.001
	2	TOC	0.55	0.027
	3	$\delta^{13}C$	0.60	0.054
Zn	1	TN	0.30	< 0.001
	2	$\delta^{13}C$	0.47	0.002

DW), and Zn (11–47 µg/g DW) concentrations in two moss species (*Sanionia uncinata* and *Hylocomium splendens*) within a Dovekie colony were up to twice as high as in specimens collected outside the colonies (Godzik, 1991), and were similar to those in the mid-Holocene *A. wormskioldii* deposit (cf. Table 3). Ombrotrophic peat bogs from the Holocene may provide the closest point of comparison to the present study. The underlying rock substrate and absence of a water table mean that the *A. wormskioldii* hummocks were not minerogenic peat. Comparison of average element concentrations in the Carey Islands peat with those in pre-industrial ombrotrophic peats elsewhere in the Northern Hemisphere shows that Cd, Br, Sr, and Zn are substantially higher in *A. wormskioldii* (Table 3). The impact of guano on Cd concentrations (~2–6 µg/g DW) was especially marked, and these concentrations fall in the lower half of the range of highly contaminated soils in a modern context (UNEP, 2010). For example, the Cd levels in *A. wormskioldii* are comparable to those in peats 25–43 km from the Rouyn-Noranda Cu smelter in Quebec (5.5–7.8 µg/g DW, Dumontet et al., 1990) and several times as high as peats affected by a Cu-Ni smelter in Finland (< 2 µg/g DW, Rausch et al., 2005).

In contrast, Hg and Cu concentrations in *A. wormskioldii* fell within ranges similar to those of other peats (see Table 3). The absence of high Hg concentrations in *A. wormskioldii* was surprising given that piscivorous seabird colonies in polar regions today have major impacts on Hg levels in nearby terrestrial and freshwater ecosystems (Blais et al., 2005; Choy et al., 2010; Zheng et al., 2015).

TABLE 3. Comparison of trace element concentrations in Northern Hemisphere ombrotrophic (rainwater-fed) peat bogs in the pre-industrial era and in the guanogenic peat at the Carey Islands. Units are µg/g DW, except for ng/g DW for Hg. LoD = limit of detection. Data shown are mean ± SD, except where ranges are presented.

	Hg	Cd	Br	Sr	Cu	Zn	Sources
Carey Islands	14 ± 5	3.5 ± 1.0	85.0 ± 16.1	139 ± 15	6.5 ± 2.4	56.0 ± 10.3	Present study
Sask Lake 4, Flin Flon, Manitoba, before 1900 AD	61 ± 21		12.8 ± 3.2	19.1 ± 2.7	7.7 ± 1.8	14.0 ± 8.4	Outridge et al., 2011
Hietajärvi bog, southern Finland, before 1100 AD		0.08 ± 0.01			1.3 ± 0.2	4 ± 2	Rausch et al., 2005
Inuvik, Northwest Territories, 6250–180 yr BP	34 ± 13		9.2 ± 2.8	23.5 ± 8.4	< 1.5 (LoD)	1.5 ± 1.0	Outridge and Sanei, 2010
Bathurst Island, Nunavut, 5900–800 yr BP	28 ± 4						Givelet et al., 2004
Swiss Jura Mountains, before 1340 AD							
– Étang de la Gruère	25 ± 4		16.3 ± 2.8				Roos-Barraclough and Shotyk, 2003
– La Tourbière de Genevez	68 ± 15		15.6 ± 2.9				Roos-Barraclough and Shotyk, 2003
Faroe Islands, before 880 AD			200–800	10–60			Shotyk et al., 2005
Norway, 6 sites, before 1000 AD	15 ± 8						Steinnes and Sjøbakk, 2005
Southern Germany, before 1440 BP		0.1–0.4					Küster and Rehfuess, 1997
Étang de la Gruère, Switzerland, 8030–240 yr BP		0.076 ± 0.022			0.5–8	1–30	Shotyk et al., 2002

The lack of a seabird effect on peat Cu concentrations is also surprising, because Cu is one of the most highly concentrated elements in the tissues of modern seabirds in the NOW polynya (Campbell et al., 2005). The singular absence of a correlation between Cu and TN, an indicator of the amount of guano deposition, suggests that Cu may not have been excreted in large quantities in the guano, possibly because of its role as an essential nutrient. However, a definitive explanation for the Cu finding is lacking.

For Hg and Cd, their increase, or lack of increase, in Arctic marine ecosystems since the pre-industrial period is the most conservative explanation for the difference between this study and others concerning the impact of seabird guano on environmental Hg and Cd levels. The modern seabirds studied by Blais et al. (2005) and Choy et al. (2010), among others, feed in Arctic marine food webs that on average contain Hg concentrations an order of magnitude higher than in pre-industrial times (Dietz et al., 2009). This conclusion was in part based on Hg trends in the feathers of raptors from West Greenland (Dietz et al., 2006), showing that this region has experienced a degree of Hg contamination similar to that found elsewhere in the Arctic. In contrast to Hg, Cd levels have not increased in Arctic marine animals since the pre-industrial period despite increased global emissions from industry (Outridge et al., 1997, 2005; AMAP, 2005). Unlike Hg, Cd is bio-diluted (i.e., decreases or is stable with increasing trophic position) in marine food webs, including that of the NOW polynya (Campbell et al., 2005). Naturally high Cd concentrations occur in zooplankton, particularly the amphipod *Themisto libellula*, in the NOW marine food web. Thus, the amounts of Cd in the mid-Holocene seabird guano may have been similar to those of the modern period, whereas Hg is likely to have been an order of magnitude lower. This pattern could explain why the Holocene seabirds, like seabirds today (e.g., Brimble et al., 2009b), delivered significant quantities of Cd to areas around the colony, but negligible amounts of Hg.

In the absence of definitive evidence, the identity of the seabird in question remains unknown but may be constrained by currently available information. The position of the colony, on a rocky plateau above high cliffs on a small offshore island, is consistent with Atlantic Puffins (Bennike et al., 2008) or with Dovekies, which usually nest in crevices on talus slopes up to 400 m above sea level and are currently the most common colony-nesting seabird in the region (Montevecchi and Stenhouse, 2002). The site would be an unusual nesting location for other Arctic-nesting seabirds such as Thick-billed Murres (*Uria lomvia*), Northern Fulmars (*Fulmarus glacialis*), Black-legged Kittiwakes (*Rissa tridactyla*), and Glaucous Gulls (*Larus hyperboreus*), which are typically found in large colonies on steep cliff faces. Black Guillemot (*Cepphus grylle*) breeds in solitary pairs or in small groups near sea level. Puffins and Dovekies are distinguished from each other by their feeding preferences, mainly fish and zooplankton, respectively (Lowther et al., 2002; Montevecchi and Stenhouse, 2002). If the calculation above of the birds' average tissue δ^{15}N of ≥ 15.7‰, based on its guano δ^{15}N values, is approximately correct, then the species was predominantly a fish-eater. Dovekies in the NOW ecosystem today, on the other hand, are distinguished from other seabirds by a relatively low tissue δ^{15}N of ~11‰ (Campbell et al., 2005). Thus, it can be inferred that puffins were the species most likely to have formed the colony.

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APPENDIX 1

The following figure is available as a supplementary file to the online version of this article at: http://arctic.journalhosting.ucalgary.ca/arctic/index.php/arctic/rt/suppFiles/4577/0

FIG. S1. Interpolated dates from ^{14}C age dating of Carey Islands peat core. (Regression equation: Calibrated date (yr BP) = $4.9 \times 10^3 + 16.1 * \text{Depth} - 2.72 \times 10^{-2} * (\text{Depth})^2$; $r^2 = 0.981$, $p < 0.001$. Dashed lines represent 95% confidence intervals.)

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