

**MERCURY DYNAMICS IN SUB-ARCTIC LAKE SEDIMENTS ACROSS
A METHANE EBULLITION GRADIENT**

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A thesis submitted in partial fulfillment of the requirements for the degree of Bachelor of

Arts (Geology)

At

Gustavus Adolphus College

2014

Mercury Dynamics In Sub-Arctic Lake Sediments Across A Methane Ebullition Gradient

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Abstract

Recent studies have suggested that Arctic warming may play a key role in enhancing carbon (C) and mercury (Hg) export from permafrost peatlands, yet the mechanisms by which Hg is mobilized during thaw remain enigmatic. To elucidate the links between these chemical systems, we investigated Hg concentrations in cores taken in organic C rich sediments in Lake Villasjön (avg. depth 1.5m) at the Stordalen Mire, Abisko, Sweden. We chose coring sites based on zones with significantly different ebullitive methane (CH₄) fluxes established in earlier studies and we hypothesized that the microbial community producing CH₄ is also potentially mobilizing Hg. Four recovered sediment cores (39-44cm long) were characterized by having roughly 30cm of organic-rich silt material on top of a transition to more clastic material in the bottom ~10cm. Cores were sub-sampled every 2cm, and the sediment samples were then freeze-dried. Sub-samples were analyzed for extractable Hg by cold vapor inductively coupled plasma mass spectrometry (2 cores) and for total Hg by thermal decomposition and cold vapor atomic fluorescence (2 cores). Mercury was most abundant in the upper portions of all sediment cores, after which concentrations decreased with depth. The highest ebullition site had Hg concentrations exceeding 80 ng_{Hg}/g_{sediment} at the core top that decreased to a low Hg concentration < 15 ng_{Hg}/g_{sediment} at the core base. The lowest ebullition site had overall lower concentrations compared to the higher ebullition sites with more intermediate values (< 50 ng_{Hg}/g_{sediment}) starting at 2cm depth, dropping to < 15 ng_{Hg}/g_{sediment} at ~ 26 cm. We found differences

(≥ 50%) in overall Hg contents of surficial sediments. Mercury content positively correlated with total organic C (TOC, $R^2=0.74$) and sulfur (S, $R^2=0.92$) in the highest ebullition site. Mercury content also negatively correlated with dissolved inorganic carbon (DIC) in the highest ebullition site ($R^2=0.71$). These relationships are only seen in the higher ebullition sites, whereas in the lower ebullition sites, Hg correlations to other geochemical data (e.g., TOC, C, nitrogen, S, DIC) were more variable. Our findings imply that (1) processes that enhance CH₄ mobilization affect Hg dynamics in sediment cores, (2) higher ebullitive regions will exhibit elevated Hg concentrations and (3) assessing overall Hg behavior in lakes requires cores in multiple locations.

Acknowledgements

The amount of collaboration on this project was incredible and if not for scientific collaboration, this project would not be what it is. First off, I thank Northern Ecosystem Research for Undergraduate's program director Dr. Ruth Varner for providing me the opportunity to do research at the University of New Hampshire (UNH) and the Abisko Scientific Research Station (ASRS). Secondly, I would like to thank Florencia Meana-Prado and Julia Bryce for their supervision of my understanding of mercury cycles. I also thank Martin Wik and Patrick Crill for teaching me about the Abisko region and Stordalen Mire, assisting in determining sampling locations, and conveying regional geochemical cycles to me in northern Sweden. I thank Joel P. Destasio, Madison J. Halloran and Joel E. Johnson for the help with field sampling and auxiliary data. A thank you goes to Jacob B. Setera for his help in analyzing sediments while I was in Sweden conducting field work. Lastly, I would like to thank Dr. Laura Triplett for supervising my research at Gustavus Adolphus College and ultimately initiating and pushing me to pursue my interest in geology and scientific research. My research at UNH and ASRS was funded by the National Science Foundation.

<u>Table of Contents</u>	<u>Pg.</u>
I. Introduction_____	6
II. Geologic Setting_____	9
III. Methods_____	12
IV. Results_____	14
V. Discussion_____	23
VI. References Cited_____	26
Figure 1 – Abisko Location_____	9
Figure 2 – Regional and local aerial photo of Tornetrask valley Stordalen Mire, respectively_____	11
Figure 3 – Sediment Core Visuals_____	15
Figure 4 – Grain Size Distribution_____	17
Figure 5 – Extractable Hg Content_____	18
Figure 6 – Total Hg Content_____	19
Figure 7 – Methane (CH ₄) Content_____	20
Figure 8 – Total Organic Carbon Content_____	21
Figure 9 – Total Sulfur Content_____	22
Figure 10 – Total Nitrogen Content_____	23

Introduction

Today, sub-Arctic environments are some of the most susceptible to changing climate. Anthropogenic influence is the primary driving factor behind this change. In addition to influencing average global temperatures, humans have also altered the chemical components of atmosphere, ocean and terrestrial environments with potential environmental contaminants. Though remote, the Arctic is not immune to anthropogenic influenced changes. Of all of the atmospherically deposited contaminants, mercury (Hg) is commonly thought to be the largest threat to Arctic environments (Klaminder et al, 2008 and Van Oostdam et al, 1999). The Arctic is particularly important for sequestering Hg, which in some forms is highly toxic to life.

Mercury's potential to contaminate Arctic ecosystems begins at the lowest levels of the food chain with bacteria, Achaea, phytoplankton and algae. Mercury can accumulate in these low level organisms through absorption and consumption of inorganic Hg, however specific processes remain to be determined. Mercury exists as methyl mercury (MeHg) and inorganic Hg in Arctic environments. Both inorganic Hg and MeHg may be assimilated by biota, however, MeHg is more readily biomagnified within the food chain (Douglas et al, 2012; Fitzgerald et al, 2007 and Mason et al, 1996). Mercury accumulation in Arctic sediments increases the 'bioavailability' of inorganic Hg to be methylated by microbial communities and thus increases the likelihood of MeHg accumulation in prey organisms and predators within the food chain (Douglas et al, 2012).

The atmosphere at higher latitudes is a long term Hg scavenger, primarily because of the presence of radioactive halogens. Radioactive halogens favor oxidation of gaseous Hg^0 into Hg species that more easily deposits onto terrestrial landscapes (Steffen et al,

2008 and Klaminder et al, 2008). As more Hg is deposited in the higher latitudes, the Arctic will act as a sink for atmospheric Hg (Ariya et al, 2004). There, most Hg will be converted to MeHg, the form most readily available to organisms (Douglas et al, 2012) and mobilized by sulfate-reducing bacteria (Gilmour et al, 1992). Sulfate-reducing bacterial communities are also thought to produce methane (CH₄). Thus, this study tries to elucidate the links between CH₄ chemical systems and Hg sequestration and possible Hg mobilization.

Sub-Arctic permafrost peatlands serve as good locations in which to study these chemical system links because peatlands sequester large volumes of carbon (C) and Hg (Biester et al, 2003; Bindler et al, 2004 and Martinez-Cortizas et al, 1999). In northern Sweden, permafrost in peatlands is currently thawing (Christensen et al, 2004 and Kokfelt et al. 2010). In these northern environments, Hg is strongly bound to organic matter (Xia et al, 1999). The melting of permafrost peatlands due to climate warming may allow for more organic matter to be susceptible to bacterial breakdown with subsequent release of Hg.

Bacterial breakdown of C to produce CH₄ is a positive feedback contributing to climate warming processes (Walter et al, 2006); warming causes CH₄, which leads to more warming. Methane has a radiative forcing of about 25 times that of carbon dioxide (CO₂) on a 100 year scale (Anthony et al, 2010). Naturally emitted CH₄ has three pathways in which it is released from lake sediments: 1) transport through emergent plants, 2) molecular diffusion and 3) ebullition (bubbling) (Anthony et al, 2010). This study solely investigates the ebullitive pathway as microbial communities in the sediment column are thought to methylate Hg along with production of ebullitive CH₄.

Ultimately, the importance of studying Hg is its potential to be mobilized and likelihood to bioaccumulate in organisms. Climate warming will cause more organic matter

to be broken down via bacterial communities. The increased bacterial activity will lead to more CH₄ release and potential Hg mobilization in lake waters as MeHg. Understanding the link between CH₄ ebullition and Hg sequestration, and potential Hg mobilization, is key to determining Hg dynamics in lake ecosystems.

Geologic Setting

Located roughly 200km north of the Arctic Circle, Abisko, Sweden lies in the discontinuous permafrost zone. The discontinuous permafrost zone is situated between fully frozen permafrost and fully melted peatland sediments. The zone of discontinuous permafrost is situated in the melting margin of permafrost. My study site, Lake Villasjön, is located 11km east of Abisko in Stordalen Mire. This setting is an excellent location for indications regarding the effects of climate change as global warming feedbacks influence permafrost and change can rapidly seen in the Stordalen Mire in the discontinuous permafrost zone.



Figure 1 - Map showing regional location of Abisko in relation to the Arctic Circle

Stordalen Mire is in the Lake Tornetrask glacial valley. Tornetrask valley lies in a structure created by multiple orogenies beginning with the Caledonian. The Caledonian orogeny began in the early Cambrian and occluded in the mid Silurian when the Lapetus Ocean off of Laurentia began to collide with Baltica (Cocks et al, 2005; Corfu et al, 2011; Roberts et al, 2002 and Gee et al, 2010). The Baltican Precambrian complex consists of sandstone, mudstone, conglomerates and dolomite and now is situated on the Sweden/Norway border (Gee et al, 2010 and Lindstrom et al, 1987). The Caledonian orogeny ultimately had three phases; the second and third phases are seen at the Abisko Thrust sheet area. The combined phases of the orogeny creating Tornetrask valley ended around 400Ma as the orogen collapsed (Gee et al, 2010). Rocks occurring in the Abisko thrust sheet include schists, gneisses, quartzite, amphibolites and occasional dolomite/ marble lenses (Corfu et al, 2011; Lindstrom et al, 1987 and Kathol et al, 1987). The Caledonian orogeny drifted north to the polar latitudes in

the early Cenozoic. Being in the polar region, uplift and the cooling climate created an environment of persistent glaciation. With fluctuating uplifts and glacio-fluvial cycles, the stepped landscape of Abisko was turned into one composed of large erosional valleys. Ultimately, Tornetrask valley was created from the erosion of exposed lower resistant rocks and gravitational forces (Lidmar-Bergstrom et al, 2007).

Being in the northern latitudes, glacial activity is prominent in the landscape. Glaciers in this region were cold-based, meaning that they were frozen at the base and exhibit low-energy erosional rates. Deglaciation started around 10,000 yBP with mountain peak ice melting first, producing remnant glaciers in valleys (Stroeven et al, 2002; Kleman et al, 1999; Andre, 2002; Dobinski, 2010 and Snowball, 1996).

As deglaciation isolated remnant glaciers on the slopes, the valley glaciers blocked the valley outlet holding back water (Shemesh et al, 2001 and Grudd et al, 2002). Till was initially deposited but as melting progressed, glacio-lacustran silts were deposited across the valley approximately 9500 – 9000 yBP (Linden et al, 2006 and Barnekow et al, 2000). Since the deglaciation, isostatic rebound has been prominent in the region lifting at estimates of 100 to 400m with the majority of rebound in the first 1000 years preceding deglaciation. Currently, rebound rates are at 4-6mm/yr while erosional rates are 0.2-5mm/kyr indicating continuous evolution of the landscape (Bakkelid et al, 1986 and Andre, 2002).

Around 9000 yBP, strong cooling occurred in the Abisko region and subjected Lake Tornetrask to drop by 400m. Eventually, around 5000 yBP, the Abisko region became a shallow wetland allowing for the first deposition of peat (Kokfelt et al, 2010). Stordalen's environment was then separated from the Tornetrask around 3400 yBP when Holocene

cooling caused reformation of mountain glaciers and further reduction in lake levels (Bigler et al, 2002; Kokfelt et al, 2010; Taylor et al, 2009 and Woo et al, 2006). Vegetation shifted towards graminoids (grasses, rushes, sedges) in the next 1000 years while forests retreated. Permafrost subsequently formed as dry moss and the raised topography allowed ice to form at depths. Frost-heave transitions allowed for palsa formations with underlain discontinuous permafrost (Kokfelt et al, 2010 and Woo et al, 2006).



Figure 2 - (Above) Aerial photo of Abisko and Lake Tornetrask region. (Below) Aerial photo of Stordalen Mire, highlighting the locality of Lake Villasjön within the mire and its local scale within the Lake Tornetrask region. Photos courtesy of Google Maps, 2013.

Lake Villasjön lies in the eastern portion of Stordalen Mire and is approximately 1.5 meters in depth throughout. As Lake Villasjön is underlain with clastic sediment, it is assumed that it is a glacial remnant lake of the most



recent glaciation ~4000yBP. At approximately 365m above sea, it currently sits 11m above the nearby Lake Tornetrask (Wik et al, 2011).

Methods

Sediment cores were taken from Lake Villasjön along a known methane ebullition gradient. The methane ebullition gradient was confirmed by Wik's research in Lake Villasjön (Wik et al, 2011). The methane ebullition gradient was found via floating bubble traps. I analyzed four locations along the methane ebullition gradient with samples from two more locations archived for future analytical work. Four cores were taken at each location to determine CH₄ concentrations, perform CH₄ incubations, analyze carbon, hydrogen, nitrogen and sulfur (CHNS), measure grain size and Hg concentration. Cores were taken using a modified AMS certified hammer corer. Most cores were approximately 30-50cm in depth with some reaching 80cm. Methane concentrations were acquired by taking sediment/gas plugs every 5cm downcore. Samples were placed in nitric acid to stop bacterial activity and head-gas was later sampled for CH₄ analysis by gas chromatography. Methane incubation samples were placed into two vials and put into 20°C and 5°C chambers to record the productivity of bacterial activity. Head-gas volumes were sampled for five consecutive days following core collection and analyzed by gas chromatography. Carbon, hydrogen, nitrogen and sulfur weight percentages were acquired via a Perkin-Elmer CHNS 2400 analyzer. Carbon, hydrogen, nitrogen and sulfur was sampled every 5cm downcore. Grain size samples were taken every 5cm and also at each distinct sediment change. Grain size data were acquired by a Mastersizer Laser particle analyzer. Samples for Hg analysis were taken every 2cm to ensure a high resolution dataset downcore.

To acquire Hg sediment samples, the core was split to minimize the possibility of contamination from the sampling tube. At each sample depth, the surface was also scraped clean with a clean polypropylene tool to reduce contamination. Each Hg sample was stored

in a double plastic baggy until further analysis took place. Samples were freeze-dried to prevent sediment decay. Once freeze-dried, samples from VM1 and VM6 were digested three times to extract Hg from particulates. Cold vapor ICP-MS was used to determine extractable Hg content ($\text{ng}_{\text{Hg}}/\text{g}_{\text{Sediment}}$). Samples from VM2 and VM5 were analyzed via thermal decomposition and cold vapor atomic fluorescence. Data was then plotted against multiple factors (i.e. grain size, CHNS, CH_4) to determine chemical and physical correlations.

Results

All cores taken in Lake Villasjön had approximately 30cm of organic-rich material on top of a clastic layer ~10cm thick. Sediment in the depths of 0-14cm was a very dark black/brown color. Within the 15-30cm depth, the color lightened slightly to brown. From 30cm depth and below, the sediment was gray. The four cores I analyzed ranged in length from 39cm to 44cm: 44cm at VM1, 39cm at VM2, 41cm at VM5 and 40cm at VM6 (Figure 3).

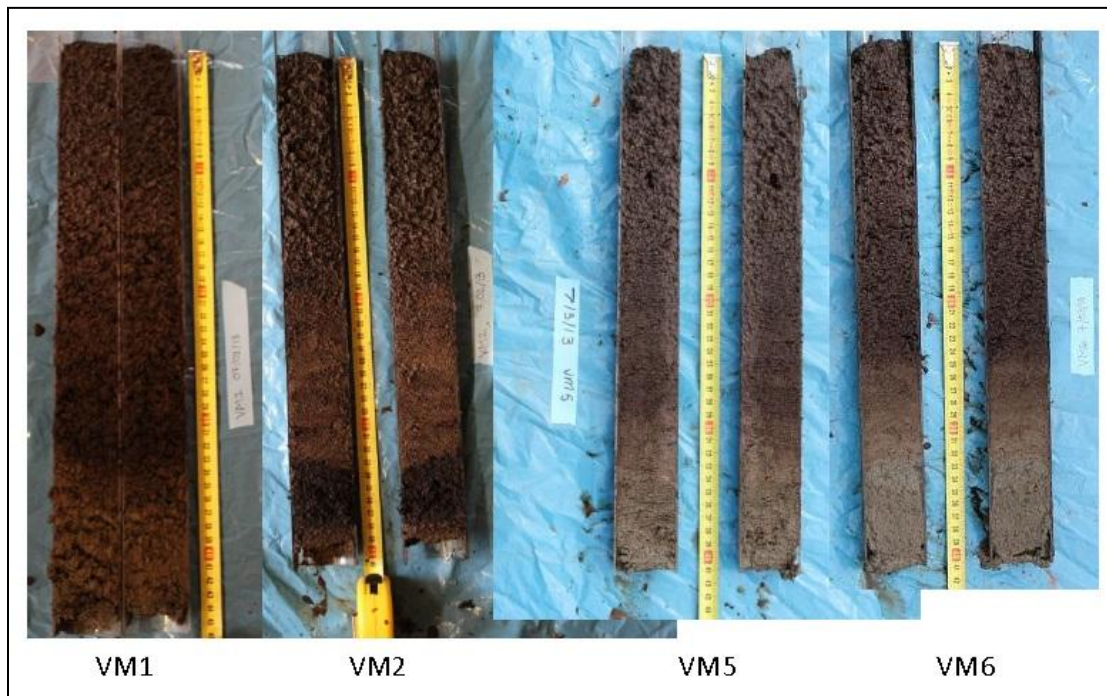


Figure 3 – Sediment cores from transect across an ebullition gradient in Lake Villasjön, Stordalen Mire, Abisko, Sweden. Organic-rich sediment is brown in color and clastic material is gray in color. Note that light settings are different in each photo. Photos courtesy of Florencia Meana-Prado.

Grain size distributions were different throughout each sediment core even while physical appearance differences were subtle. At site VM1, clay content stayed relatively constant through the core, varying from 3.1% to 9.8% in the total grain size distribution percentage. Sand and silt content were about equal in the upper portions of the sediment columns, however volume percentages diverge at 30cm depth as silt content increases to

84% and sand content decreases to 8% of total sediment volume. Sediment at VM2 showed a similar distribution however one major difference is the spike in clay content at 25cm depth to 15.5%. Site VM5 has almost exactly the same grain size distribution as site VM1 with clay content staying low, only ranging from 3.4% to 5.1% of total volume. Sand and silt content within VM5 were similar until a depth of 30cm as silt content increases to 76% and sand content declines to 20% of total sediment volume. VM6 clay content was comparable to the other cores as it was near or below 5% of total sediment volume percent. VM6's silt and sand content diverge in the upper portions from 5cm to 20cm depth (silt at ~33%, sand at ~65%) and then were more similar in the lowest 15cm (Figure 4).

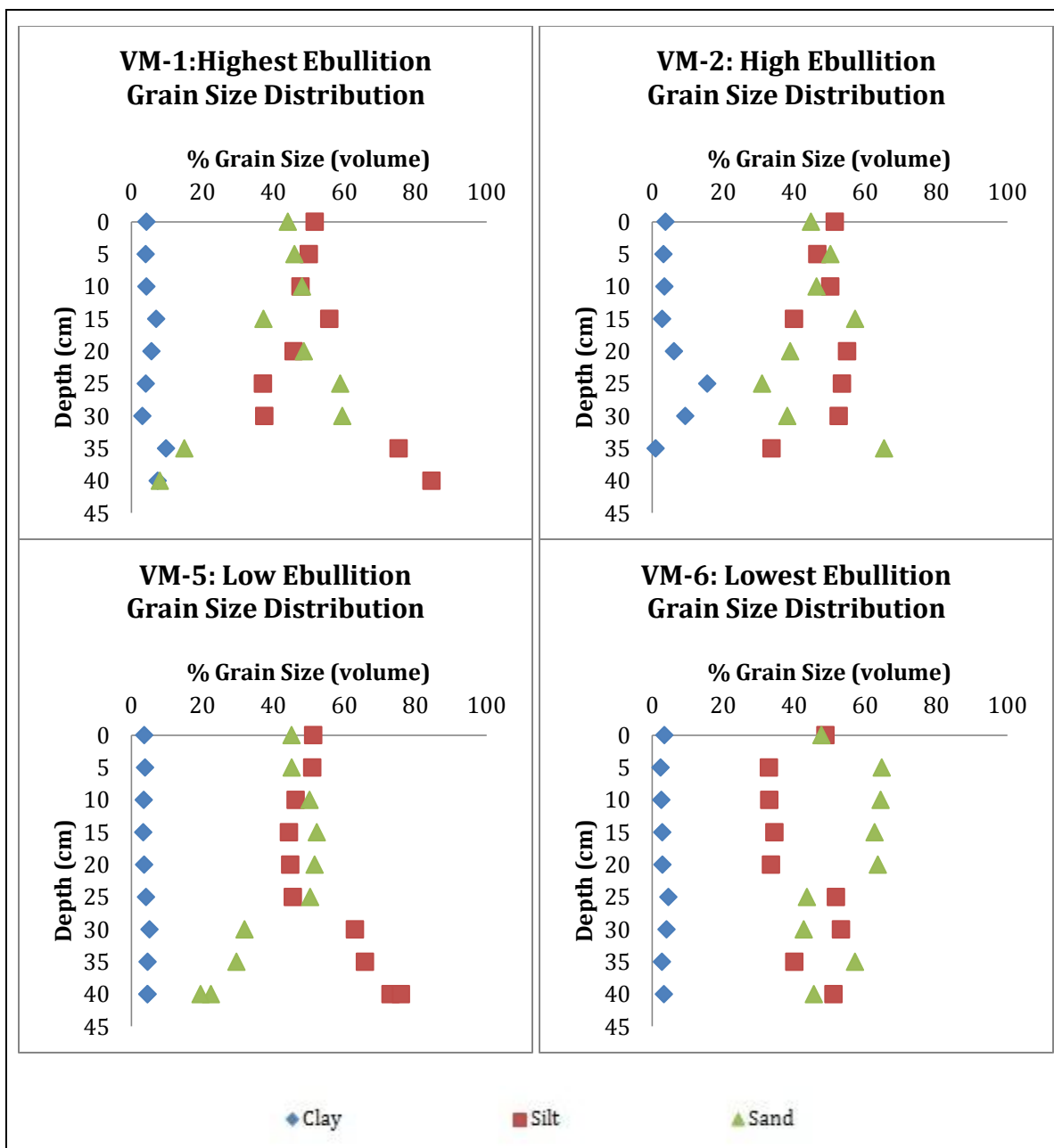


Figure 4 – Grain Size distributions by percent volume at four sites along a known CH₄ ebullition gradient in Lake Villasjön, Stordalen Mire, Abisko, Sweden. VM1 has highest ebullition rates while VM6 exhibits the lowest ebullition rates.

Mercury (Hg) content was measured as total Hg (sites VM2 and VM5) and extractable Hg (sites VM1 and VM6). In both analyses, the site at the higher ebullitive region (sites VM1 and VM2) had elevated Hg content.

In the extractable Hg cores, VM1 had its highest Hg content in the upper portions of the sediment sequence, approximately 72ng/g at

the surface and a peak at 83.1ng/g at a depth of 5cm; Extractable Hg content gradually decreased with depth. This gradual decrease in extractable Hg content is also seen in VM6, though that core has lower initial Hg content of 43ng/g in the upper sediments (Figure 5). The large outlier value of 103.7ng/g in the extractable Hg content of VM6 was likely a result of surface contamination. Statistical standard error for all values was 10.277% and was calculated via averaging error values from sediment standard runs (Figure 5).

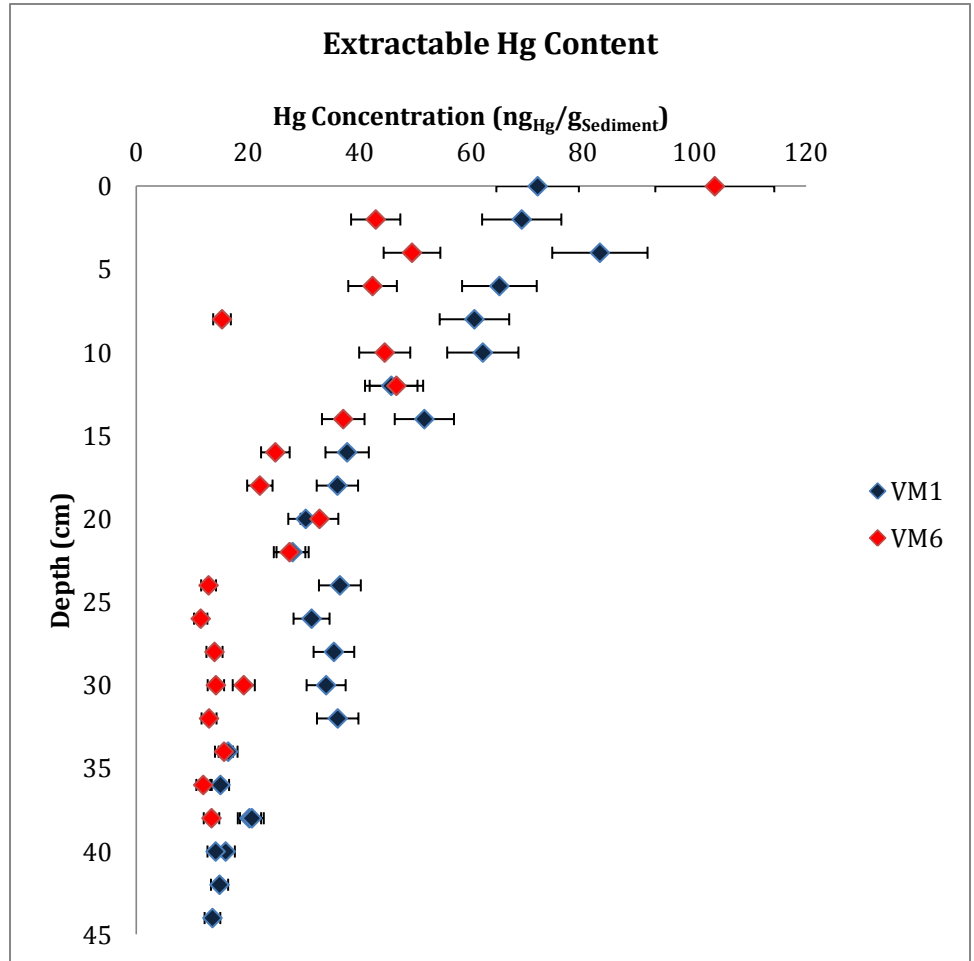


Figure 5 – Comparison of extractable Hg content in sediment cores from sites VM1 and VM6 along a known ebullition gradient in Lake Villasjön, Stordalen Mire, Abisko, Sweden. Error Bars (10.3%) were acquired via calculating error from machinery fluctuation on standard samples. The VM6 surface Hg value is an outlier likely caused from surface contamination. VM1 is located at highest ebullitive region whereas VM6 is located near

Total Hg content exhibited a similar trend to the extractable Hg content. The higher ebullitive site (VM2) had elevated Hg content compared to the lower ebullitive site (VM5). One outlier from the trend of gradual decrease in Hg is at site VM2 at 32cm depth (Figure 6). This elevated

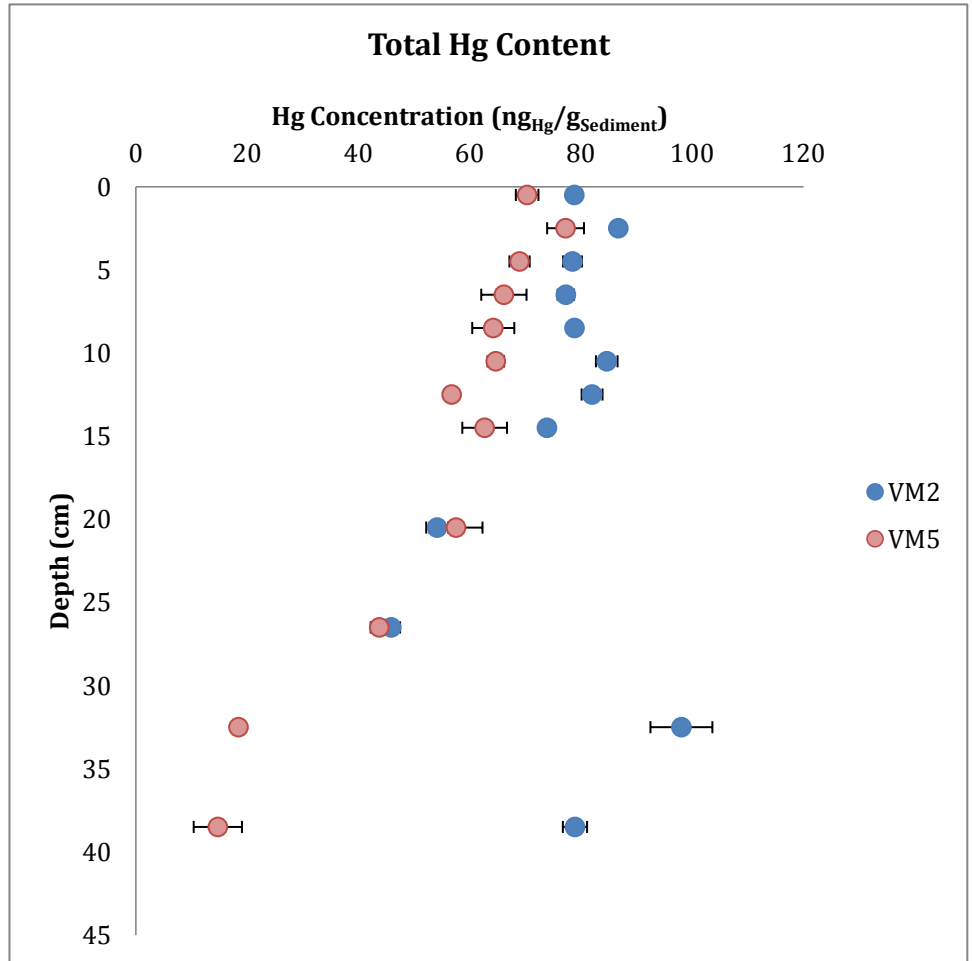


Figure 6 – Total Hg content acquired from sites VM2 and VM5 in Lake Villasjön, Stordalen Mire, Abisko, Sweden. Individual error bars were calculated from triple runs of each sample; Displayed on each data point. Site VM2 is at a known higher ebullitive region whereas site VM5 is at a known lower ebullitive region.

value in Hg content is not seen in the other cores.

Total Hg content was more elevated compared to the extractable Hg content because total Hg data takes into account the particulate material whereas the extractable Hg content only accounts for readily available, digestible Hg content. Standard error bars are shown on Figure 6 and are specific to the individual sample.

Methane content down core varied because each site had different rates of ebullition. Site VM1 had the lowest content of CH₄ with a maximum value of 1269.5 ug CH₄ g⁻¹_{ds} at a depth of 26cm. The trend in all cores for CH₄ content revealed that in the upper portions of core, CH₄ is depleted. It

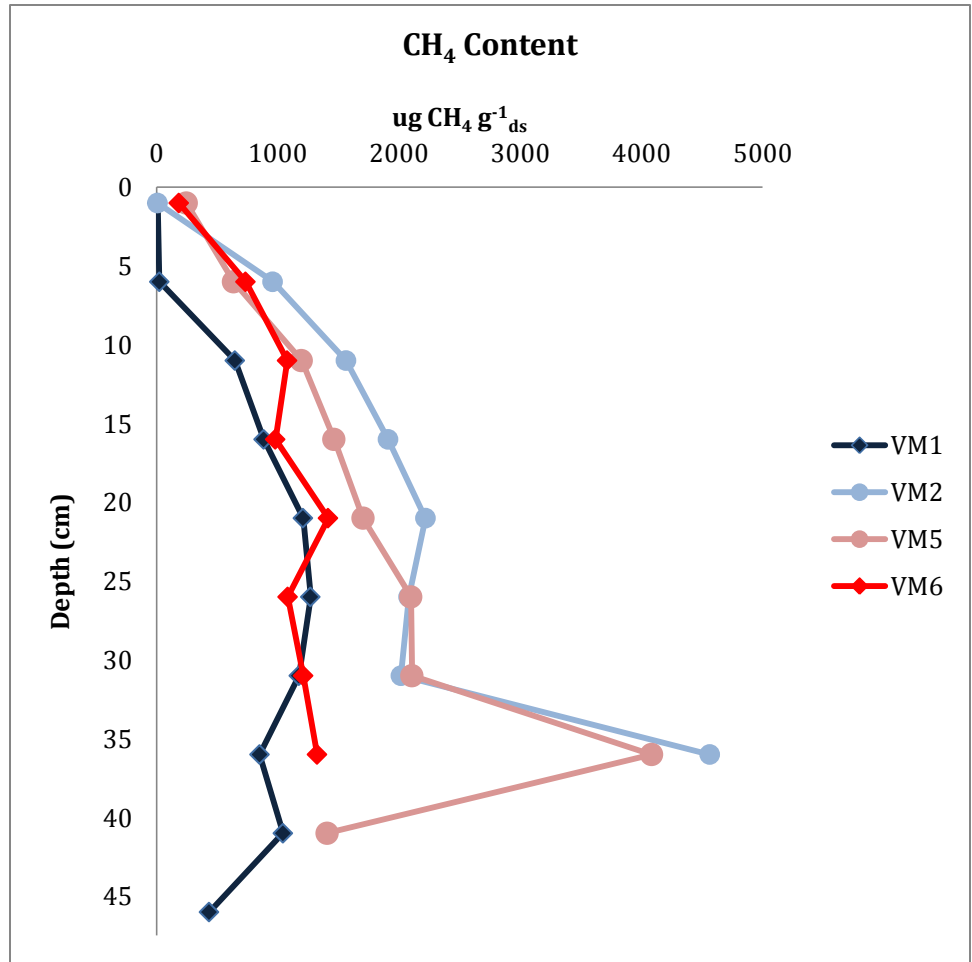
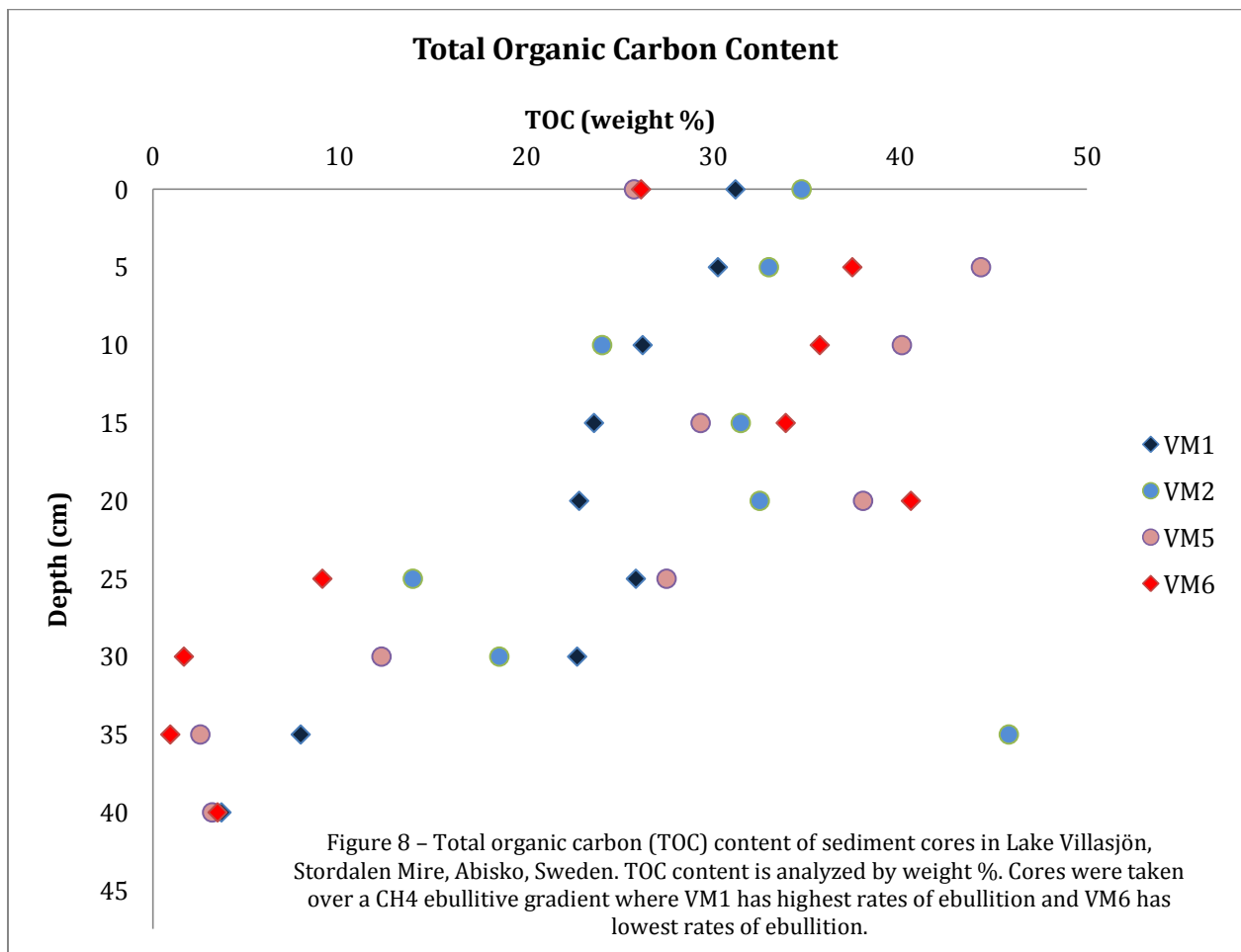


Figure 7 – Graph showing CH₄ content of sediment cores in Lake Villasjön, Stordalen Mire, Abisko, Sweden. CH₄ content was acquired via collecting headspace CH₄ gas at each sediment plug depth. Sediment cores were taken from a known CH₄ ebullitive gradient where VM1 has highest ebullition and VM6 has the lowest ebullition.

increased in CH₄ content going down core to a middle portion and then is depleted again at the base of the core. For example, CH₄ content at site VM1 began with 8 ug CH₄ g⁻¹_{ds} at a depth of 1cm and increased to 1269.5 ug CH₄ g⁻¹_{ds} at depth 26cm and then decreased again to 433.6 ug CH₄ g⁻¹_{ds} at depth 46cm. Sites VM2 and VM5 exhibit a large peak in CH₄ content to approximately ~4000 ug CH₄ g⁻¹_{ds} at 36cm depth. Site VM6 showed the same trend as VM1 but had slightly elevated CH₄ content throughout the whole core (Figure 7).

Further analysis revealed the total organic carbon (TOC), nitrogen (N), and sulfur (S) content within the sediment cores. In almost all cases, levels of TOC, N, and S were elevated in the upper portions of the cores and then decreased with depth. Total organic carbon, N and S were analyzed by calculating weight percent in each sample. TOC in the upper portions of cores varied from ~25 weight % to ~35 weight %. Total organic carbon slightly increased with 5-10cm depth and then diminished at a relatively steady rate to the bottom portions of the cores. Total organic carbon of all cores at 40cm depth was ~3.5 weight %. VM2 showed a max peak, unlike any other cores, at 35cm depth with a TOC weight % of 45.8 (Figure 8).



Sulfur content showed a similar distribution to TOC in all cores. A slight increase in S content occurred at the surface of the core, which then decreased going down core. Contrarily to TOC weight %, total S only had a minor weight % within the sediment core. Values were from 0.47 to 0.92 weight % in the upper 5cm of the cores (Figure 9).

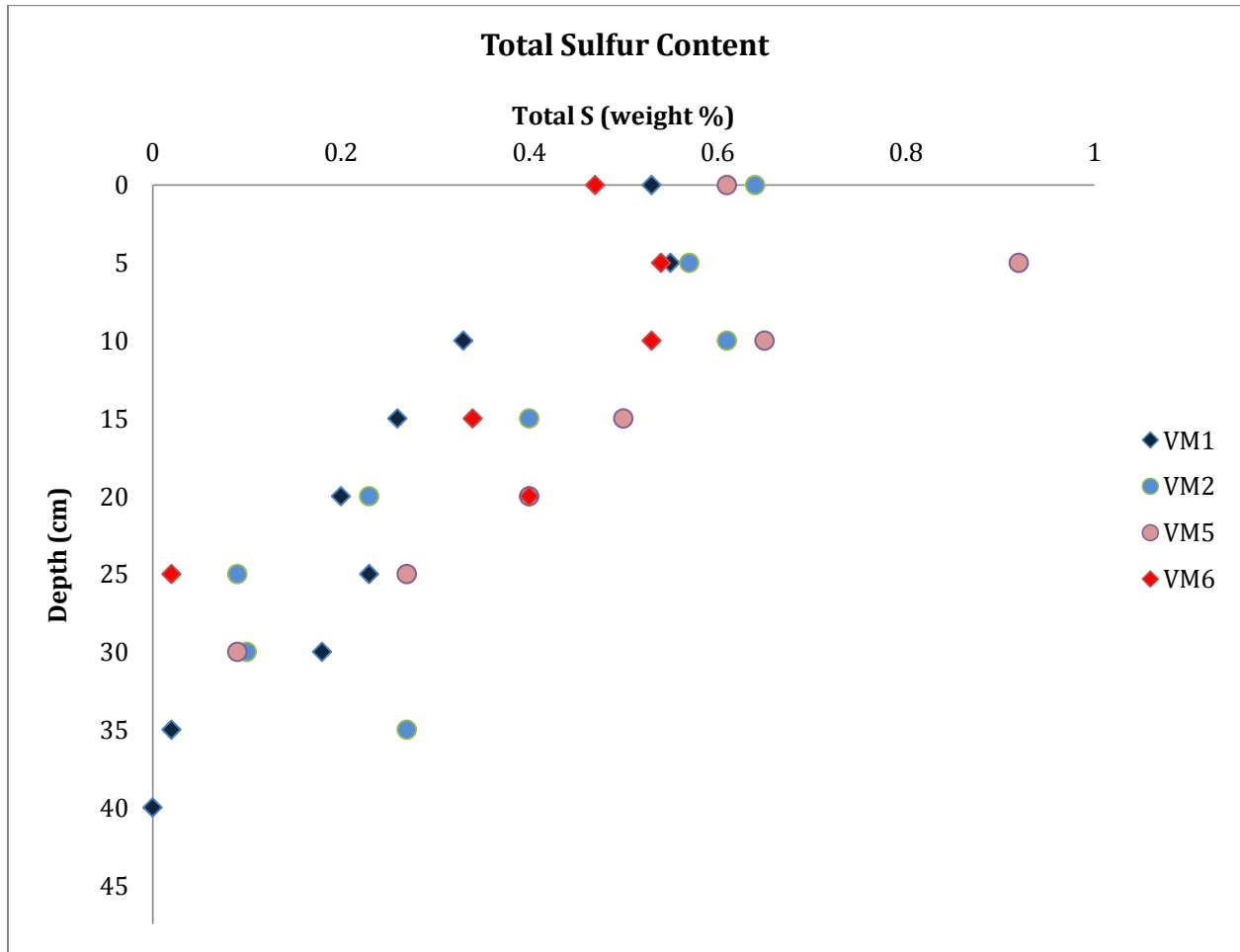


Figure 9 – Total Sulfur content of lake sediment within Lake Villasjön, Stordalen Mire, Abisko, Sweden. Total S content analyzed via weight %. Sediment cores were taken over a CH₄ ebullition gradient where VM1 has highest ebullition rates and VM6 has lowest ebullition rates

Accordingly, nitrogen (N) content showed a similar distribution to TOC and S content down core in all sediment cores. Maximum N content was seen at a depth of 5cm in 3 cores, with the most extreme case VM5 showing a max N weight % value of 2.98. VM6 has a maximum N value at 20cm depth. Sediment at VM2 showed a different trend in the

bottom portion as it decreases to ~1.1 weight % N at depth 25cm and then increases again to ~1.8 weight % N at 35cm depth (Figure 10).

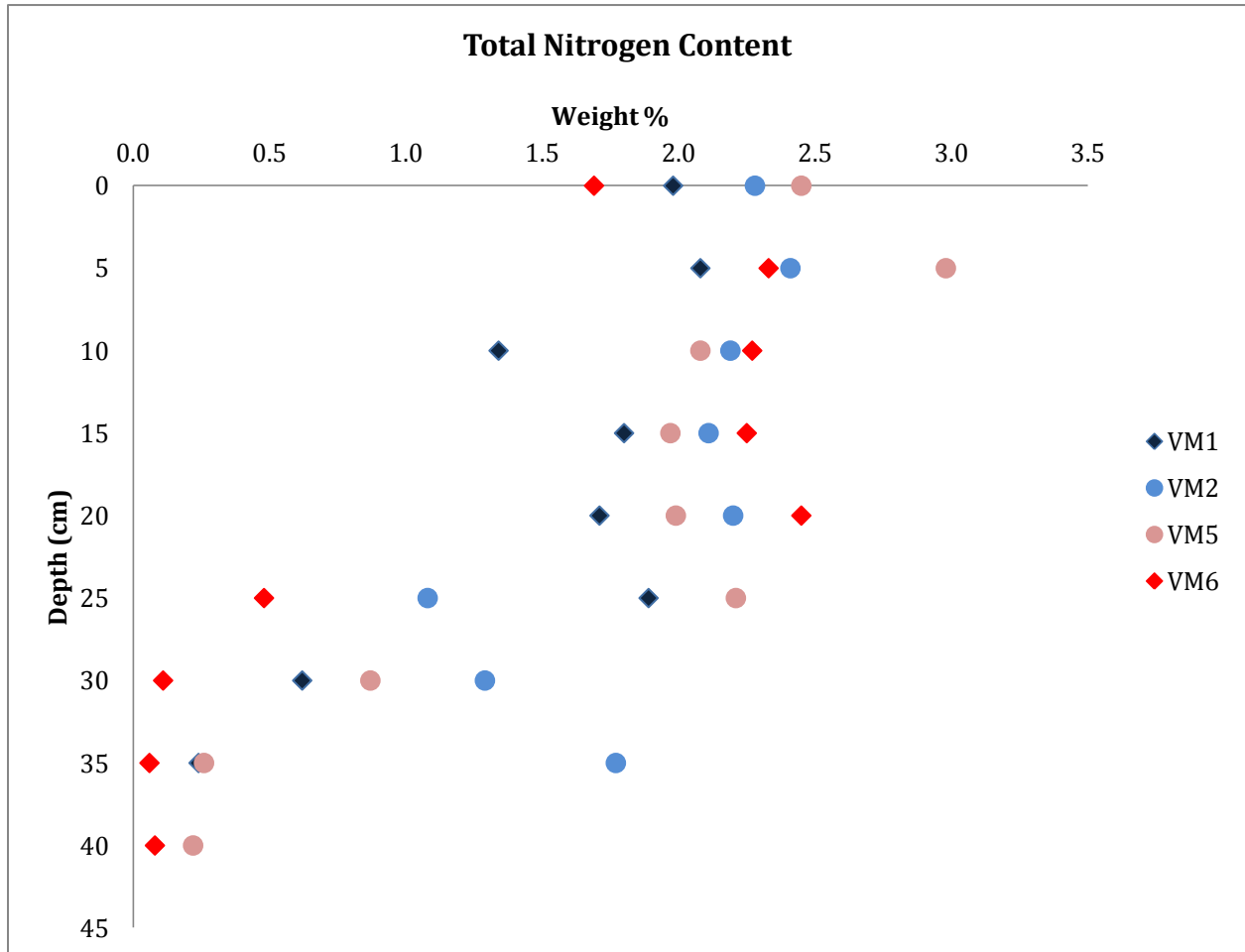


Figure 10 – Graphic showing total nitrogen (N) content within sediments from Lake Villasjön, Stordalen Mire, Abisko, Sweden. Nitrogen content is shown as weight %. Sediment cores were taken over a CH₄ ebullition gradient where VM1 has highest ebullition rates and VM6 has lowest ebullition rates.

Discussion

Elevated Hg concentrations are seen in both of the higher ebullition sites in the VM transect. One possible factor that can be related to these concentration differences is the grain size distribution throughout Lake Villasjön. Three cores show similar sand to silt ratios in the top sediments and changes in deeper sediments. With Hg most prominent in the upper portions of the cores, the upper most portion of grain size percentages are most important.

Lake Villasjön is a closed lake, meaning there are no inputs or outlets of water. There is speculation, although, that there may be groundwater movement from the southeastern portion of Lake Villasjön to the northwestern bay, flowing directly parallel to my chosen transect. Thus when analyzing extractable and total Hg content and its attachment to clastics, if groundwater movement were to sort suspended grain particles, larger grains would be present near VM5 and VM6 and smaller grains would be near VM1 and VM2. Grain size can be directly related to extractable Hg and total Hg because with more surface area to be digested in chemical analysis, Hg content is going to be elevated. Overall with this theory, where there are smaller grains in the sediment column, one would expect higher Hg concentrations and vice-versa.

Keep in mind the difference between extractable and total Hg content relates to the methods in which each were analyzed. Extractable Hg values for VM1 and VM6 were acquired via cold vapor ICP-MS after three digestion processes. Total Hg content values were acquired via thermal decomposition and cold vapor atomic fluorescence. Total Hg content takes into account the Hg content within the clastic (inorganic material) and also the organic material in the sediment, whereas the extractable Hg content only measures

the Hg bound/attached to the clastics and organics. The different methods arose from time constraints within my research program. Also note that I specifically measured bulk Hg, an unknown amount of which may potentially be methylated. Actual methylated Hg values are yet to be determined.

Methane is produced from the breakdown of organics by bacterial activity which feed on nutrients in the organics. The relationship of CH₄ and the primary nutrients (TOC, S and N) can be seen as a direct correlation exists at site VM2 at 35cm depth. TOC, S, and N content increased at 35cm depth, also seen with a rapid spike in CH₄ content at 35cm depth. The increase in nutrients likely supplied the bacterial community with the resources needed to produce more CH₄ than in other depths of the core. This relationship was only seen in the lower portion of cores at VM2 and VM5: in the upper portions of all cores the CH₄ content decreased near the surface. The decrease in CH₄ content near the surface is likely related to the release of CH₄ during ebullition. The upper portion of the sediment column throughout all of Lake Villasjön was unconsolidated and organic rich. Thus the potential for the sediment column to hold the CH₄ in bubble form decreased near the surface. The lower portion of the sediment column was much more compact and consolidated, making a better environment for CH₄ storage. Overtime, CH₄ will travel upward in the sediment column and will ultimately be released through ebullition.

Ultimately, elevated Hg content was seen where there was an abundance of primary nutrients. The elevated Hg content was likely from the strong binding of Hg to organic material. This correlation was seen best in the highest ebullition site VM1. Hg positively correlates with TOC and Total Sulfur in VM1 with R² values of 0.74 and 0.92, respectively. Methogenesis of CH₄ and Hg methylation likely correspond with one another as

correlations with TOC and Total Sulfur were highest in the VM1 site. Where more ebullition was occurring, one can expect more bacterial activity. With more bacterial activity, more methylation of Hg was likely to occur and ultimately elevate bulk Hg concentrations in the sediment column.

Overall, the findings imply that (1) processes that enhance CH₄ mobilization may also affect Hg dynamics in sediment cores and (2) assessing overall Hg behavior in lakes requires cores in multiple locations.

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