

1 **A multi-proxy palaeolimnological study to reconstruct the evolution of a coastal**
2 **brackish lake (Lough Furnace, Ireland) during the late Holocene.**

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11 **Abstract** - This study examines the evolution of Lough Furnace, a coastal brackish lake in the west
12 of Ireland, using high-resolution sensors in the water column and palaeolimnological examination
13 of the sediment archive. Palaeoenvironmental reconstructions suggest that meromixis formed as a
14 result of sea level rise prior to ca. 4,000 cal. yr BP. Increased seawater inflow has progressively led
15 to permanent water stratification, which caused the onset of anoxia, making the monimolimnion a
16 harsh environment for biological life. Diatom floristic interpretations suggest a progressive upcore
17 increase in salinity, which is paralleled by a reduction in cladocera remains. Diagenetic processes
18 have not altered the sediment organic matter signature. Organic matter mainly derives from
19 freshwater DOC and appears to be linked to the presence of peat bogs in the catchment as
20 confirmed by the C/N ratio. Upcore variations in the C/N ratio with a ca. 800-year periodicity have
21 been interpreted as the result of alternating dry and wet climatic phases during the late Holocene,
22 which appear synchronous with the NAO and long-term solar cycles. The current hydrology is
23 largely controlled by freshwater inflow, which determines permanent meromictic conditions.
24 Overturns are rare, requiring a specific combination of factors such as exceptionally dry and warm
25 summers followed by cool autumns. According to the climate projections for the next century in
26 Ireland, permanent meromictic conditions will probably continue.

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1. Introduction

Coastal lagoons formed at the land-sea interface are rare habitats representing only 5.3% of European coasts (Barnes, 1980). Coastal lagoons are common around the shores of the Baltic, Mediterranean and the Black Sea (Doody, 2005), whilst on the northwest European Atlantic they are naturally rare (Barnes, 1989; Joint Nature Conservation Committee, 1996).

The land-sea interface is an exceptionally dynamic zone that shows strong non-linear physical and biological forces from both terrestrial and marine environments (Talley *et al.*, 2003). Lagoons are paralic systems (Guelorget & Perthuisot, 1992), characterised by marine, brackish and freshwater deposits. These environments are generally unstable and susceptible to variations in hydrological components making them the most changeable and vulnerable environments on Earth (Viaroli *et al.*, 2007). The main natural hydrological components include droughts, flooding, storms and sea-level rise (Kennish & Paerl, 2010), all of which are likely to be impacted by increased global warming (IPCC, 2007; Dunne *et al.*, 2008). The presence of both fresh and marine water combined with lagoon geomorphology may lead to the development of ectogenic meromictic conditions, where an external source of saline water into a fresh water lake restricts water column circulation to a surface layer (Walker & Likens, 1975; Hakala, 2004). Salinity is the primary stratification driver and temperature and thermal circulation processes are impeded. Therefore, overturns are limited to the top layer (*i.e.* mixolimnion), whilst the bottom of the lake (*i.e.* monimolimnion) can develop anoxia that represents a threat to aquatic life.

Climate fluctuations have occurred over different time-scales, spanning decades to millennia. Lagoons are generally short-lived coastal features formed during the Holocene (Kjerfve, 1986) and represent unique habitats vulnerable to climate change. In particular they are endangered

52 by progressive sea level rise, which has been recorded over the Holocene and is expected to
53 continue into the future (IPCC, 2007; Bates *et al.*, 2008; Dunne *et al.*, 2008). Palaeoclimatic records
54 suggest the existence of multiple periods of rapid climate change during the Holocene in North
55 Atlantic Ocean land masses (Mayewski *et al.* 2004; Wanner *et al.*, 2008). A transition to generally
56 warm but relatively unstable conditions characterised the late Holocene (Moros *et al.*, 2004 (Turney
57 *et al.*, 2006; Swindles *et al.*, 2010) that have been associated with solar variability (Mauquoy *et al.*,
58 2008), changes in the strength of the thermohaline circulation (Bianchi & McCave, 1999) and the
59 North Atlantic Oscillation (NAO) (Nesje & Dahl, 2003; Trouet *et al.*, 2009). Changes in relative sea
60 level (RSL) are part of a complex pattern involving seawater volume and mass change, tectonics
61 and dynamic changes due to external forcing exerted on the sea surface (Chao *et al.*, 2002; Mörner,
62 2006) while Glacio-Isostatic Adjustment (GIA) also played an important role (Horton 2006;
63 Miettinen *et al.*, 2007). A strong north-south trend in glacio-isostatic loading is evident in the
64 British Isles, where uplift is evident in central and western Scotland and Northern Ireland and is
65 coincident with submergence in southwest England and southern Ireland (e.g. Shennan & Horton,
66 2002; Roberts *et al.*, 2006; Smith *et al.*, 2006; Gehrels, 2010). The extent to which globally
67 averaged sea levels have risen over the Holocene is unclear (Edwards, 2006). A number of global
68 (Klemann & Wolf, 2007; NOAA, 2012) and local (e.g. Tushingham & Peltier, 1992; Shennan &
69 Horton, 2002; Brooks & Edwards, 2006; Vink *et al.*, 2007; Engelhart & Horton, 2011;) RSL
70 databases of the Holocene have been collated. Brooks & Edwards (2006) estimate a rise from ca. -6
71 m ca. 7,000 cal. yr BP to ca. -2 m approximately 5,000 cal. yr BP for Ireland and British Isles.
72 However, despite a range of quantitative relative sea level data (e.g. Sinnott, 1999; Devoy *et al.*,
73 2006), precise and reliable information about changes since the early Holocene for parts of the Irish
74 coastline remains incomplete (Brooks *et al.*, 2008).

75 A number of ecological studies have been conducted and published on Irish lagoons (e.g.
76 Parker, 1977; Parker & West, 1979; Pybus & Pybus, 1980; Healy *et al.*, 1982; Norton & Healy,

77 1984; Healy, 1997; Healy, 1999a, b; Oliver, 2005; Roden & Oliver, 2010). However, these
78 inventorial surveys provided only a snapshot examination of physico-chemical and biological data.
79 Long-term data sets are needed to aid in the understanding of coastal ecosystems (Talley *et al.*,
80 2003). For almost all lakes, palaeolimnology is the only means of obtaining data on past trends
81 (Battarbee, 1999). Coastal lagoons provide a suitable sedimentary environment for palaeoecological
82 study because they are relatively sheltered from wave and tide exposure (Bennion & Battarbee,
83 2007) and are influenced by changes affecting both freshwater and marine environments (Ryves *et*
84 *al.*, 2004). Therefore, sediments contain the ontogeny (*i.e.* development through time) of basins
85 themselves. Sediments also record information on atmospheric deposition and its terrestrial
86 watershed (O'Sullivan, 2004) as well as changes derived from the marine environment (e.g. marine
87 transgression). Palaeolimnology of coastal lakes also assists in determining the presence of marine
88 or non-marine deposition, which in turn can be potentially linked back to climate patterns
89 (Battarbee *et al.*, 2002; Lotter, 2005). Many palaeoecological studies have successfully
90 reconstructed the evolution of coastal lagoons by combining a number of proxies (e.g. Müller &
91 Mathesius, 1999; Weckström, 2006; Cearreta *et al.*, 2007; Blázquez & User, 2010). However, only
92 a very small number of studies have been conducted on coastal lagoons in Ireland (Buzer, 1981;
93 Holmes *et al.*, 2007).

94 In a European context lagoons are recognised as vulnerable systems (Airoldi & Beck, 2007).
95 The Habitats Directive (92/43/EEC) aims to maintain biodiversity through the conservation of
96 natural habitats and by defining Special Areas of Conservation (SACs). Additionally, lagoons are
97 included in the Water Framework Directive (WFD) (2000/60/EC) as priority habitats for
98 conservation (Annex I). The Directive aims to achieve good ecological status in all relevant waters
99 between 2015 and 2027 and requires baselines of reference conditions prior to anthropogenic
00 impact to be identified. Moreover, the identification of natural background conditions and longer-
01 term natural variability is often essential for conducting informed lake management programs

02 (Smol, 2008). In the absence of long-term data, and considering the dynamic nature of coastal
03 lagoons, palaeolimnological reconstructions offer perhaps the only source of information regarding
04 baseline reference conditions.

05 The aim of this paper is to reconstruct the evolution of Lough Furnace, a brackish coastal
06 lagoon on the western coast of Ireland, during the late Holocene using a multiproxy
07 palaeolimnological approach. Particular attention was focussed on the reconstruction of meromixis
08 formation as a result of the relative sea level rise. The palaeolimnological investigation of this
09 hydrologically complex system was furthermore augmented using high-resolution time series data
10 on salinity, temperature and dissolved oxygen measured through the water column over a two-year
11 period, to better understand modern and future system dynamics. The influence of climate
12 variability on the palaeohydrology of Furnace and the future hydrological scenarios given the
13 predicted global warming make this study highly relevant.

14

15 **2. The study area**

16 Lough Furnace is located in the Burrishoole catchment (N 53°55'22", W 9°34'20") at the north-
17 eastern corner of the Clew Bay (northwest Ireland) (Figure 1a). The geology of the catchment is
18 dominated by metamorphic rocks of late Precambrian age (Long *et al.*, 1992), represented by
19 schists, gneiss and quartzites (Whelan *et al.*, 1998). Catchment land cover consists of peat bog and
20 forestry (Bossard *et al.*, 2000). The upper part of the catchment is considered to be one of the best
21 examples of an active blanket bog (NPWS, 2001), which represents 71% of the catchment today
22 (Bossard *et al.*, 2000) while forest plantations occupy 23% of the catchment (Forest Inventory and
23 Planning System, unpublished data). Pollen records from the Late Glacial suggest early
24 development of woodland, its subsequent decline and then the development of peat and bog ca.
25 5,000 cal. yr BP (Browne, 1986). Anthropogenic influences of fire and grazing probably accelerated
26 deforestation (Coxon *et al.*, 1991). From the 13th to the 18th centuries the Burrishoole Channel was

27 an important port, which facilitated trade with northern Mayo through the Nephin Beg mountains
28 (Synge, 1963). Since the mid-1950s the Burrishoole catchment has been an important site of
29 fisheries research, with fish census and catchment monitoring being conducted by the Marine
30 Institute.

31 Furnace is part of the Clew Bay Complex SAC (site code 1482) and is described as a good
32 example of a deep, stratified, saline coastal lake in a very natural state (NPWS, 2001). The lake has
33 a surface area of 141 ha, a maximum depth of 21 m, a diameter of 1850 m, and it is approximately
34 800 m wide on average. Furnace is located to the south of the catchment (Figure 1b) and receives
35 highly coloured drainage waters ($52\text{-}112\text{ mg l}^{-1}\text{ PtCo}$; Marine Institute, unpublished data) that
36 strongly limit the photic zone depth (ca. 1.7 m). The Burrishoole Channel connects Furnace to the
37 sea in Clew Bay and semidiurnal tides flow northward into the lake.

38 Furnace is a permanently meromictic lake and only a single water column overturn is known
39 to have occurred on the 12th September 1995 (Salmon Research Agency, 1995). This caused a
40 major fish kill that wiped out trout and char that were caged in the lake at the time. Measurements
41 of oxygen revealed an epilimnion depleted in oxygen ($<2\text{ mg l}^{-1}\text{ DO}$) whilst the temperature profile
42 showed isothermal conditions (ca. 17°C). The summer of 1995 was exceptionally hot with average
43 air temperatures of ca. 17°C and low rainfall (175 mm) (Marine Institute, unpublished data). The
44 water temperature within the hypolimnion from June to August 1995 was $4\text{-}5^{\circ}\text{C}$ warmer than any
45 previously recorded measurements (Salmon Research Agency, 1995). Due to the negligible input of
46 freshwater in the summer of 1995, the halocline progressively disappeared and stratification was
47 maintained by the thermocline only (Salmon Research Agency, 1995). As the weather cooled in the
48 early autumn, isothermal conditions were mainly responsible for the mixing event, which moved
49 anoxic water upward from the monimolimnion. An examination of summer rainfall records (*i.e.* the
50 sum of precipitation in June, July and August) from 1960 (Marine Institute, unpublished data)
51 suggests that rainfall in 1976, 1983 and 2006 were lower than 1995, but no overturns were

52 documented. Therefore, it is likely that the combination of the disappearance of both halocline and
53 thermocline together with the contribution of a northerly wind caused the 1995 event (Russell
54 Poole, pers. comm.). Similar lagoon dynamics were documented by Lamont *et al.*, (2004), who
55 found that the upwelling of anoxic water depends on the coincidence of strong winds and low levels
56 of precipitation, which results in a weak salinity stratification. The fish kill associated with the 1995
57 overturn was probably due to the upwelling of hydrogen sulphide rather than anoxic waters. The
58 permanent or extended stratification of Furnace causes the accumulation of H₂S and its lethal
59 effects on fish are known (e.g. Lamont *et al.*, 2004; Luther *et al.*, 2004).

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62 **3. Methods**

63 ***3.1 Monitoring water quality***

64 The water column in Furnace was monitored by the Marine Institute by means of an Automatic
65 Water Quality Monitoring Station (AWQMS) (Lakeland Instrumentation) (Figure 1c). A Datasonde
66 DSX5 (Hydrolab OTT) with temperature, dissolved oxygen and conductivity sensors collected
67 water column measurements from 0 m to 13 m depth. Data were recorded at two minute intervals
68 by a Campbell Scientific CR1000 datalogger, with an average of 0.17 m between measurement
69 intervals (depending on weather conditions). A backup and continuous temperature record was also
70 collected using tidbits (Onset). Data were collated for the period January 2009 to December 2010.
71 Hydrological data analysis involved the comparison of in-lake parameters with climatic data.
72 Rainfall, air temperature and wind speed data recorded at the Furnace meteorological station were
73 provided by the Marine Institute together with Furnace and Feeagh water level (OTT water level
74 recorders) and tidal data recorded by tidal gauges at Belmullet (Co. Mayo). Plots of salinity,
75 temperature dissolved oxygen (DO) were constructed using DPlot 2.3.0.7 (Hyde Soft Computing).
76 The Wedderburn number (Stevens & Lawrence, 1997), an index for determining lake stability, was

77 calculated using lake bathymetry, water salinity, water temperature and wind speed data by means
78 of Lake Analyzer Web (Read *et al.*, 2011).

79

80 **3.2 Sediment core sampling**

81 A 146 cm sediment core was collected from the central part of Furnace (11 m depth; Figure 1c) in
82 the winter 2008 using a rodless piston corer (Chambers & Cameron, 2001). The sediment was
83 extruded vertically and subsampled at 1 cm resolution.

84

85 *3.2.1 Chronology*

86 The sediment core was dated using both radiometric (^{210}Pb , ^{137}Cs and ^{241}Am) and AMS radiocarbon
87 techniques. Radiometric analysis was performed at the Bloomsbury Environmental Isotope Facility
88 (BEIF) at University College London (UCL). Samples were analysed using an ORTEC HPGe GWL
89 detector. Dates were determined using the Constant Rate Supply (CRS) model (Krishnaswami *et*
90 *al.*, 1971; Appleby & Oldfield, 1978), whilst sediment accumulation rates (SARs) were calculated
91 using unsupported ^{210}Pb and sediment density data and were expressed both as $\text{g cm}^{-2} \text{y}^{-1}$ and cm yr^{-1} .

93 Accelerated mass spectrometry (AMS) radiocarbon analysis was performed at the
94 CHRONO Centre, Queen's University Belfast (QUB). Four bulk sediment samples were selected in
95 the absence of macrofossils. Calibration of radiocarbon dates was conducted using INTCAL09
96 (Reimer *et al.*, 2009) by means of Calib 6.0 software (Stuiver *et al.*, 2005). Two sigma (2σ) age
97 ranges (95.4% probability) were utilised (Björck & Wohlfarth, 2001). Dates obtained from the two
98 different dating techniques were matched using a linear regression and estimated dates through the
99 sediment core were calculated.

00

01 *3.2.2 Lithostratigraphy, stable isotopes and geochemistry*

02 After a preliminary visual inspection in order to identify variations in the lithological composition,
03 organic matter and carbonate content were determined as loss on ignition (*i.e.* LOI₅₅₀ and LOI₉₅₀)
04 according to Heiri *et al.* (2001).

05 Carbon and nitrogen stable isotopes sample preparation was based on the combination of
06 methods proposed by Talbot (2001) and Wolfe (2001). Analysis was carried out at the
07 Geochemistry Laboratory, Trinity College Dublin (TCD) with a Thermo Delta Continuous Flow
08 Isotope Ratio Mass Spectrometer (CF-IRMS) using International Atomic Energy Authority
09 reference standards (IAEA-NO3 for nitrogen, USGS-24 and IAEA-CH-6 for carbon). Elemental
10 weight percentage composition of carbon and nitrogen was used to calculate the C/N ratio. Stable
11 isotopes results are expressed using the delta notation for carbon ($\delta^{13}\text{C}$) and nitrogen ($\delta^{15}\text{N}$). A
12 linear correlation was conducted between $\delta^{15}\text{N}$ and the C/N ratio as well as $\delta^{15}\text{N}$ and nitrogen (%)
13 using SigmaPlot 11.0 (Systat Software 2008) to test for the presence of sediment diagenesis
14 (Thornton & Mc Manus, 1994). To support diatom inferred palaeosalinity (see below), salinity
15 reconstruction was additionally derived from the $\delta^{13}\text{C}$ record according to the equation [*i.e.* salinity
16 ‰ = $(\delta^{13}\text{C}+30.5)/0.54$] proposed by Emeis *et al.* (2003). The authors examined the relationship
17 between $\delta^{13}\text{C}$ and salinity and proposed that organic matter stable carbon isotopes could be used to
18 detect and quantify salinity changes in surface water when fossil diatom assemblages are not well
19 preserved.

20 Geochemical analysis (*i.e.* Fe, Mn, Cu, Mg) was carried out at Dundalk Institute of
21 Technology using Flame Atomic Absorption Spectroscopy (FL-AAS) following US EPA method
22 3051a for sample preparation. Results are expressed as mg g⁻¹ dry weight (DW).

23

24 3.2.3 Fossil diatoms

25 Fossil diatoms were prepared and concentrations determined according to Battarbee *et al.* (2001).
26 Slides were prepared using Naphrax[®] as a mounting medium. Diatom identification and

27 enumeration was carried out using a Leica DME microscope at 1000x magnification. Taxonomic
28 identification was achieved mainly according to Snoeijs' series (Snoeijs, 1993; Snoeijs & Vilbaste,
29 1994; Snoeijs & Potapova, 1995; Snoeijs & Kasperovičienė, 1996; Snoeijs & Balashova, 1998) and
30 Krammer & Lange-Bertalot (1986-1991) floras. A total of 66 taxa had abundances >1% and
31 occurred in more than two samples (see supplementary data). The diatom morphological dissolution
32 index (F index) (Flower & Lokhoshway, 1993; Ryves *et al.*, 2006) was calculated to quantify
33 frustule dissolution. Diatom taxa were divided into groups according to their salinity tolerance and
34 their habitat. Zonation of the diatom data was performed with constrained cluster analysis, CONISS
35 method using the Psimpoll 4.27 software (Bennett, 2008). The number of significant zones was then
36 identified by applying the broken-stick model (Bennett, 1996). A diatom conductivity transfer
37 function was applied using the European Diatom Database (EDDI, 2001). Diatom-inferred
38 conductivity ($\log \mu\text{S cm}^{-1}$) reconstructions were carried out using the weighted average (WA) and
39 locally weighted-weighted averaging (LWWA) numerical methods. Results were also converted to
40 salinity according to the equation: $\text{salinity } (\text{‰}) = 0.67 \times 10^{-3} \times \text{conductivity } (\mu\text{S cm}^{-1})$ (Pawlowicz,
41 2008; Pieters & Lawrence, 2009). The modern analogue technique (MAT) (Overpeck *et al.*, 1985)
42 was run to ascertain the ratio of analogues between fossil and modern datasets.

43 44 *3.2.4 Fossil cladocera and sponge spicules*

45 Fossil cladocera sample preparation followed Frey (1986). Because of the low abundance of
46 cladocera remains, results are expressed as total number of fragments g^{-1} wet weight (WW). Sponge
47 spicules were encountered during cladocera counts and expressed as spicules g^{-1} WW. No detailed
48 taxonomic classification was conducted.

49

50 **4. Results**

51 *4.1 Modern hydrology*

52 Marked variations in water levels in Furnace are evident (Figure 2a). The total water level
53 amplitude was approximately 1.4 m with a minimum of 0.6 m and a maximum of 2.0 m. Variations
54 in water level are the result of both tidal inflow from the south and inflow from the upstream lake to
55 the north along with some short tributary streams flowing directly into Furnace. Water level
56 changes in Furnace with fortnightly periodicity imposed by tides, which are estimated to intrude in
57 a volume equivalent to 6-25% of the total lake volume each fortnight (Parker, 1977). Furnace water
58 level maxima follow a similar pattern to spring high tides with time lags evident. This could be the
59 result of the presence of the Burrishoole channel, which delays tidal inflow into the lake.
60 Similarities with the water level in the upstream lake Feeagh are evident. Water level in Feeagh is
61 determined by rainfall levels and increases in the Feeagh level automatically creates a greater
62 freshwater inflow into Furnace. Rainfall was higher in 2009 than in 2010 (1794 mm and 1222 mm,
63 respectively) (Figure 2b). A flash-flood in the Burrishoole catchment occurred on the 2nd July 2009
64 when 67 mm of rainfall fell in less than 24 hours. Excessive rainfall led to increased water levels in
65 Feeagh that subsequently led to a rise in water levels in the downstream Furnace. Additionally,
66 increased rainfall between August 11th and the 8th of September (289 mm) determined a water level
67 increase of ca. 0.6 m in Feeagh, which led to an increase (ca. 1.1 m) in Furnace. Similar patterns
68 were evident in 2010.

69 The salinity profile confirms Furnace is a meromictic lake (Figure 2c). The profile is
70 characterised by the presence of a denser and more saline permanent monimolimnion. The surface
71 water salinity is generally below 5‰ whilst the monimolimnion is ca. 18‰. The mixolimnion
72 generally covers the top 4 m of the water column and it deepens mainly as a result of increased
73 precipitation. Differences in rainfall patterns between 2009 and 2010 appear to determine the
74 variations in the mixolimnion depth. In fact, reduced precipitation in 2010 results in a reduced
75 mixolimnion depth, which is generally above 3 m with the exception of October 2010. Higher
76 surface water salinity is also evident and freshwater conditions (<0.5‰) only appeared in October

77 as a result of increased rainfall in September 2010.

78 During 2009 temperatures of ca. 12°C are evident in the monimolimnion below 6 m depth
79 (Figure 2f), whilst surface water is influenced by seasonal air temperature. Winter (January to mid
80 March) lowers surface water temperature (3°C at 2 m depth). Isothermal conditions characterise
81 May, with temperatures in the region of 12°C through the water column, and the highest
82 temperatures at the water column top are reached from mid-June to August (ca. 16-20°C). From
83 September to the end of the year surface water temperatures gradually decrease reaching ca. 4°C at
84 the end of December. A similar pattern is evident for 2010 with temperatures gradually increasing
85 in March leading to isothermal conditions in April. Warming of the mixolimnion gradually occurs
86 from mid April to September and highest temperatures (19°C) were recorded in June. Reduced
87 precipitation and highest lake water salinity determined an increase in bottom water temperatures
88 from ca. 8°C in May to ca. 13°C in July.

89 Dissolved oxygen (DO) concentrations are closely linked to salinity stratification, which
90 determines anoxia of the bottom water (Figure 2e). In 2009 DO concentrations were <1% below ca.
91 6 m depth showing anoxic conditions. In contrast, surface water oxygen concentrations were
92 generally over 75% DO and over-saturated conditions to 120% DO are evident in spring and
93 summer. Oxygenation of the monimolimnion is evident in 2010. Waters up to 75% DO replace
94 anoxic conditions from March to July, when oxygen levels rapidly decline and anoxic conditions
95 appear again below 5 m depth. At the surface over-saturated conditions are evident from January to
96 September 2010. This is coincident with lower rainfall and a shallower mixolimnion.

97 The Wedderburn number (W) is a parameter used to estimate the internal response of a lake
98 to wind forcing helping to determine the probability that upwelling occurs. W values greater than 0
99 indicate that upwelling does not occur, whilst values lower than 0 suggest that upwelling is likely.
00 The W number was calculated for Furnace during 2009 and 2010 (Figure 2f) and results are
01 fragmentary due to the absence of data. High W values are mostly evident in January, February,

02 September and December 2009 and coincide with the mixolimnion deepening. W is closer to 0 in
03 July and August 2009. Lower values around 0 are evident in 2010 with the exception of few peaks
04 between January and March.

05

06 *4.2 Sediment Core Chronology*

07 The equilibrium between the activity of total and supported ^{210}Pb occurs at approximately 9.5 cm
08 depth. The maximum activity of the unsupported ^{210}Pb is recorded at 2.5 cm. The radionuclide ^{137}Cs
09 exhibits a well-resolved peak at 2.5 cm depth and ^{241}Am shows a distinct peak at 3.5 cm depth. The
10 lack of two distinct ^{137}Cs peaks might be related both to the slow sedimentation rate and to the
11 sediment subsampling resolution adopted (1 cm). The CRS model indicates that the top 8.5 cm
12 cover the past 115 years with 8.5 cm depth representing the year 1883. Artificial radionuclides are
13 in good agreement with the CRS model, which places the 1963 layer just under 3 cm depth.

14 Results from four radiocarbon dates indicate that the core dates to between 3,689-3,830 cal. yr
15 BP ($3,472 \pm 20$ ^{14}C yr) at 139 cm depth (Figure 3). Samples are in chronological order and good
16 linearity ($R^2 = 0.993$) resulted.

17 The ^{210}Pb and AMS dating techniques were compared using linear regression. An age-depth
18 model is shown in Figure 3. Good linearity ($R^2 = 0.985$) resulted suggesting that the combined ^{210}Pb
19 and ^{14}C dating methods provide reliable results. The estimated chronology, expressed as cal. yr BP,
20 calculated from linear regression is used for the interpretation of change in the core physical,
21 chemical and biological proxies.

22 SARs calculated from the CRS model and AMS radiocarbon dates are in agreement. Low
23 SARs (< 0.019 $\text{g cm}^{-2} \text{ yr}^{-1}$) were derived from the ^{210}Pb chronology and from the AMS radiocarbon
24 dates (< 0.048 cm yr^{-1}) and suggest that 1 cm of sediment is deposited in approximately 28 years (on
25 average).

26

27 **4.3 Lithology and stable isotopes**

28 Results of lithology (LOI₅₅₀, LOI₉₅₀), stable isotopes ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$), carbon, nitrogen and the C/N
29 ratio are shown in Figure 4.

30 Cores of sediment extracted from Furnace were brownish black in colour (Hue 5YR 2/2;
31 Oyama & Takehara, 1967) and exhibited homogeneous soft peaty-silt composition with no visible
32 changes. Strong hydrogen sulphide smells were evident during sampling, indicating the presence of
33 anoxia in the monimolimnion. LOI₅₅₀ ranges between 17 and 30% and an increasing trend is evident
34 from ca. 3,300 cal. yr BP upcore.

35 $\delta^{13}\text{C}$ is steady from the core bottom to ca. 3,200 cal. yr BP around ca. -27.5‰ and a slight
36 $\delta^{13}\text{C}$ increase to -26.7‰ is evident ca. 500 cal. yr BP. The top of the core shows a small $\delta^{13}\text{C}$
37 reduction. $\delta^{13}\text{C}$ -inferred salinity ranges between 0.2 and 2.0‰ indicating a small increase through
38 time. However, because the equation was developed on a very different system (*i.e.* the Baltic Sea;
39 Emeis *et al.*, 2003), results are treated cautiously.

40 A reduction in $\delta^{15}\text{N}$ values is evident from the core bottom to ca. 2,500 cal. yr BP (to 2.5‰)
41 with two peaks at ca. 3,900 cal. yr BP (3.6‰) and ca. 3,300 cal. yr BP (2.9‰). An increase is
42 evident to the core top where the highest value (4.0‰) is recorded.

43 Elemental carbon ranges from 10 to 20%, nitrogen ranges from 0.9-1.3%, while C/N ratios
44 range between 9 and 17 through the core. A C/N increase from the core bottom (13 C/N) to ca. 17 at
45 ca. 1,900 cal. yr BP is evident. The C/N trend is not uniform and it is characterised by five distinct
46 lows with ca. 800 yr intervals.

47 **4.4 Metals**

48 Metal profiles are shown in Figure 5. Iron concentrations vary from a maximum of 25.3 to a
49 minimum of 7.2 mg Fe g⁻¹. A decline in concentrations at the core bottom (ca. 4,000 cal. yr BP) is
50 evident with a reduction from 25.3 mg Fe g⁻¹ to 10.6 mg Fe g⁻¹ at ca. 3,100 cal. yr BP. From thereon
51

52 Fe concentrations are relatively stable to the top of the core. Manganese concentrations are
53 generally much lower than those of iron, ranging from 6.5 to 0.2 mg Mn g⁻¹. A similar reduction in
54 concentrations upcore are evident also for Mn. The Fe/Mn ratio exhibits an overall increasing trend
55 in the core ca. 4,100 cal. yr BP with ratios increasing from ca. 4 to ca. 40. From ca. 1,000 cal. yr BP
56 the Fe/Mn ratio increases to the core top reaching maximum values (ca. 70 Fe/Mn). Copper
57 concentrations range from 0.3-0.6 mg Cu g⁻¹. The maximum Cu/Mn value (3.4) is recorded at the
58 core top (ca. 300 cal. yr BP). Magnesium ranges between 1.2 and 2.6 mg Mg g⁻¹ in the core and a
59 slight increasing trend from ca. 3,600 cal. yr BP to the core top is evident.

60

61 **4.5 Fossil diatoms**

62 A summary diagram of diatom species occurrence (>2%; n=23) is presented in Figure 6. The
63 broken stick model suggested two statistically significant zones: Zone 1 - ca. 4,300-2,900 cal. yr BP
64 and Zone 2 - ca. 2,900-500 cal. yr BP. Fossil assemblages in zone 1 are mainly dominated by
65 *Achnantheidium minutissimum* (15%), *Cyclotella shumannii* (12%), *Cyclotella radiosa* (10%) and
66 *Brachysira vitrea* (7%). A decreasing trend characterises some species such as *Denticula tenuis* (6
67 to 3%), *Aulacoseira alpigena* (8 to 3%), *Cyclotella comensis* (8 to 2%), *Diatoma tenuis* (7 to 1%)
68 and *Tabellaria flocculosa* (6 to 2%), whilst *Fragilaria exigua* (3 to 8%) increases. The assemblages
69 in zone 1 are also represented by the presence of *Cyclotella cyclopuncta*, *Eunotia incisa*, *Cocconeis*
70 *placentula*, *Fragilaria capucina*, *F. capucina* var. *gracilis*, *Staurosirella pinnata* and
71 *Pseudostaurosira brevistriata* which have abundances lower than 2%. The dominant species in
72 zone 2 is *Achnantheidium minutissimum* (max 19%) together with *Cyclotella comensis* (max 14%),
73 *Aulacoseira alpigena* (15%) and *Fragilaria exigua* (13%). Differences with respect to zone 1 are
74 evident for *Cyclotella shumannii*, *C. radiosa* and *Brachysira vitrea*, which are lower in abundance
75 in zone 2, whilst *Denticula tenuis* disappears. An increase in *Tabellaria flocculosa* (up to 7%) is
76 evident at the top of zone 2. Species such as *Cyclotella cyclopuncta*, *Eunotia incisa*, *Cocconeis*

77 *placentula*, *Fragilaria capucina*, *F. capucina* var. *gracilis*, *Staurosirella pinnata* and
78 *Pseudostaurosira brevistriata* maintain abundances similar to zone 1. The appearance of species
79 with more brackish affinities, such as *Cyclotella choctawhatcheeana*, *Cocconeis scutellum*,
80 *Diatoma moniliformis*, *Ctenophora pulchella*, *Nitzschia valdestriata* and *Rhoicosphenia abbreviata*
81 (all <5% abundance) characterise zone 2.

82 Silica dissolution was generally evident in all taxa classified including strongly silicified
83 species (e.g. *Navicula radiosa*, *Pinnularia neomajor*). Weakly silicified species such as *Asterionella*
84 *formosa* are only present at the core top. Dissolution was present from ca. 4,300 cal. yr BP to ca.
85 600 cal. yr BP, where the F index varies between 0.4 and 0.8 (Figure 6). At the core top the
86 preservation index is closer to unity (1.0) indicating a well-preserved assemblage.

87 Diatom ecology is summarized as benthic or planktic and revealed a system dominated by
88 benthic species (Figure 6). Planktic forms ranged between 15 and 50%. No change of species life
89 form dominance is evident throughout the core. According to salinity tolerances, taxa were grouped
90 into (I) freshwater species, (II) freshwater species with brackish water affinity, (III) brackish and
91 (IV) marine species (Figure 6). Freshwater species were abundant through the entire core (ca. 40%).
92 A greater abundance of freshwater taxa with brackish water affinity was evident (ca. 40-50%).
93 Species with more brackish water affinity were generally less abundant (<20%), however, increases
94 were evident for both fresh-brackish and brackish species from ca. 2,500 cal. yr BP upward. Marine
95 species were present in very low abundances (<3%). To reconstruct past conductivity the combined
96 EDDI conductivity training set was applied, although fossil taxa found in Furnace only represented
97 between 26 and 56% of the species included in training set. WA diatom inferred conductivity (DI-
98 conductivity) ranges between 2.9-3.4 log $\mu\text{S cm}^{-1}$ (i.e. 794-2511 $\mu\text{S cm}^{-1}$) and there is a slight
99 increase in DI-conductivity through the core (Figure 6). Similar values (3.0-3.7 log $\mu\text{S cm}^{-1}$; 1000-
00 5011 $\mu\text{S cm}^{-1}$) resulted from the LWWA. The Modern Analogue Technique (MAT) provided
01 MinDC values between 105 and 149, suggesting no close analogues in the training set (EDDI,

02 2001).

03

04 ***4.6 Cladocera and sponges***

05 Simple quantitative measures of cladocera fragments and sponge spicule concentrations were used
06 in this study (Figure 6). A clear overall decreasing upcore trend in cladocera remains is evident with
07 drop from 2.1×10^3 fragments g^{-1} at ca. 4,300 cal. yr BP to 0.3×10^3 fragments g^{-1} WW at ca. 3,900
08 cal. yr BP. From here to the core top fragment concentrations remain below 1×10^3 fragments g^{-1}
09 WW except for a peak between 2,400 and 2,300 cal. yr BP. Higher concentrations of sponge
10 spicules are evident. Values reduce from 9.166×10^3 spicules g^{-1} WW at ca. 4,300 cal. yr BP to 0.4
11 $\times 10^3$ spicules g^{-1} WW at approximately 1,800 cal. yr BP. From here to the core top concentrations
12 are below 0.4×10^3 spicules g^{-1} WW with spicules absent in many samples.

13

14 **5. Discussion**

15 ***5.1 Modern hydrology: stability and mixing events***

16 Lagoons are subject to natural hydrological dynamism due to their location and their relatively
17 shallow depth (Gouze *et al.*, 2008). Climatic patterns together with basin morphology and
18 freshwater or saline inflows determine large fluctuations and rapid changes in the physical and
19 chemical characteristics. The characteristics can reduce dissolved oxygen (Levin *et al.*, 2009; Pena
20 *et al.*, 2010), which is progressively depleted by organic matter degradation (Kountoura &
21 Zacharias, 2011).

22 The hydrology of Furnace reflects the effect of both tidal and freshwater inflow. It is evident
23 that freshwater inflow determines the overall lake water levels while High Spring Water (HSW)
24 leads to periodically higher water levels. The period between July and September 2009 was
25 characterised by 562 mm of rainfall and led to a progressive water level increase in the upstream
26 Lough Feeagh and a subsequent rise in water levels in Furnace that persisted until mid-October

27 2009. The increased water level minimised the tidal effect with the exception of HSW. Higher
28 water levels associated with catchment drainage appear to preclude salt-water inflow at High Neap
29 Water (HNW). In contrast, following dry spells when water levels are lower in Furnace the tidal
30 effect is more pronounced even at HNW (Brett, 2010).

31 When meromixis occurs, denser brackish water forms the monimolimnion, which is overlain
32 by a mixolimnion of less saline water deriving from the watershed and precipitation (Hakala, 2004).
33 In Furnace, the freshwater inflow is a primary driver in forming a strong halocline that varies its
34 depth through seasons as a result of the balance of catchment runoff and seawater inflow. Furnace is
35 a saline lake with a natural but modified tidal inlet (Healy *et al.*, 1997). A weir holds back the lake
36 water and helps maintain permanent and stable meromictic conditions (Healy *et al.*, 1997; NPWS,
37 2001). The deep water is well protected from recirculation and is stagnant (Parker, 1977). The
38 detailed hydrological data confirmed that Furnace is permanently stratified due to the presence of a
39 strong halocline, which prevents the water column mixing. This condition has lead to extended
40 anoxia in the monimolimnion, which was additionally confirmed by a typical hydrogen sulphide
41 smell. The Wedderburn number (W) (Shintani *et al.*, 2010) calculated for Furnace confirmed that
42 water column overturn by wind forcing is improbable. On the other hand, a partial overturn
43 appeared in spring and summer 2010. Reduced precipitation determined isohaline conditions
44 through the water column as recorded in March, at the beginning of April and July. In parallel, DO
45 concentration increased in the monimolimnion. It is interesting to note how the thermocline reduced
46 in this period leading to isothermal conditions through the water column, which has the potential to
47 mix the water column. Additionally, the oxygen profile suggests that the temporary oxygenation of
48 the monimolimnion might be interpreted as the result of reduced rainfall that allowed more tidal
49 oxygenated water to flow into Furnace. To date, Furnace water column recording is still ongoing to
50 obtain a long-term monitoring period to detect the existence of seasonality of partial overturns and
51 better interpret the influence of the climate on hydrology.

52 Predictions of future climate change permit hypotheses of potential lake responses. An
53 increase in the frequency and intensity of extreme precipitation and a significant increase of 0.01
54 mm/year in mean precipitation were found to have occurred at Furnace between 1960-2009 (Fealy
55 *et al.*, 2010). A tendency towards wetter winters and drier summers has been observed over the last
56 few decades (Murphy & Washington, 2001; Parsons & Lear, 2001), and projected climatic changes
57 suggest a further increase in precipitation during the winter months and reductions during summers
58 in Ireland (Dunne *et al.*, 2008; Fealy *et al.*, 2010). Drier summers have the potential to weaken the
59 halocline in Furnace, and, as a result, stratification may only be maintained by the thermocline. One
60 might think that future predicted climatic conditions could result in more frequent overturns in
61 Furnace, however, temperature projections derived from three global circulation models and two
62 emissions scenarios (A2 and B2) suggest that the greatest warming will be experienced in the
63 autumn and spring seasons by 2080 (IPCC, 2000; Fealy *et al.*, 2010). An increased tendency for
64 overturns is therefore unlikely in Furnace, since hot summers followed by cool autumns are needed
65 to reach isothermal conditions through the water column and determine the water column mixing.
66 Permanent meromictic conditions will potentially characterise Furnace in the future, with rare water
67 column overturns caused by a combination of temperature, precipitation and wind.

68

69 ***5.2 Reconstruction of meromixis during the late Holocene***

70 Due to their location, coastal lakes are in contact with the sea, receiving saline water inflows and
71 meromixis can be considered as part of lake evolution (Hakala, 2004). The development of
72 ectogenic meromixis in Furnace is mainly a result of the tidal influence, which has progressively
73 increased in the past and it is probably related to sea-level rise.

74

75 ***5.2.1 The onset of meromixis***

76 Measurement of redox-sensitive metals (Fe, Mn and Cu) in lake sediment can enable reconstruction

77 of redox potential at the water bottom and the onset of meromixis in coastal areas (e.g. Sparrenbom
78 *et al.*, 2006; Nichol *et al.*, 2007). Variation in redox conditions determines metal migration from the
79 sediment to the water column and this process can also lead to the exhaustion of the pools in the
80 sediment (e.g. Kristiansen *et al.*, 2002; Kristensen *et al.*, 2003). A shift from oxygenated to oxygen-
81 depleted bottom waters is suggested by reductions in sediment manganese and iron at ca. 3,400 cal.
82 yr BP in Furnace. Mn is known to be very soluble compared to most inorganic ions (Davison,
83 1993). Under anoxic conditions it reduces from Mn^{4+} to Mn^{2+} and leaves the sediment accumulating
84 in oxygen-depleted bottom waters. Reduced manganese tends to disappear first (Kristensen *et al.*,
85 2003), which is also the case in Furnace, where it reaches background concentration at ca. 3,400
86 cal. yr BP, whilst Fe concentrations decrease more steadily upcore. The Fe/Mn ratio is a useful tool
87 to reconstruct variations in redox conditions (Boyle, 2001). In particular, reducing conditions are
88 suggested by an inverse correlation between Fe and the Fe/Mn ratio ($r = -0.44$, $p = 0.02$). This is the
89 case of metal profiles in Furnace, which are reinforced by the upcore increase of copper. Copper is
90 generally negatively correlated with Fe and Mn (Basaham, 1998) and its accumulation in sediments
91 has been described as an indicator of anoxia (Sohlenius *et al.*, 2001; Kawakami *et al.*, 2008). Under
92 anoxic conditions Cu^{2+} shows a strong affinity for complexing with sulphur and it precipitates as
93 copper sulphide (Cu_xS_y), which is stable (Balistrieri *et al.*, 1994; Jung *et al.*, 1996). Sulphate
94 reduction in organic-rich coastal marine sediments usually leads to the precipitation of pyrite (FeS_2)
95 (Skei *et al.*, 2003), and copper sulphides may co-precipitate even in the presence of iron sulphides
96 (Oakley *et al.*, 1980). However, the relative stability of Fe and the increase of Cu from ca. 2,100
97 cal. yr BP suggest that iron sulphide precipitation did not occur in Furnace. Indeed, Basaham (1998)
98 demonstrated that FeS_2 precipitation did not take place in the anoxic Al-Arbaeen lagoon (Red Sea)
99 sediments, where Cu was preferentially associated with organic matter. Therefore, it can be
00 assumed that because of the ability of Cu to form stable organic complexes (Shank *et al.*, 2004) and
01 the presence of high concentrations of complexing organic ligands in Furnace humic waters, copper

02 is preferentially associated with organic matter in Furnace sediments (e.g. Hullebusch *et al.*, 2003;
03 Sparrenbom *et al.*, 2006). In summary, the upcore increase in the Cu/Mn ratio, which is an indicator
04 of reducing conditions and anoxia formation (e.g. Jung *et al.*, 2006), supports the parallel increase
05 in the Fe/Mn ratio in Furnace as suggested by Pearson correlation ($r= 0.94$, $p<0.01$).

06 Carbonate precipitation is a common process in coastal systems (López & Lluch, 2000) and
07 the main forms are calcium and magnesium carbonates. In particular, magnesium calcite
08 $[\text{CaMg}(\text{CO}_3)_2]$ deposits are characteristic of brackish and saline sediments (Stumm & Morgan,
09 1981). A slight increase of the LOI₉₅₀ measurements (13 to 19%) is evident, and it might suggest an
10 accumulation of carbonates from ca. 3,500 cal. yr BP. Additional support is given by the parallel
11 increase in Mg that might be associated with the carbonate phase. Seawater inflow potentially
12 isolates bottom water, thus impeding water column mixing, and in turn determines increased calcite
13 precipitation and the onset of anoxia. It is accepted that organic matter preservation is enhanced by
14 permanent bottom water anoxic conditions (Meyers & Teranes, 2001). Therefore, the increase of
15 LOI₅₅₀ approximately 3,500 cal. yr BP, which parallels LOI₉₅₀, might be a result of anoxia
16 formation leading to enhanced organic matter accumulation in the sediment.

17

18 *5.2.2 Biological response*

19 Diatoms in Furnace inhabit the water column surface and littoral zones and their productivity is
20 probably constrained by peat-coloured waters coming from the catchment. The floristic composition
21 reflects the strong freshwater influence. The ecological subdivision of taxa into salinity groups
22 indicates that the fossil record is mainly represented by benthic and planktic freshwater species
23 together with fresh-brackish taxa able to tolerate slightly brackish conditions. The diatom
24 assemblages show a slight progression to fresh-brackish and brackish water affiliated taxa from
25 approximately 1,800 cal. yr BP onwards. Similar shifts with increasing salinity in coastal areas have
26 been documented in a number of studies (e.g. Westman & Hedenström, 2002; Emeis *et al.*, 2003;

27 Bao *et al.*, 2007). In particular, the Furnace diatom profile resembles diatom associations described
28 by Bao *et al.* (2007), who found species assemblages mostly dominated by the freshwater epiphyte
29 *Achnantheidium minutissimum* during the evolution of the Traba coastal wetland (Spain). The
30 increase in *Diatoma tenuis* from ca. 900 cal. yr BP might also indicate increased influx of marine
31 water to the lake as suggested by Mills *et al.* (2009).

32 Quantitative reconstructions of diatom-inferred (DI) conductivity were constrained by poor
33 frustule preservation and poor training set representation and thus the inferences are not wholly
34 reliable (Ryves *et al.*, 2006; Ryves *et al.*, 2009). Silica dissolution has altered fossil diatoms causing
35 partial loss of the record and it might be a factor responsible for the large number of unclassified
36 taxa. Valve dissolution depends on a number of factors such as pH, salinity, temperature, depth,
37 water permanence, meromixis and sediment accumulation rates (SARs) (e.g. Lewin, 1961; Marshall
38 & Warakomski, 1980; Hurd *et al.*, 1981; Barker *et al.*, 1994; Reed, 1998; Bidle *et al.*, 2002; Ryves
39 *et al.*, 2006; Roubex *et al.*, 2008). Diatom frustules are preserved in the sediment only if SARs are
40 high enough to prevent opal dissolution in opal-unsaturated bottom waters (Bohrmann, 1986; Emeis
41 *et al.*, 2003). Furnace had very low SARs (ca. 0.04 cm yr⁻¹ on average). A further limitation in
42 determining DI-conductivity was the lack close analogues between fossil taxa in Furnace and taxa
43 in the used calibration datasets. Fossil taxa constituted between 26-56% of the training set
44 assemblages. However, WA DI-conductivity increases from ca. 750 to 2,511 $\mu\text{S cm}^{-1}$ (ca. 0.5-1.7‰
45 salinity) and the oligohaline conditions are in harmony with the floristic changes and upcore
46 increases in brackish species. A salinity transfer function developed by Martin (2001) for Scottish
47 lagoons was also applied, however, there was a poor match (7%) between modern and fossil
48 assemblages. Reed (2007) proposed that transfer functions may only be accurate for sites included
49 in the training set while Saros (2009) commented strong seasonal salinity in lagoons may result in
50 poor ecological inferences when annual mean chemical conditions are employed in transfer
51 functions. Emeis *et al.* (2003) proposed that organic matter stable carbon isotopes could be used to

52 detect and quantify salinity changes in surface water when fossil diatom assemblages are not well
53 preserved. Salinity reconstruction based on $\delta^{13}\text{C}$ in Furnace suggests an increase from ca. 0.2‰ to
54 2‰. The $\delta^{13}\text{C}$ inferred salinity is of a similar range to the diatom-inferred increase, however the
55 inferences diverge at the core top.

56 An increase in mixolimnion salinity is also suggested by the parallel reduction of cladocera
57 remains. The study of Hofman & Win (2000) showed that prior to the Littorina transgression, the
58 Baltic Sea had a rich cladoceran and chironomid fauna typical of freshwater habitats, which reduced
59 and disappeared after that event. The cladocera remains in Furnace sediments could represent the
60 remains of individuals transported by freshwater inflows from the upstream lake and/or could be
61 autochthonous species able to withstand a certain degree of salinity (e.g. Sarma *et al.*, 2000; Green
62 *et al.*, 2005). However, no cladocera were found in modern water samples and the decline in
63 cladocera remains in Furnace is coincident with a progressive increase in salinity reconstructions.
64 Sponges, which are sensitive to extended anoxic conditions (Boundy-Sanders *et al.*, 1999; Bond &
65 Wignall, 2005), provide additional support to the hypothesis of meromixis formation. A decrease in
66 sponge remains parallels dissolved oxygen reduction in the monimolimnion inferred from redox-
67 sensitive metals. The decline in spicules from ca. 2,000 cal. yr BP might be driven by prolonged
68 anoxia and hydrogen sulphide formation (Engstrom & Wright, 1984; Thauer *et al.*, 2007). This
69 potentially made the monimolimnion a harsh environment precipitating benthic community collapse
70 and supporting only bacterial activity (Grall & Chauvaud, 2002; Hakala, 2004; Conley *et al.*, 2007).
71 However, further inferences would require systematic taxonomic and ecological sponge spicule
72 classification.

73

74 5.2.3 Sources of organic matter

75 Allochthonous organic matter includes a variety of ligno-cellulosic debris and soil-derived organic
76 material (Meyers, 1997) including particulate organic carbon (POC) and dissolved organic carbon

77 (DOC). In particular, fluxes of DOC from peatlands assume particular relevance since high DOC is
78 leached to surface runoff and groundwater (Barry & Foy, 2008). In Furnace, the majority of land-
79 derived material passes through the upstream lake Feeagh and the terrigenous material becomes
80 diluted with autochthonous material (Parker, 1977). The comparison of sediment $\delta^{13}\text{C}$ values with
81 C/N ratios showed ranges typical of organic inputs to coastal environments (Lamb *et al.*, 2006)
82 indicating a strong land-derived influence. $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values from Furnace are typical of
83 sediments from peat-dominated catchments (e.g. Skrzypek *et al.*, 2008; Diefendorf *et al.* 2008) and
84 a number of studies have revealed similar signals in lagoons and estuaries (e.g. Müller & Mathesius,
85 1999; Müller & Voß, 1999). The nature of the sediment organic matter is further confirmed by
86 similar results from the upstream catchment lake (Dalton *et al.*, 2010).

87 Organic matter degradation is minimal under anoxic conditions (Meyers, 1997) and sediment
88 $\delta^{13}\text{C}$ is considered to be more conservative in comparison to $\delta^{15}\text{N}$ and the C/N ratio (Thornton &
89 Mc Manus, 1994). However, microbial reworking during early diagenesis has the potential to
90 modify the original $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ value (Lehmann *et al.*, 2002; Lamb *et al.*, 2006). Yamaguchi *et*
91 *al.* (2010) found similar $\delta^{13}\text{C}$ increases in a saline meromictic lake and attributed this to preferential
92 microbial decomposition of low- $\delta^{13}\text{C}$ labile components such as amino acids. Furthermore, a shift
93 to positive $\delta^{13}\text{C}$ values has been linked to progressive salinity increases (Emeis *et al.*, 2003).
94 Additionally, in coastal waters the pH-dependent equilibrium between CO_2 ($\delta^{13}\text{C}$ -8‰) and HCO_3^-
95 ($\delta^{13}\text{C}$ 0‰) tends more to bicarbonate composition and in-lake derived organic matter would then
96 reflect lower $\delta^{13}\text{C}$ values (Emeis *et al.*, 2003).

97 As a result of permanent water stratification anoxic conditions may form and denitrification
98 leads to a release of ammonium with a consequent $\delta^{15}\text{N}$ rise ($> +10\%$) (Ogawa *et al.*, 2001). In
99 similar conditions, ammonification resulting from the bacterial reduction of organically-bound
00 nitrogen can also lead to a reduction in $\delta^{15}\text{N}$ values (Teranes & Bernasconi, 2000). This could be
01 the case at the bottom of the Furnace core, where fractionation in the order of ca. 1‰ determines a

02 $\delta^{15}\text{N}$ upcore reduction from 3,800 to ca. 2,300 cal. yr BP. The nitrogen stable isotope trend parallels
03 the reduction in redox-sensitive metals and it might suggest ammonification at the sediment-water
04 interface. Interpretation of the increase of $\delta^{15}\text{N}$ from ca. 1,000 cal. yr BP, when it is assumed that
05 the lagoon hydrodynamics were similar to modern conditions (*i.e.* permanent meromixis), proved
06 inconclusive. Diagenetic processes might be responsible (Yamaguchi *et al.*, 2010), however
07 correlations between $\delta^{15}\text{N}$ with the C/N ratio ($r= 0.0459$, $p= 0.810$) and between $\delta^{15}\text{N}$ and nitrogen
08 ($r= -0.163$, $p= 0.390$) proposed by Thornton & Mc Manus (1994) suggested no significant
09 relationships and thus no organic matter alteration by diagenesis.

10

11 ***5.3 Sea level rise during the late Holocene***

12 Mid-late Holocene sea level is estimated to be much lower than today. The highest quality sea-level
13 data (*i.e.* primary index points) available from Ireland are mainly located along the southern coast,
14 whilst fewer reliable sea-level index points are found along eastern and western coastlines (Brooks
15 & Edwards, 2006). Confirmation of a sea level rise is suggested by a palaeolimnological study
16 based on fossil diatoms in Lough Hyne (south-western Ireland) that documented a gradual marine
17 transgression from approximately ca. 4,000 cal. yr BP (Buzer, 1981). Furthermore, a sedimentary
18 record from the south coast of Ireland suggest a marine transgression of the inner coastal area from
19 ca. 7,400 cal. yr BP with a sea level of 5.7 m lower than the Ordnance Datum (OD) (Devoy *et al.*,
20 2004). The sediment sequence suggested a sea level rise of 1.19 m, passing from -1.82 m (OD) at
21 4,210 cal. yr BP to -0.63 m (OD) at 3,118 cal. yr BP. Shaw (1985) reconstructed a RSL of -0.36 m
22 (OD) 2,103 cal. yr BP that increased to -0.10 m (OD) 746 cal. yr BP. These RSL dates are
23 synchronous with the response in sediment proxies in Furnace. Sediment reconstructions suggest
24 the formation of anoxia at ca. 3,400-3,200 cal. yr BP due to the increasing inflow of marine water.
25 The progressive expansion of the more dense and stagnant monimolimnion and catchment and
26 upstream inputs may account for the later increase in brackish diatoms and disappearance of sponge

27 spicules. At approximately 4,000 cal. yr BP, the mean sea level is estimated to be ca. 1.8 m lower
28 than today (Devoy *et al.*, 2004), and only highest spring tides coupled with strong onshore winds
29 were potentially able to flow into Furnace. The lake hydrology was mostly governed by freshwater
30 discharge from the catchment and salinity was low. Following the progressive increase in the mean
31 sea level, tidal inflows became more frequent and seawater started accumulating in the deepest part
32 of the lake determining the formation of anoxia. The monimolimnion gradually reached modern
33 conditions, where even HNW can flow into Furnace.

34

35 ***5.4 Late Holocene climate variability***

36 The C/N ratio has been employed to reconstruct hydrological changes in coastal environments, for
37 example, change in lake levels (Wolfe *et al.*, 2011), isolation of coastal basins from the sea (Mills *et*
38 *al.*, 2009) and fluctuations in relative sea level and river discharge (Lamb *et al.*, 2007). Periodic
39 reductions of the C/N ratio may indicate dry periods characterised by reduced precipitation and
40 lower runoff, consequently resulting in lower C/N ratio values mainly deriving from autochthonous
41 phytoplankton. Ombrotrophic peatlands in northern Europe have long been recognized as excellent
42 climate archives (e.g. Barber *et al.*, 2003; Turney *et al.*, 2006; Swindles *et al.*, 2010). In particular,
43 palaeoclimatic studies on peat bogs in Ireland and UK have provided detailed reconstructions of the
44 alteration of wet-dry periods. Turney *et al.* (2006) analysed Irish bogs and lake tree populations and
45 a series of dry phases were recognized (ca. 4,100, ca. 3,200, ca. 2,650, ca. 1,700 and ca. 800 cal. yr
46 BP) indicating a significant reduction in moisture delivery and/or significantly reduced water tables.
47 More recently, widespread water deficits in summer months (2,752-3,102 cal. yr BP, 1,802-2,272
48 cal. yr BP, 1,482-1,702 cal. yr BP and 1,850-2,000 AD) have been revealed by Swindles *et al.*
49 (2010) from Northern Ireland. Similarly, alternate floods and droughts were identified by Barber *et*
50 *al.* (2003), who defined a series of phase-shifts to wetter climates from three sites in Ireland and the
51 UK. In particular, the record from the Abbeyknockmoy Bog (Co. Galway) provided a series of

52 major dry periods that corresponded to ca. 4,100, ca. 3,350, ca. 2,900, ca. 2,350, ca. 750 cal. yr BP.
53 The coincidence between Furnace C/N minima (at ca. 4100, 3300, 2400, 1700, 600) and the
54 inferred dry phases from peat bogs is marked. Offsets in dates might be in part due to intrinsic
55 chronological errors and/or differences in regional climates.

56 It is well known that the NAO influences European climate (Hurrell, 1995; Wanner *et al.*,
57 2001; Scaife *et al.*, 2008) and a number of studies suggested the existence of interannual to multi-
58 centennial NAO cycles (e.g. Mann *et al.*, 1998; Rimbu *et al.*, 2003, Magny, 2004). Magny (2004)
59 investigated synchronous changes in lake-levels from French pre-Alps and the Swiss plateau. Lake
60 level changes were assumed to be climatically driven and a rather unstable Holocene climate
61 indicated 15 high water level phases attributed to wetter conditions. Wet phases detected in
62 Southern Europe surprisingly correspond to dry phases inferred from Furnace C/N ratio. This
63 suggests that the NAO might represent an important driver in Furnace palaeohydrology, and it is
64 supported by similar findings in the upstream lake Feeagh (Jennings *et al.*, 2000).

65 Shifts to minimum C/N ratio values occur with ca. 800-year periodicity in Furnace. A number
66 of studies have detected a periodicity in solar activity of ca. 800 years (e.g. Schulz *et al.*, 2004;
67 Turney *et al.*, 2005; Ma, 2007) and these appear to be synchronous with the multi-centennial
68 variations in the NAO recorded in many studies (e.g. Magny, 2004; Hu *et al.*, 2003; Lamy *et al.*,
69 2006). Karlén & Kuylenstierna (1996) studied a possible correlation between changes in the climate
70 of Scandinavia and changes in solar irradiation. The authors demonstrated that for most of the last
71 9,000 years a good correspondence between the timing of cold events and the timing of major $\delta^{14}\text{C}$
72 anomalies due to low solar irradiation exists. Similar findings have been proposed by Bernárdez *et*
73 *al.* (2008), who reconstructed the climatic variability during the late Holocene in the NW Iberian
74 Peninsula and described how a high irradiance controlled precipitation and induced high run-off and
75 increased riverine influx. Therefore, it seems that changes in the solar activity have played a major
76 role in Holocene climate oscillations over the North Atlantic area (Magny, 2004).

77

78 **6. Conclusions**

79 The multiproxy palaeolimnological reconstruction of Lough Furnace suggests a response to
80 progressive sea level rise and meromixis formation during the late Holocene. Prior to ca. 4,300 cal.
81 yr BP lake salinity was probably close to freshwater conditions. The increased influence of marine
82 water into the basin relative to freshwater input has determined the onset of meromixis, which has
83 become progressively more stable. This determined the formation of anoxia in the monimolimnion
84 from ca. 3,400 cal. yr BP. Anoxia strongly limited the development of benthic communities and it
85 appears that from ca. 2,000 cal. yr BP onwards permanent meromictic conditions persisted. Prior to
86 3400 cal. yr BP the lake was probably holomictic and mixing events were likely more frequent than
87 today. Since meromixis formation water column stratification has become progressively more
88 permanent and only occasional overturns (coupled with dry summers) have occurred. Modern
89 hydrological monitoring data show that meromixis is mainly dependent on freshwater inflow and
90 temporary isohaline conditions are evident in spring and summer 2010. Climate projections indicate
91 that Furnace overturns might potentially remain rare in the future. Furthermore, this study outlines
92 how the hydrology of the Burrishoole catchment appears to be dependent on the North Atlantic
93 Oscillation and long-term solar cycles, with an alternation of dry and wet local climatic phases
94 recorded during the late Holocene.

95

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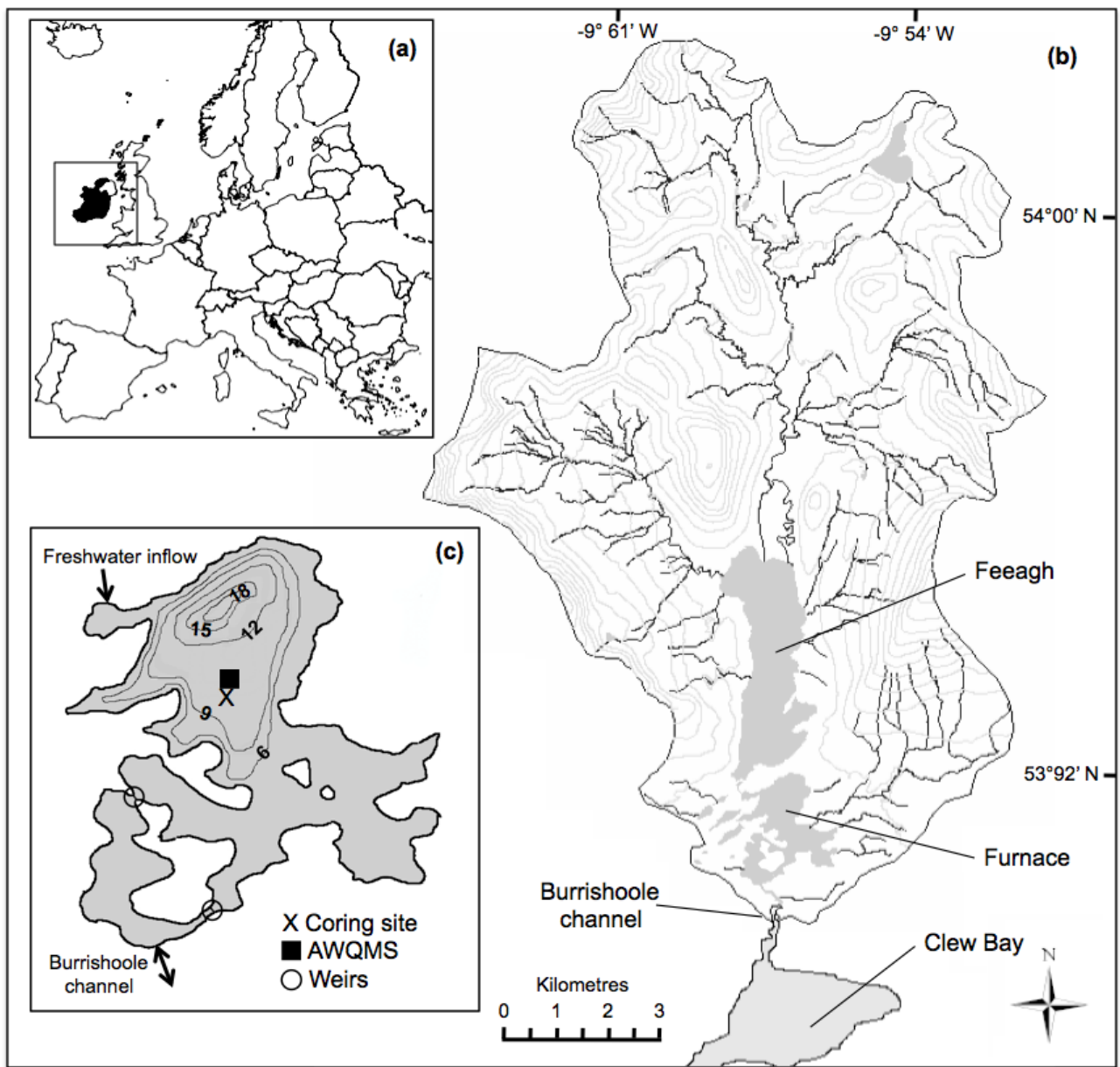
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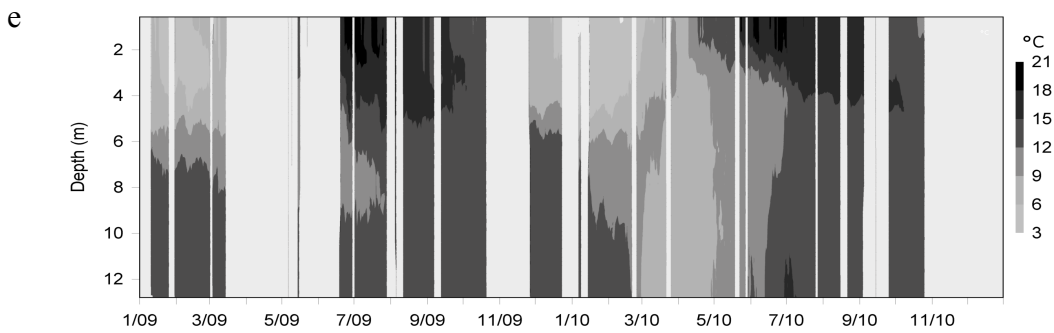
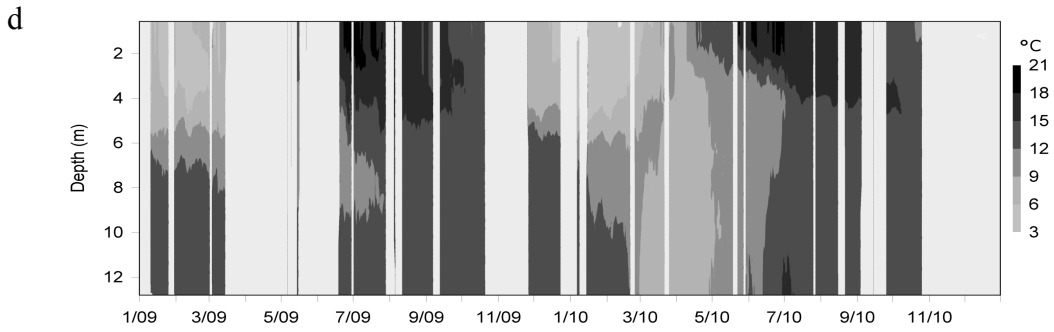
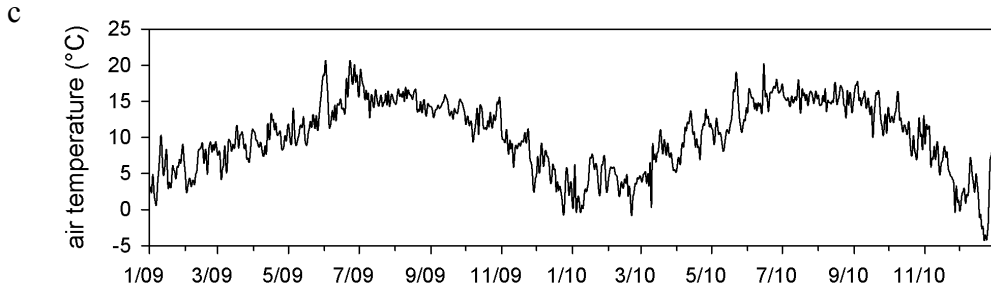
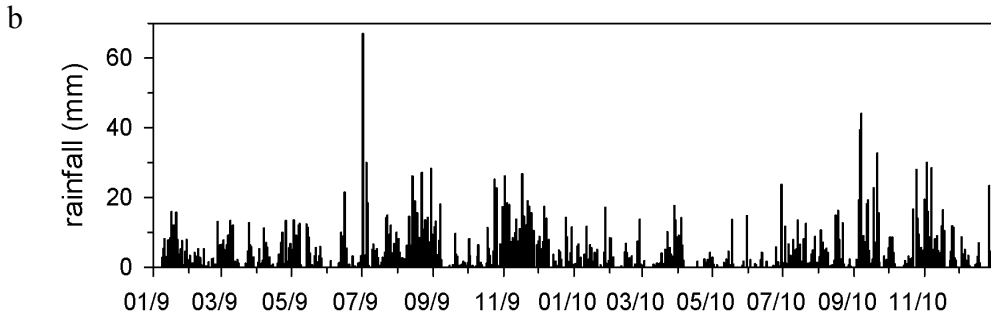
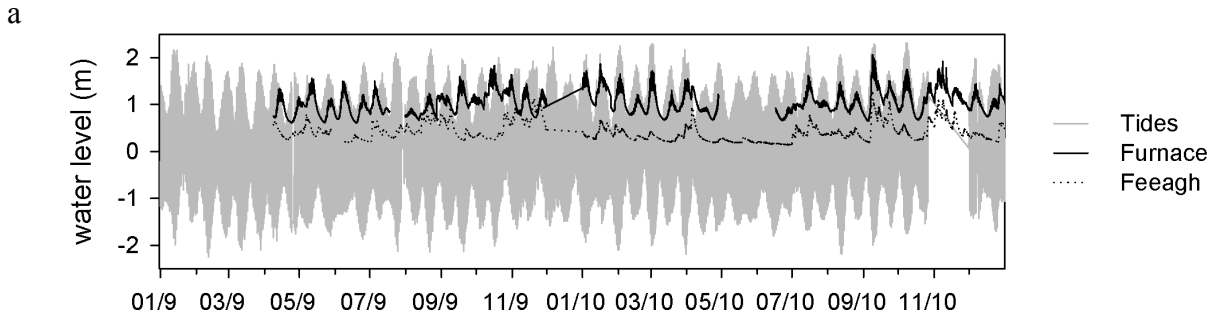
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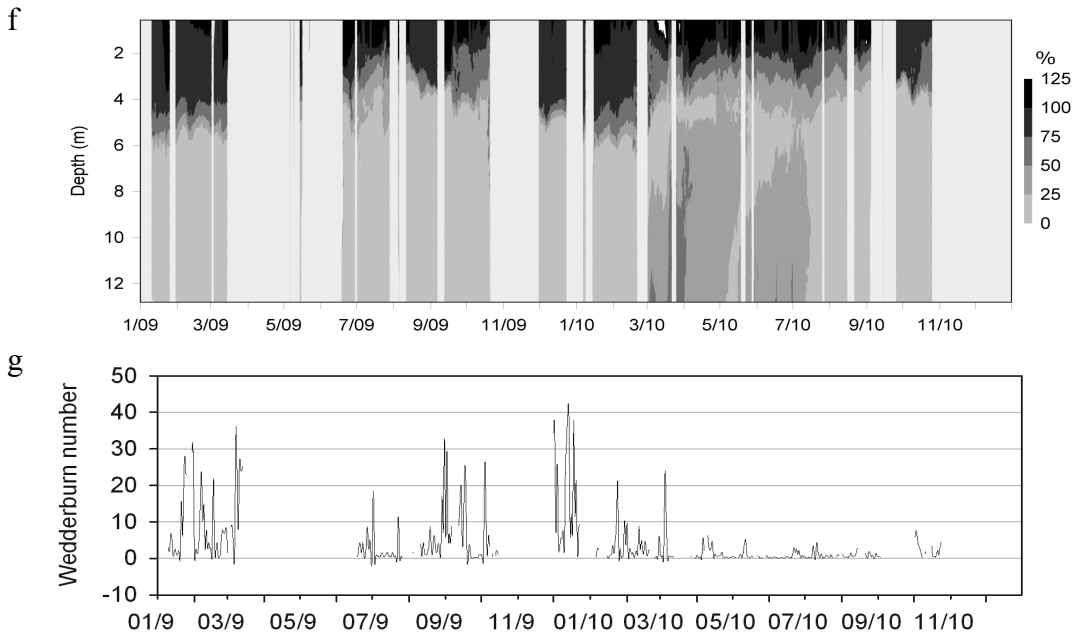
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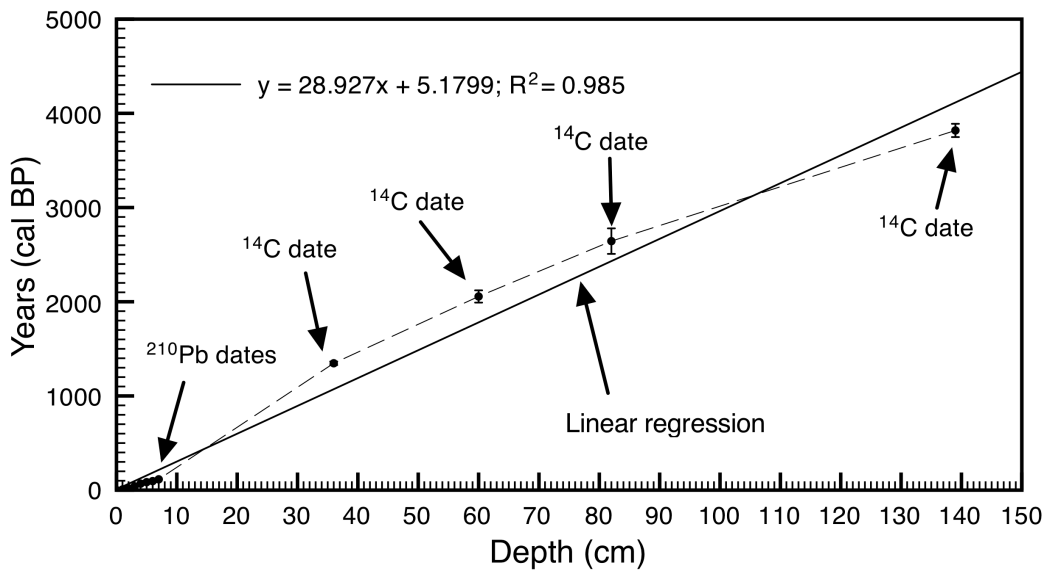
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Figure 1 – (a) Location of Ireland, (b) the Burrishoole catchment, (c) bathymetry (m), the coring site, the automatic water quality monitoring system (AWQMS) and weirs position in Lough Furnace.

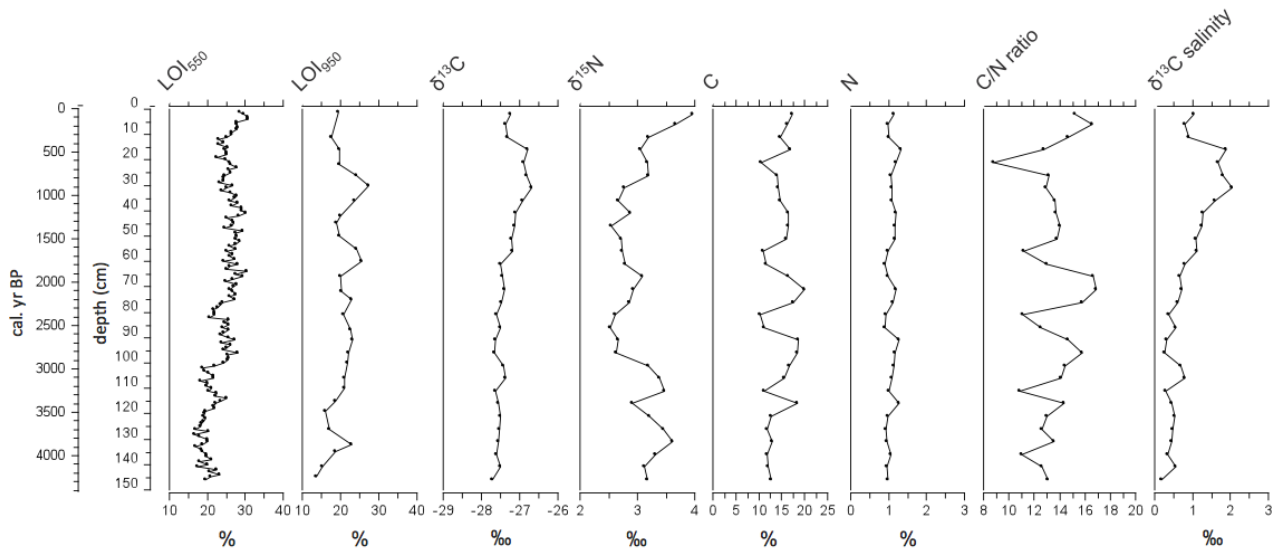




89 **Figure 2** – (a) Furnace (dark line) and Feagh (dotted line) water level and tides (grey line) (m); (b)
 90 rainfall (mm); (c) air temperature (°C); (d) Furnace salinity (‰); (e) temperature (°C), (f) dissolved
 91 oxygen (%) and (g) the Wedderburn number.



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 96 **Figure 3** – Age-depth model of combined ^{210}Pb and AMS radiocarbon dates.

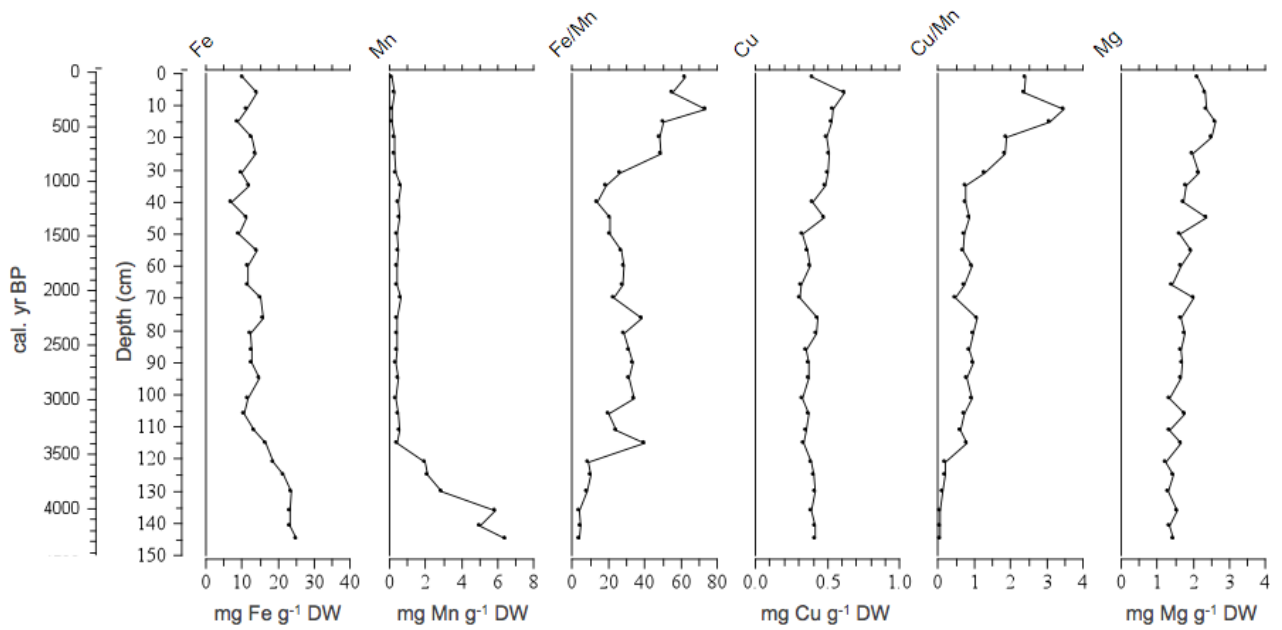


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00 **Figure 4** – LOI₅₅₀ (%), LOI₉₅₀ (%), carbon and nitrogen stable isotopes (‰), carbon and nitrogen
 01 (%), C/N ratio and δ¹³C-inferred salinity (‰) in Furnace.

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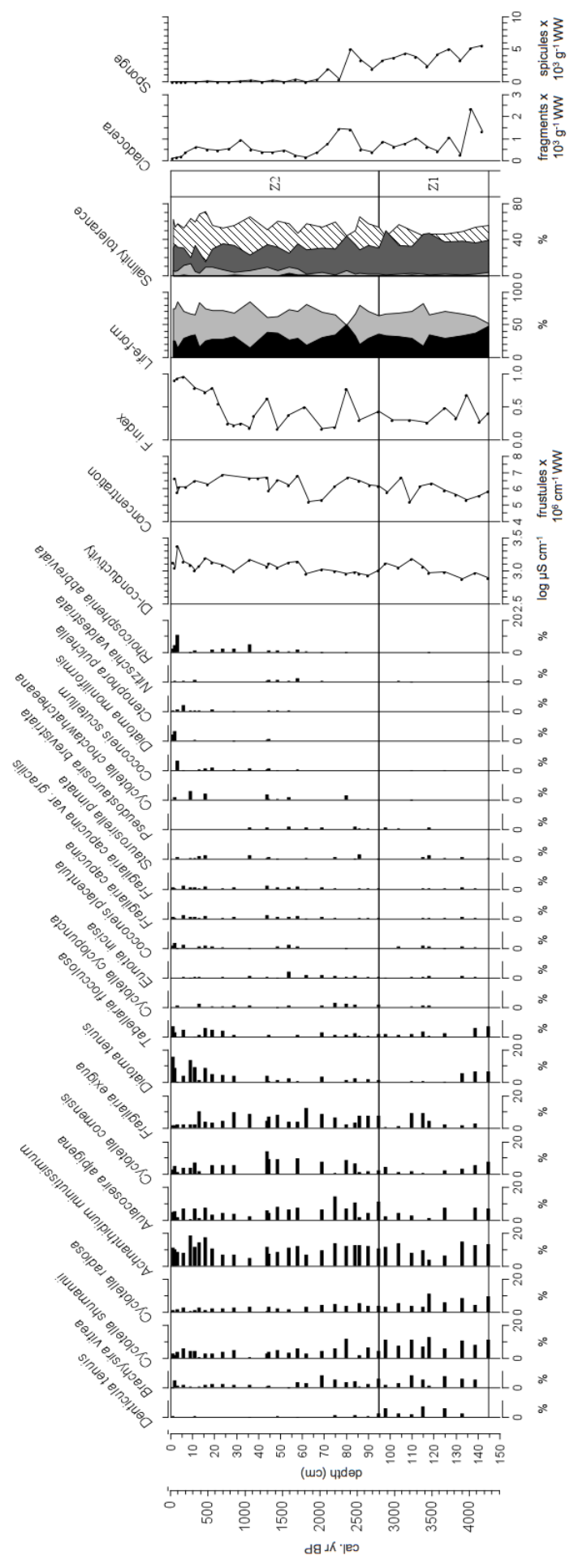
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05 **Figure 5** – Iron, manganese, Fe/Mn mass ratio, copper, Cu/Mn mass ratio and magnesium in
 06 Furnace.

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09 **Figure 6** – Summary of the most abundant (>2%) diatom taxa (n=23), DI-conductivity (WA; log
10 $\mu\text{S cm}^{-1}$), diatom concentration (frustules $\times 10^6 \text{ g}^{-1} \text{ WW}$), diatom dissolution index (F index),
11 diatom life-form (black=planktic; gray=benthic), salinity tolerance (black=marine; light
12 gray=brackish; dark grey=freshwater; black pattern=fresh-brackish), cladocera (total fragments \times
13 $10^3 \text{ g}^{-1} \text{ WW}$) and sponge spicule (spicules $\times 10^3 \text{ g}^{-1} \text{ WW}$) concentration in Furnace. Zones were
14 calculated by means of CONISS and the broken stick model (Bennet, 1996).