

Response and recovery of a *Sphagnum* peatland from long-term human-induced alkalinisation

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Abstract

Northern peatlands are significant terrestrial carbon stores but are increasingly threatened by human activities. Ombrotrophic peatlands, being naturally acidic, are particularly vulnerable to alkaline pollution. Despite their importance, the effects of alkalinisation on peatlands remain insufficiently studied. In Estonia, alkaline pollution from a cement industry and oil shale power plant emissions have degraded several peatlands since the 19th century. Although some sites have recovered in recent decades, more severely impacted areas remain in poor condition.

We investigated the effects of alkalinisation on Varudi peatland, a forested site in northeast Estonia, which was exposed to 125 years of alkaline emissions from a nearby cement factory. Using a multi-proxy, high-resolution palaeoecological approach combined with a precise and reliable age-depth model, we reconstructed changes in environmental, chemical, botanical, and hydrological conditions over the past millennium. Our findings revealed three successional phases: during the mid-13th century CE, land clearance and increased mineral deposition caused the site to transition from a bog to a poor fen phase between approximately 1250-1570 CE; and while the cement factory operated without efficient filters, the site became a pine-dominated fen between 1871-1995.

After the installation of filters in 1996, peatland pH returned to near pre-disturbance levels, and some recovery was observed. However, the site remains degraded. Our results indicate that alkalinisation significantly disrupts peatland functioning, reducing carbon storage and altering vegetation communities. These effects can persist for decades even after the source of contamination is removed, underscoring the need for more comprehensive monitoring of peatlands impacted by alkaline pollution globally.

1. Introduction

Despite only covering c. 3% of the Earth's surface, northern peatlands contain >500 gigatonnes of carbon (Bridgham *et al.*, 2008; Yu *et al.*, 2010; Yu, 2012). Their capacity to accumulate and store carbon results from the waterlogged and acidic nature of their soils (Clymo *et al.*, 1998). These conditions preserve organic material, which accumulates and may be stored indefinitely (Harenda *et al.*, 2018). Since their initiation, peatlands have slowly removed carbon from the atmosphere, imparting a weak but persistent cooling effect upon global climate over millennial timescales (Frolking *et al.*, 2006).

Despite being recognised as a valuable tool for climate change mitigation, peatlands still receive little protection, regionally or nationally (Rawlins and Morris, 2010). As of 2018, approximately 10% of the remaining peatlands worldwide are in a degraded state (Leifeld and Menichetti, 2018), while in Europe this rises to 25% (Tanneberger *et al.*, 2021). Such disturbance can disrupt the fragile hydrological balance that maintains the carbon sink function of peatlands and may cause them to shift from sinks to sources of atmospheric carbon, exacerbating climate change (Leifeld and Menichetti, 2018).

Estonia is one of Europe's most peat-rich countries, with peatlands covering c. 22.5% of its land area (Oru and Oru, 2008). Due to this abundance, peat is a significant natural resource for Estonia, and has been heavily exploited, particularly after the Industrial Revolution (Paal *et al.*, 2010; Łuców *et al.*, 2022). The rise in anthropogenic pollution since this time has caused substantial changes in global geochemistry such that this era is informally termed 'the Anthropocene' (Fiałkiewicz-Kozieł *et al.*, 2018; Waters *et al.*, 2023). During this time in Estonia, emissions from industrial sources were characterised by high levels of calcium-rich particulate matter, with most of the emissions concentrated in the northeastern industrial region of the country (Liblik *et al.*, 1995; Karofeld, 1996). Ombrotrophic peat bogs, which are the dominant type of peatlands in Estonia, being naturally acidic and nutrient-poor ecosystems are particularly sensitive to alkaline atmospheric pollution (Paal *et al.*, 2010). These emissions caused significant changes in the geochemical and botanical composition of bogs adjacent to pollution sources, which resulted in dramatic increases in pore-water pH and losses of bog-specific vegetation (including *Sphagnum* mosses) near affected sites (Paal *et al.*, 2010; Vellak *et al.*, 2014). This was often followed by encroachment of *Pinus sylvestris* (Scots

60 pine) onto polluted sites alongside other species typical of nutrient-rich alkaline environments
61 (Pensa *et al.*, 2004, 2007; Ots and Reisner, 2006; Kaasik *et al.*, 2008; Kask *et al.*, 2008).

62

63 By the 1990s, industrial emissions in Estonia began to fall, following a decline in power
64 generation and improved filtration systems in factories (Liiv and Kaasik, 2004). Following
65 these reductions, polluted peatland sites began to show signs of recovery, with acidic
66 conditions and bog-specific vegetation returning (Karofeld, 1996; Kaasik *et al.*, 2008; Paal *et al.*, 2010). However, in more heavily polluted sites, this recovery has been slow, and the
67 impact of past alkaline pollution persists to this day in some areas (Ots and Reisner, 2006). It
68 remains unclear whether current levels of atmospheric pollution are sufficiently low to permit
69 their full recovery in the future, or how long this process will take (Paal *et al.*, 2010). Despite
70 growing concerns over alkaline pollution and its potential future effect on peatlands,
71 particularly concerning their role as carbon reservoirs, research exploring the effects of
72 alkalisation upon peatland ecosystems and their subsequent recovery has been limited.

74

75 Atmospheric pollution remains a significant threat to peatland ecosystem functioning
76 (Bobbink *et al.*, 1998; Turetsky and St Louis, 2006; Osborne *et al.*, 2024). The effects of alkaline
77 pollution upon peatlands have been relatively overlooked relative to those of acid rain due to
78 its effects being more localised (Vellak *et al.*, 2014; Sutton *et al.*, 2020). However, nearly two
79 billion tonnes of alkaline residues are emitted into the atmosphere each year (Gomes *et al.*,
80 2016). Despite environmental standards curbing emissions in recent decades, in some areas
81 these regulations are not consistently enforced or are merely declarative (Abril *et al.*, 2014;
82 Ivanov *et al.*, 2018). Following global reductions in acid rain since the 1980s, the relative
83 proportion of alkaline pollutants in airborne particulate matter has increased in the UK, much
84 of Europe, North America and China since 1986 (Turetsky and St Louis, 2006; Sutton *et al.*,
85 2020). Additionally, climate change may exacerbate the effects of increased alkalinity in
86 boreal regions, as permafrost thawing may cause the expansion of areas of open water,
87 increasing surface runoff and infiltration in some regions (Walvoord and Kurylyk, 2016). This
88 may allow for longer contact times between surface water and carbonate bedrock,
89 accelerating weathering and raising the pH of surface waters which may then enter peatlands
90 in the surrounding catchment (Schindler, 1997; Osterkamp *et al.*, 2000; Lehmann *et al.*, 2023).

91

In this study, we focus on the effects of alkalinisation resulting from over 125 years of intense emissions from a nearby cement factory upon Varudi soo (bog), a formerly ombrotrophic peatland in northeastern Estonia. By employing a high-resolution, multi-proxy palaeoecological framework, we reconstruct changes in the chemical, botanical, hydrological and environmental conditions of the site over the past millennium to address the following questions:

1. What is the current state of a heavily polluted raised bog almost 30 years after the reduction in alkaline pollution?
2. How has alkaline pollution altered the ecosystem functioning of the site and how does this compare with pre-disturbance conditions?
3. To what extent has this ecosystem function recovered 30 years after removing the point source of pollution?
4. Can we identify critical transitions that can be broadly applied to assess peatland condition and recovery following alkaline pollution?

2. Methods and materials

2.1. Study area

Varudi bog (59°26'19"N, 26°35'13"E) is located in Lääne-Virumaa, northeastern Estonia, consisting of fen-bog habitats. The site is approximately 10 km south of the coast of the Gulf of Finland and Baltic Klint (Figure 1) and spans c. 12.6 km². The site is primarily a forested *Sphagnum* bog interspersed by numerous bog pools and hummocks, with an overstory of *Pinus sylvestris*. Varudi peatland receives 478 mm of rainfall per year, has a mean annual temperature of 7.3 °C, and prevailing winds are from the southwest and south. The underlying bedrock is composed of Cambrian and Lower Ordovician siliciclastic sedimentary rocks and Middle Ordovician limestones (Sibul *et al.*, 2017) covered with a relatively thin layer of glacial and post-glacial sediments.

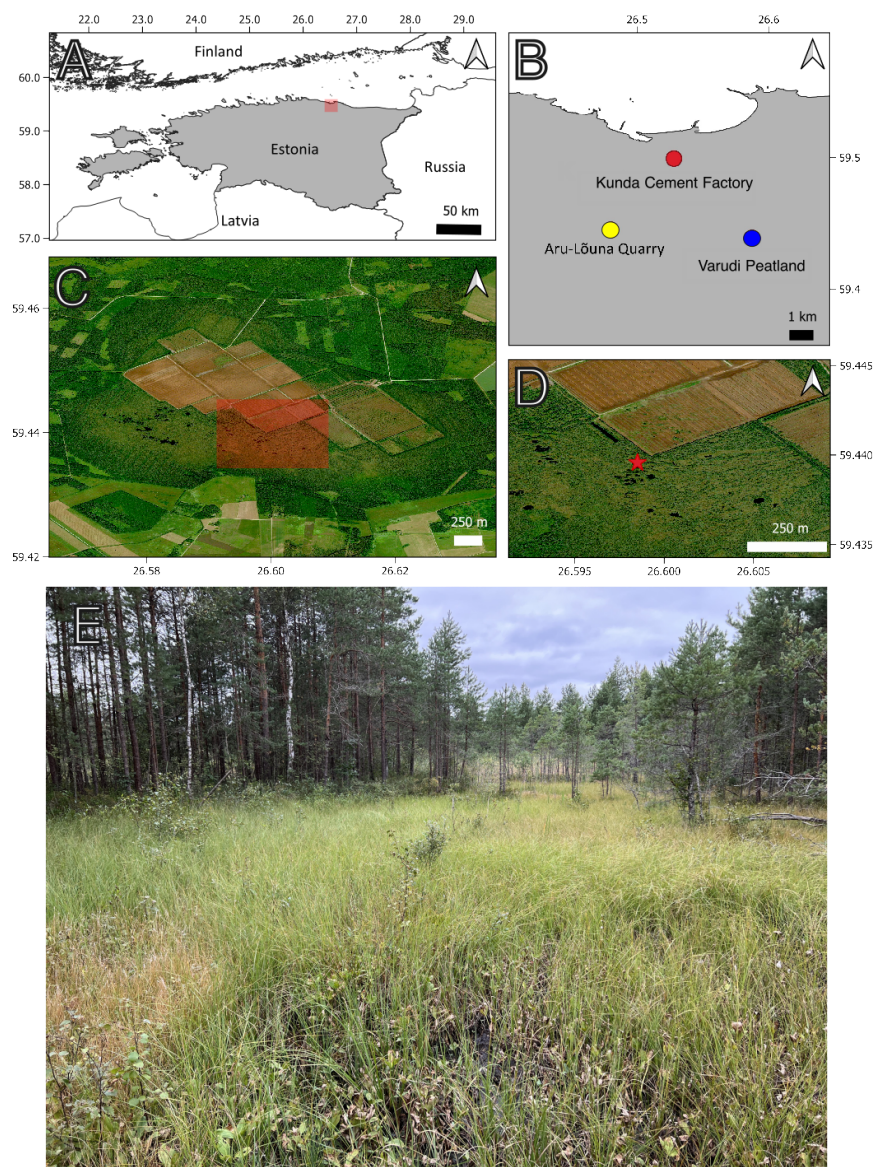


Figure 1: Map of study locations. A. Modern-day Estonia (in Gray). The red shaded square in map 1A indicates the area mapped in map 1B. B. Locations of sites relevant to this study (Kunda Cement Factory: Red, Aru-Lõuna Quarry: Yellow, Varudi peatland: Blue). C. Satellite image of Varudi bog (© Microsoft) showing the extent of peat cutting and drainage that has taken place over the past century. The red-shaded area indicates the area mapped in map 1D. D. Close-up of the location from where core VAR1 was recovered (red star). E. Coring location.

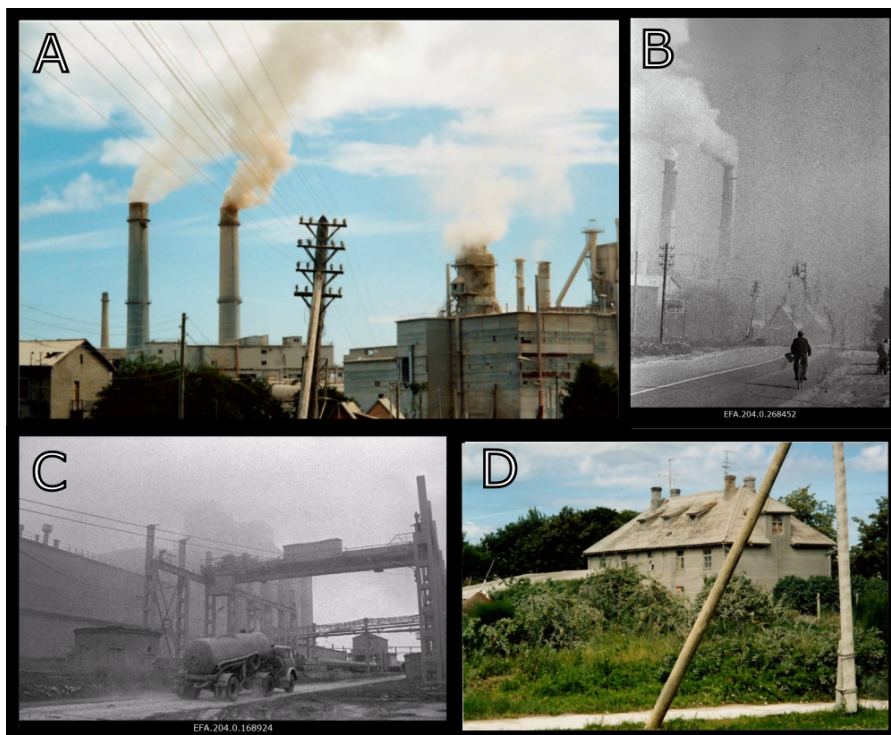
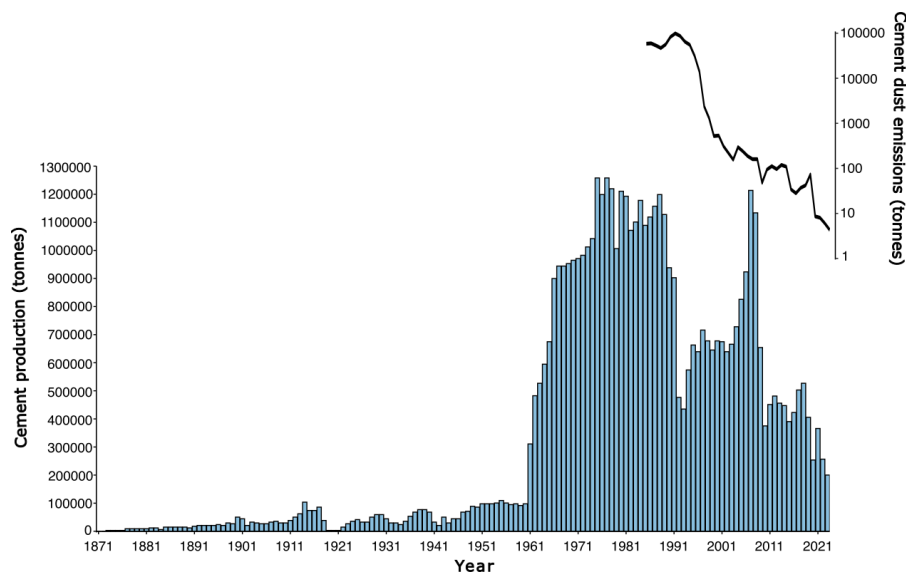


Figure 2. Photographs of the Kunda Cement factory and locale taken during the 1980s and 1990s, showing A. Cement dust emissions from the chimneys. B. Photograph of chimneys and factory surroundings. C. Photograph of the factory workings, showing substantial cement dust deposition in the surrounding area. D. Photograph showing cement dust deposition upon house near the factory. Photography by: A and D: Atko Heinsalu taken in the early 1990s; B: Estonian National Archives Photo Database (code EFA.204.0.268452) August 1994 Albert Truuväart. C: Estonian National Archives Photo Database (code EFA.204.0.168924) August 1989 Tiit Veermäe.

Varudi was selected for study due to its proximity (7.5 km NW) to the Kunda Nordic Tsement Factory (henceforth Kunda Cement Factory) (Figures 1 and 2), which has been operational since the 1870s. During the late 1970s, cement production peaked at c. 1.2 million tonnes per year (Figure 3). The dust emissions from the cement plant have fluctuated between 45,000 and 99,000 tonnes per year during the last decades of the 20th century, with the highest dust emissions recorded in 1991 (Ots and Mandre, 2012). In addition to emissions from the cement factory, the site has also received emissions from nearby alkaline generating industries and oil shale power plants, including the Balti power plant, located approximately 100 km west, and the Aru-Lõuna limestone quarry, situated 8 km to the east (Karofeld, 1996, Figure 1) which supplies raw materials for cement production at Kunda. However, significant emission reductions occurred after 1996, following the installation of pollution control filters, which

148 lowered emissions from 14,000 tonnes in 1996 to 530 tonnes in 2000 and 8 tonnes in 2020
149 (Figure 3).
150 The input of alkaline cement dust emissions has generated the drastic pH increase of bog's
151 water. During 1996-1997, the pH levels taken from peat pools at Varudi varied from 7.6 to
152 8.5, whereas in natural conditions bog pools have a pH in the range of 3-4. Disturbances at
153 the site are compounded by drainage and peat harvesting for horticulture, which continues
154 to the present day, with harvesting affecting around 40% of the former centre of the site
155 (Figure 1C). There is limited documentary information available related to the timing of
156 drainage and peat cutting at Varudi, although historic maps (ETOMESTO, 2025; MAA-JA
157 RUUMIAMET, 2025) indicate that most of the extensive drainage took place before 1977 and
158 after 1944, probably relating to the large-scale drainage projects undertaken in Estonia in the
159 1960s under Soviet rule (Paavilainen and Päivänen, 1995). Peat extraction matching the areal
160 extent of present-day works at the site by 1988. Prior to this, there is documentary evidence
161 for drainage in the northern, eastern and western margins of the site having taken place
162 around 1866 – 1911, likely indicative of the systematic forest drainage that took place in
163 Estonia around this time (Paavilainen and Päivänen, 1995).



164
165 Figure 3. Cement annual production and emissions by the Cement Factory in Kunda. Data adapted from Trumm
166 *et al.*, (2010), digitised from a figure at page 209 using WebPlotDigitiser: <https://automeris.io/WebPlotDigitizer/>
167 Emissions data (available from 1985 to 2023) sourced from Raukas (1993); Pärtma (2023) and Heidelberg
168 Materials Kunda AS (2023). Note the logarithmic scale for emission values.
169

170 The chemical composition of the cement dust emissions from Kunda consists primarily of CaO
171 (12 – 17%), SiO₂ (6 – 9%), and several other trace metals, including lead (Pb, c. 60 mg kg⁻¹),
172 cadmium (Cd, c. 0.9 mg kg⁻¹), and zinc (Zn, 129 mg kg⁻¹) (Mandre and Ots, 1999). The pH of
173 the cement dust in the water solution ranged between pH 12.3 to 12.6 (Mandre and
174 Korsjukov, 2007).

175 2.2. Coring method 176

177 In August 2022, an 86 cm peat core (VAR1) was extracted from Varudi peatland using a 1-
178 meter-long Wardenaar peat corer (Wardenaar, 1987, coring location: 59°26'23"N,
179 26°35'55"E, Figure 1D). The core site was located near an actively harvested peat area but is
180 within an intact section of the site in what was originally the bog's central raised dome.
181 Vegetation was characterised by *Pinus sylvestris*, with the ground cover dominated by
182 *Eriophorum* sp., *Menyanthes trifoliata*, *Vaccinium* sp. and scattered *Betula nana*. Both
183 *Sphagnum* and brown mosses were also present. Despite its proximity to a drainage ditch,
184 the coring location was representative of the overall condition of the site.

185
186 Following recovery, the core was wrapped in plastic and transported to the Faculty of
187 Geographical and Geological Sciences at Adam Mickiewicz University, Poznań, Poland for
188 analysis. The core was stored at 4 °C prior to sub-sampling.

189 2.2. Dating methods and age-depth model 190

191 Identifiable above-ground plant macrofossils were picked from 1 cm thick sub-samples taken
192 at various depths throughout the core, following frameworks by Piotrowska *et al.* (2011) and
193 Nilsson *et al.* (2001). *Sphagnum* stems, branches and leaves were preferentially used for
194 dating where present. Where these were not available, above-ground remains of ericaceous
195 plants (leaves, stems, seeds) were used instead. Initial samples were taken at 20 cm intervals
196 throughout the core to establish a baseline chronology, which was then used to determine
197 where additional samples would be selected. A total of 12 samples were sent for radiocarbon
198 analysis. Each sample was pre-treated using the acid-base-acid approach and analysed by
199 accelerator mass spectrometry (AMS) at the Poznań Radiocarbon Laboratory, Poland.

200
201 To provide a reliable chronology for recently accumulated peat, ²¹⁰Pb and ²³⁹⁺²⁴⁰Pu analyses
202 were used on material from the upper 40 cm of the core at 1 cm contiguous resolution,

203 following methods outlined by Appleby (1998). Peaks in the activity of $^{239+240}\text{Pu}$, which are
204 linked to nuclear fallout events (e.g., 1950s atmospheric nuclear tests), were also measured
205 as independent time-markers to validate and supplement the age-depth model (Mroz *et al.*,
206 2017; Cwanek *et al.*, 2021). Samples were processed at the Polish Academy of Sciences'
207 Institute of Nuclear Physics, Kraków, Poland using an AlphaAnalyst™ 7200 spectrometer
208 (Mirion Technologies). Quality control and accuracy was ensured by measuring blanks and
209 certified reference material (IAEA 447, IAEA 385) alongside the core samples (results are
210 provided in [Table S1](#). The activity concentration of ^{210}Pb (T 1/2 = 22.3 yr) was estimated by
211 measurement of its decay product - ^{210}Po (T 1/2 = 138.4 d), while $^{239+240}\text{Pu}$ was measured
212 directly. Activity concentrations of ^{210}Pb and $^{239+240}\text{Pu}$ are reported in units of Bq kg^{-1} .

213

214 The age-depth model was constructed by integrating all ^{14}C , ^{210}Pb , and $^{239+240}\text{Pu}$ data within a
215 Bayesian framework in R using the package 'rplum' (rplum package, Aquino-López *et al.*, 2018;
216 Blaauw *et al.*, 2021; R working group, 2023). rplum generates maximum age probabilities at
217 user-defined intervals (here every 1 cm), together with maximum and minimum ages based
218 upon calculated 95% credible intervals. This method allows for the integration of ^{14}C dates
219 with the ^{210}Pb dates without the need for re-modelling (Aquino-López *et al.*, 2018, 2020).
220 Radiocarbon dates were calibrated in rplum using the INTCAL20 curve, with post-1950
221 samples using the BOMB1 curve for the Northern Hemisphere (Reimer *et al.*, 2020; Uno *et al.*,
222 2013). In this study, the resultant ages are expressed as calendar years (cal) CE, with 0 BP
223 equal to 1950 cal CE.

224 2.3. Palaeoenvironmental proxies

225 2.3.1. Testate Amoebae

226

227 Samples were processed following a modified version of protocols by Hendon and Charman
228 (1997). Samples were placed into 50 ml centrifuge tubes filled with deionised water and
229 agitated for c. 10 minutes. These were sieved through a 300 μm mesh and the smaller fraction
230 was retained. Sieved samples were centrifuged at >3000 rpm for 5 minutes and a sub-sample
231 of the resultant material was transferred to a microscope slide for identification at 400x
232 magnification. Samples were not heated or micro-sieved, to retain small but ecologically
233 sensitive species, as recommended by Avel and Pensa (2013). A minimum count of 100 tests
234 for each sample was considered statistically significant (Payne and Mitchell, 2008). Tests were
235 identified to the species level where possible, with reference to Siemensma (2023) and Mazei

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237 and Tsyganov (2006) and were later pooled into taxonomic groups defined by Amesbury *et al.* (2016). The relative abundance (%) of each taxa count was calculated for each sample. 238
 239 Water table depth was reconstructed using the pan-European tolerance down weighted with
 240 inverse de-shrinking transfer function model by Amesbury *et al.* (2016), based on a training
 241 set of 1302 samples spanning 35° of latitude and 55° of longitude. Small species (<10-25 µm,
 242 broadly oval-shaped) not included in these groupings but present in the core from Varudi
 243 were grouped under the '*Cryptodiffugia oviformis*' group. We also reconstructed changes in
 244 pH using a transfer function developed by Šímová *et al.*, (2022), based on over 250 samples
 245 from east-Central Europe. The calibration dataset includes samples from peatland sites
 246 ranging from acidic bogs to calcareous fens, with a pH range of 3.5-7.9 making it suitable for
 247 reconstructing pH in sites that have likely undergone substantial changes in pH over time,
 248 such as Varudi. The transfer function was constructed using maximum likelihood calibration
 249 of Gaussian response curves (MLRC) with squared chord distance as the dissimilarity measure.
 250 The full dataset transfer function (mineral and ombrotrophic sites combined) was tested by
 251 leave-one-out cross-validation, achieving a RMSEP₁₀₀ of 0.45 pH units and an R² of 0.86. Both
 252 reconstructions were performed using the 'Rioja' package in R (Juggins, 2019). Comparisons
 253 between the modern calibration and fossil datasets using the 'compare.datasets' function in
 254 Rioja indicate good levels of overlap, with no non-analogue assemblages identified in the
 255 fossil data.

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Moved down [1]: Reconstructions were performed using the 'Rioja' package in R (Juggins, 2019).

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256
 257 Stratigraphically Constrained Cluster Analysis (CONISS) was used to quantitatively define
 258 stratigraphic zones in the sub-fossil testate amoeba data (Grimm, 1987). This method is used
 259 to determine statistically significant zones, reflecting changes in testate amoebae community
 260 composition. The data were square root transformed prior to applying CONISS. The numbers
 261 of statistically significant zones were determined using Broken-Stick modelling.

262
 263 We applied the framework outlined by Burge *et al.* (2023) to identify significant ecological
 264 transitions in the testate amoeba communities due to disturbance. Sub-fossil data were
 265 Hellinger transformed and analysed using the 'prcurve' function in the 'analogue' package in
 266 R (Simpson and Oksanen, 2016). Principal response curves (PrC), which reduce multi-
 267 dimensional community data to a single-dimensional curve, calculating the (dis)similarity
 268 between sample scores indicative of the difference between samples (De'ath, 1999; van Den
 269 Brink and Braak, 1999) provided the best fit for our data. A generalised additive model (GAM),

an approach effective in capturing rapid and non-linear changes in palaeoecological studies, was applied to the PrC data to account for temporal autocorrelation (Auber *et al.*, 2017; Beck *et al.*, 2018; Burge *et al.*, 2023). Given the abrupt changes in our data, we also applied an adaptive spline GAM following Burge *et al.* (2023). However, this method does not account for temporal autocorrelation and thus remains incompatible with the GAM framework as outlined by Simpson (2018).

2.3.2. Plant macrofossils

Plant macrofossil analysis followed procedures adapted from Mauquoy *et al.* (2010). Samples of approximately 5 cm³ were sieved through a 200 µm test sieve and the larger fraction was retained. Botanical composition was estimated as percentages under a low-powered microscope at between 10–100 times magnification, using a 10 x 10 grid eyepiece graticule to aid quantification of plant remains. Seeds, fruits, spindles, leaves, and wood were also identified at the species level where possible and counted as individual counts. Identification was aided by identification guides (Katz *et al.*, 1965, 1977; Grosse-Brauckmann, 1972, 1974; Tobolski, 2000; Mauquoy and van Geel, 2007; Bojnanský and Fargašová, 2007). *Sphagnum* remains were identified to sub-generic sections, with 100 leaves examined per sample, where possible, to calculate the relative abundance of each sub-section as a percentage of the total leaves identified.

2.3.3. Pollen, non-pollen palynomorphs and microscopic charcoal analyses

Past changes in vegetation cover at the landscape scale were assessed using pollen analysis (Seppä and Bennett, 2003). A total of 22 samples were prepared following the laboratory procedures outlined by Berglund and Ralska-Jasiewiczowa (1986). Each 1 cm thick sample, measuring 1 cm³, was sub-sampled at 5 cm intervals throughout the core. The samples were treated with 10% potassium hydroxide (KOH) to remove humic compounds before acetolysis. A *Lycopodium* tablet (batch no: 280521291, 18,407 spores per tablet; Manufacturer: Lund University) was added to each sample to calculate pollen concentrations, following methods by Stockmarr (1971). Samples were transferred to microscope slides and mounted in glycerine jelly for analysis. Pollen, spores, and selected non-pollen palynomorphs (NPPs) were identified and counted using a high-powered stereo microscope. Identification was based on established atlases and keys (Pollen: Moore *et al.*, 1991; Beug, 2009; NPPs: van Geel, 1978;

307 van Geel and Aptroot, 2006; Miola, 2012). Although a target of 500 terrestrial pollen grains
308 was aimed for per sample, this count was not always achievable due to low pollen
309 concentrations in some core sections. The relative abundance of spores and NPPs was
310 calculated as a proportion of the terrestrial pollen sum (TPS), which includes both arboreal
311 (AP) and non-arboreal (NAP) pollen, excluding aquatic and wetland plant spores, Ericaceous
312 pollen, and NPPs.

313 In addition to pollen counts, microscopic charcoal particles ($> 10 - < 100 \mu\text{m}$) were counted
314 from pollen slides as past fire activity, both natural and anthropogenic (Finsinger and Tinner,
315 2005), while spheroidal carbonaceous particles (SCPs) counted as indicators of industrial
316 activity (Patterson III *et al.*, 1987; Swindles, 2010). Microcharcoal concentrations per cm^3
317 were calculated by dividing the number of particles counted with the number of *Lycopodium*
318 spores and multiplying this by the total number of particles counted, and accumulation rates
319 (reported as particles $\text{cm}^{-3} \text{yr}^{-1}$) were calculated by dividing the pollen concentration with the
320 rPlum-derived age increments per cm slice.

321 2.3.4. Apparent rates of carbon accumulation

322

323 To measure apparent rates of carbon accumulation (aCAR), contiguous 1 cm-thick sub-
324 samples were taken throughout VAR1. The volume of each wet sample was determined by
325 water displacement, and then the samples were dried in an oven at 105°C until no further
326 weight loss occurred. Dry bulk density was calculated by dividing the wet volume by the dry
327 mass of each sample. Organic matter content (derived from LOI) was determined by ashing
328 the samples at 550°C for six hours, following the method of Chambers *et al.* (2011).

329 The carbon content of each sample was estimated indirectly by multiplying the LOI content
330 by 0.52, based on the average ratio of organic carbon (OC) and LOI in ombrotrophic peat from
331 multiple studies (Ball, 1964; Dean, 1974; Gorham, 1991; Clymo *et al.*, 1998). Carbon density
332 was calculated by multiplying the dry bulk density (g/cm^3) by the percentage of carbon
333 content, as described by Chambers *et al.* (2011). Apparent carbon accumulation rates (aCAR)
334 were then calculated by dividing the carbon density from each peat slice by the sedimentation
335 rate, determined from the age-depth model (Young *et al.*, 2019, 2021).

336 2.3.5. μXRF -Core Scanning (ITRAX)

337

338 To identify the section of the core affected by cement dust pollution, we followed methods
339 similar to those used by Varvas and Punning (1993) to assess pollution histories from Estonian
340 lake sediments. They identified rapid increases in micro-element concentrations associated
341 with alkaline fly-ash emissions, accompanied by a decrease in organic matter, indicating the
342 presence of particulate emissions from oil-shale combustion by power plants.

343

344 In this study, the concentrations of geochemical elements throughout the core were
345 measured using an ITRAX μ XRF core scanner equipped with a molybdenum X-ray tube.
346 Element concentrations were quantified as counts per second, based on the number of
347 secondary fluorescence detected for each element over a given period. Measurements were
348 taken at 5 mm intervals (30 kV, 50 mA, exposure time: 30 seconds per step) at the Institute
349 of Geography, University of Bremen, Germany. The scanner identified the activity of the
350 following elements: Al, Si, P, S, Ar, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, Se, Br, Rb,
351 Sr, Y, Ba, Pt, Pb, Bi, and Fe.

352

353 The mean concentrations of each element were calculated for each 1 cm slice of the core. To
354 account for variability in element concentrations due to sedimentation throughout the core,
355 the results were normalised by dividing the total counts by the sum of the coherent and
356 incoherent peaks, following the approach of Orme *et al.* (2015) as recommended by Longman
357 *et al.* (2019). The chemical signature of cement dust was identified based on the composition
358 of clinker emissions from the Kunda Cement factory, as detailed by Klõšeiko *et al.* (2011)
359 (Clinker emissions being the emissions produced by the production of clinker, the primary
360 ingredient of cement, created by the high-temperature heating of limestone and other
361 materials). Due to the uneven and unconsolidated nature of the top 6 cm of the core, this
362 section was not scanned and was removed prior to analysis.

363

364 Principal Component Analysis (PCA) was used to summarise patterns of variation in the
365 geochemical data, using the 'vegan' package in R (Oksanen *et al.*, 2019). To mitigate scaling
366 effects, the data were standardised to z-scores. The analysis was conducted with varimax
367 rotation in correlation mode, to explore correlations between elements and organic matter
368 percentage (Silva-Sanchez *et al.*, 2014). The number of components to retain for analysis was
369 determined using a Broken-Stick model.

3. Results.

3.1. Dating and age-depth model.

A total of 13 radiocarbon dates were analysed from core VAR1, four of which were post-bomb dates extending to a depth of 19.5 cm. The oldest dated section of the core produced a date of > 1000 yr. Radiocarbon and calibrated dates are shown in Table 1.

Table 1. Uncalibrated and calibrated radiocarbon dates from core VAR1, including depths and materials used for dating. PMC = Percent modern carbon

Code	Depth (cm)	Radiocarbon Age + error	Material dated	Calibrated dates + uncertainties
Poz-164520	5.5	104.51 ± 0.33 pMC	Seeds, ericaceous leaves	68.3% probability 1950 – 1957 (5.6%) 2010 – 2012 (62.7%) 95.4% probability 1956 – 1957 (10.6%) 2009 – 2012 (79.9%) 2012 – 2013 (5%)
Poz-164521	9.5	110.06 ± 0.67 pMC	Seeds, ericaceous leaves	68.3% probability 1957 – 1958 (7.5 %) 1997 – 1999 (50%) 1999 – 2000 (10.8%) 95.4% probability 1958 (11.2 %) 1996 – 2001 (85.2 %)
Poz-164662	15.5	110.36 ± 0.48 pMC	Seeds, ericaceous leaves	68.3% probability 1958 (7.3%) 1997 – 1999 (56.9%) 2000 (4.1%) 95.4% probability 1958 (11.9%) 1997 – 2000 (80.3%)
Poz-161987	19.5	118.8 ± 0.67 pMC	<i>Sphagnum</i> stems, leaves, branches	68.3% probability 1986 (68.3%) 95.4% probability 1959 (5.4%) 1960 (3.4%) 1986 - 1989 (86.7%)
Poz-162663	26.5	80±30	<i>Sphagnum</i> stems, leaves, branches	68.3% probability 1697 - 1724 (22.0%) 1814 - 1837 (20.1%) 1881 - 1913 (26.1%) 95.4% probability 1691 - 1729 (26.2%) 1809 - 1922 (69.3%)

Poz-164664	34.5	80±35	<i>Sphagnum</i> stems, leaves, branches	68.3% probability 1695 - 1725 (21.6%) 1813 - 1839 (19.6%) 1878 – 1916 (27.1%) 95.4% probability 1686 - 1725 (26.4%) 1805 - 1929 (69.0%)
Poz-161988	39.5	105±30	<i>Sphagnum</i> stems, leaves, branches	68.3% probability 1695 - 1725 (19.9%) 1813 - 1839 (17.5%) 1846 – 1853 (4%) 1869 - 1872 (1.4%) 1878 - 1916 (25.5%) 95.4% probability 1683 - 1737 (25.9%) 1803 - 1937 (69.5%)
Poz-163962	49.5	540±30	<i>Sphagnum</i> stems, leaves, branches	68.3% probability 1329 – 1338 (8.5%) 1397 - 1428 (59.8%) 95.4 % probability 1323 - 1357 (26%) 1392 - 1437 (69.5%)
Poz-161989	59.5	865±30	<i>Sphagnum</i> stems, leaves, branches	95.4% probability 1153 - 1263 (87.2%)
Poz-163963	69.5	875±30	<i>Sphagnum</i> stems, leaves, branches	95.4% probability 1126 - 1231 (79.2%) 1243 - 1258 (1.8%)
Poz-161990	79.5	910±30	<i>Sphagnum</i> stems, leaves, branches	68.3% probability 1047 - 1085 (28.6%) 1097 - 1103 (2.8%) 1126 - 1179 (30%) 1192 - 1205 (1.8%) 95.4% probability 1041 – 1214 (95.4%)
Poz-161992	85.5	985±30	<i>Sphagnum</i> stems, leaves, branches	68.3% probability 1022 - 1048 (26.8%) 1084 - 1128 (34.7%) 1140 - 1150 (6.8%) 95.4% probability 994 – 1055 (38.7%) 1076 – 1158 (56.7%)

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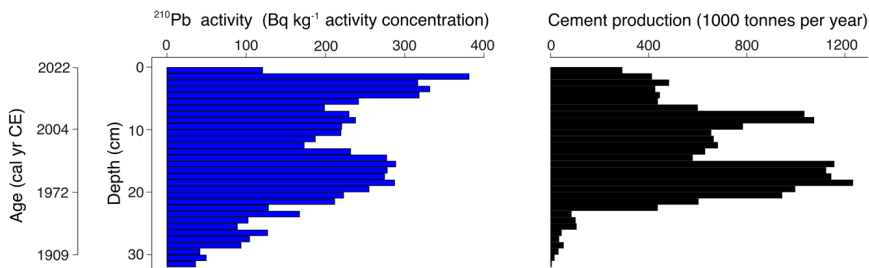


Figure 4. Comparison between ^{210}Pb activity concentrations throughout core VAR1 and cement production. Values interpolated from a GAM fitted to annual cement production data from Kunda Cement Factory (Figure 2; Trumm *et al.*, (2010)) at the same age frequency as the VAR1 age depth model (Figure 5) using a GAMM. Age ranges are based upon the median ages from the age-depth model.

The ^{210}Pb activity profile throughout core VAR1 (Figure 4) declined throughout the core, although not in a typical monotonic pattern, with a substantial increase from 12.5 cm to 19.5 cm, indicating that the rate of ^{210}Pb accumulation was not constant throughout the core. This spike is due to the enrichment of ^{210}Pb from fly-ash fallout from the cement factory and other sources (Vaasma *et al.*, 2014; 2017). Comparison of the ^{210}Pb activities and cement production rates applied to the age-depth model support this (Figure 4) with a significant correlation ($r_b = .487$, $p = <.0001$), indicating that ^{210}Pb accumulation in the core cannot be solely attributed to precipitation. From 32 cm core depth, ^{210}Pb activities achieve low levels, although they do not reach the background activity. Unsupported ^{210}Pb was calculated using linear regression of the last 5 samples, showing that the samples assumed background activities following the final measured samples.

The $^{239+240}\text{Pu}$ activity profile reflects the history of atmospheric deposition at the site (Figure 5). A clear peak at 28.5 cm corresponds to the onset of nuclear testing in 1945, followed by a second, larger peak at 22.5 cm, likely associated with the peak in global fallout from bomb testing in 1963, before the signing of the Partial Test Ban Treaty (Cwanek *et al.*, 2021). Smaller peaks at 16.5 cm suggest possibly the 1986 Chernobyl fallout (Ketterer *et al.*, 2004), and at 5.5 cm potentially originate from the 2011 Fukushima disaster (Bossew, 2013).

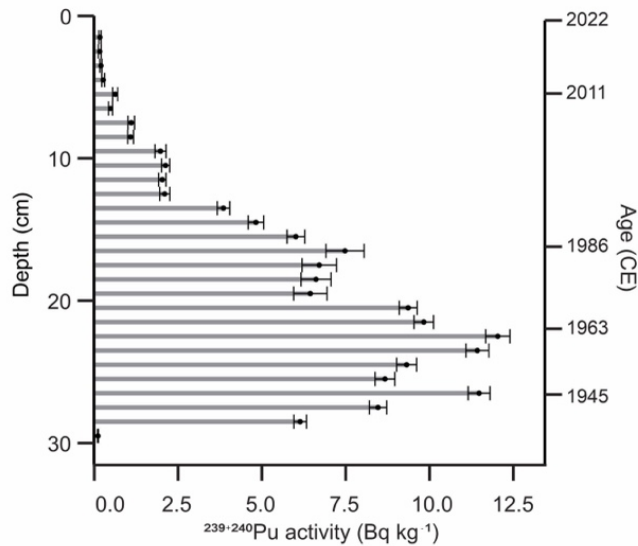
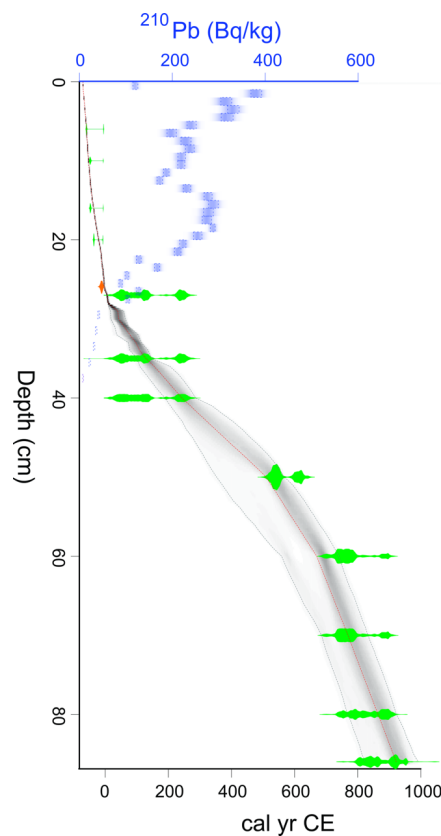


Figure 5. $^{239+240}\text{Pu}$ activity with depth in core VAR1. Selected median dates from the rPlum-derived age-depth model show good correspondence between known fallout events. Each peak may be tentatively related to a known nuclear fallout event, as shown on the right y-axis.

The age-depth relationship for core VAR1 (Figure 6) was calculated by aligning ^{210}Pb and $^{239+240}\text{Pu}$ data with the calibrated radiocarbon dates. [Model diagnostics are shown in Figure S1](#). Considering the enrichment of ^{210}Pb from industrial fallout, the assumption of a constant unsupported ^{210}Pb supply was violated. Despite this, anchoring the model with the known 1963 peak in $^{239+240}\text{Pu}$ activity and adding a constant but uncertain reservoir effect of c. 15 years for radiocarbon dates improved the model alignment with the peaks in $^{239+240}\text{Pu}$. A radiocarbon reservoir effect is possible in *Sphagnum* peatlands, due to the recycling of 'old' gaseous carbon by mycorrhizal fungi associated with ericaceous plants near the peat surface. However, this does not usually occur when individual plant remains are dated, as was the case for our study (Piotrowska *et al.*, 2011). A more likely explanation could be the uptake of old carbon by vegetation of dissolved carbonates, particularly in the more recent samples (Madeja and Lafowski, 2008) where old carbon may derive from the buried cement dust.

The resulting age-depth model (Figure 6) indicates that peat accumulation rates were stable from the base of the core until around 1255 cal CE, after which accumulation slowed to around 0.22 yr cm^{-1} , remaining low until around 1940 cal CE, where there was a sharp acceleration to c. 2 yr cm^{-1} .

430



431

432 Figure 6. Age-depth model for core VAR1, including calibrated radiocarbon dates and uncertainty (green) ^{210}Pb
433 activity (Bq kg^{-1}) in blue and the calendar date for the 1963 nuclear treaty peak (orange). Uncertainties for the
434 model are shown as the shaded area.

435 3.2. Peat physical and chemical properties

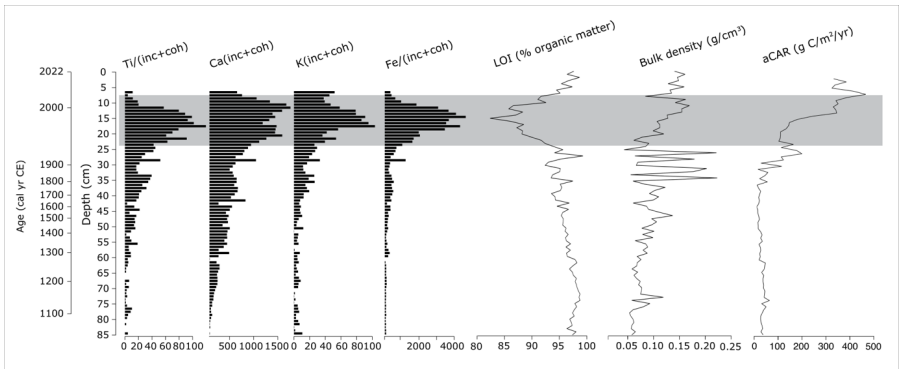
436

437 Figure 7 shows that activities of elements associated with clinker dust pollution began to
438 increase around c. 1873 cal CE, marking the beginning of cement production in Kunda. There
439 is evidence of increasing lithogenic dust deposition throughout the core, beginning as early
440 as the mid-12th century. There is a clear negative correlation between the counts of lithogenic
441 elements associated with cement dust (Ti, Pb, Ca, K, Fe, Cr) and organic matter content, most
442 noticeably between c. 1942 - 2006 cal CE. Organic matter content falls sharply until c. 1988
443 cal CE, suggesting the accumulation of cement dust occurred during this period, mirroring the
444 results of studies in NE Estonia lakes impacted by oil shale powerplants (Varvas and Punning,
445 1993; Punning *et al.*, 1996; Koff *et al.*, 2016).

446

447 The aCAR for the Varudi core is illustrated in Figure 7, which also shows the trends in LOI %,
448 bulk density, and carbon accumulation throughout the core. Over the past millennium, the
449 average aCAR for the entire core is $148.7 \text{ g OC m}^{-2} \text{ yr}^{-1}$. This is especially high in the upper
450 section of the core, where carbon accumulation rates peak at $342.2 \pm 231.7 \text{ g OC m}^{-2} \text{ yr}^{-1}$. For
451 most of the record (c. 1045 to 1910 cal CE), the mean aCAR is $72.9 \pm 28.9 \text{ g OC m}^{-2} \text{ yr}^{-1}$, aligning
452 more closely with average values reported for Northern peatlands (Roulet *et al.*, 2007). The
453 rate of carbon accumulation is relatively stable from the base of the core until c. 1280 cal CE,
454 with aCAR averaging $94.5 \pm 19.8 \text{ g OC m}^{-2} \text{ yr}^{-1}$. After this point, aCAR falls to $51.3 \pm 19.7 \text{ g OC}$
455 $\text{m}^{-2} \text{ yr}^{-1}$, remaining low until c. 1840 cal CE. After this date, aCAR increases significantly,
456 reaching an average of $256.6 \pm 221.2 \text{ g OC m}^{-2} \text{ yr}^{-1}$.

457



458

459 Figure 7. Comparison of selected elements detected by the μ XRF core scanner, as indicated by the PCA
460 analysis, for the detection of dust fallout from the Kunda Cement Factory. In addition to peat physical property
461 parameters: Loss on ignition (LOI%), bulk density and apparent carbon accumulation rates (aCAR). μ XRF Data
462 are presented as counts per second and normalised by dividing the sum of incoherent (inc) and coherent (coh)
463 activities. The shaded area represents the section of core where most of the cement dust is concentrated. The
464 uppermost aCAR sample was removed from the figure to aid interpretation.

465

466 The broken stick model shows that the two first components together explain a significant
467 proportion (48.3%) of the total variance in the peat's chemical composition (PCA1: 25.5%,
468 PCA2: 22.8%). These components highlighted elements associated with clinker dust
469 deposition from the Kunda Cement Factory and are significantly negatively correlated with
470 peat LOI content (Figure 8).

471

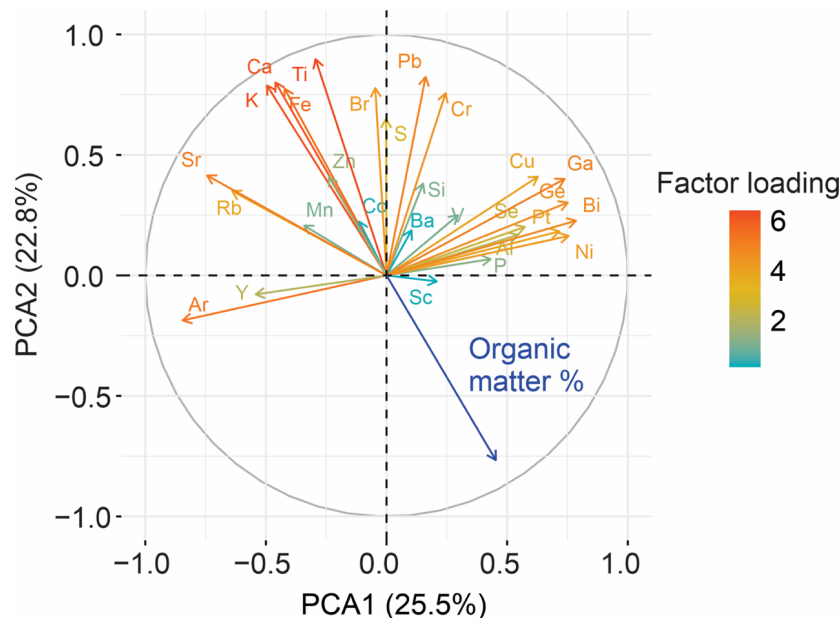


Figure 8. Biplot of first and second principal components showing factor loadings of individual elements scanned across core VAR1. Positively correlated variables point to the same side of the plot, while negatively correlated elements point to the opposite sides. Peat organic matter % (Derived from Loss on Ignition) content is shown in blue. The colour of the lines represents the sum factor loading of each variable for both axes, representing how strongly each variable contributes to the principal component.

3.3. Palaeoecological reconstructions

3.3.1. Plant Macrofossils

The results of plant macrofossil analysis are illustrated in Figure 9. The plant macrofossil data show three major phases of vegetation change, corresponding broadly with shifts observed in the testate amoeba and palynological records (Figures 10, 11 and 12).

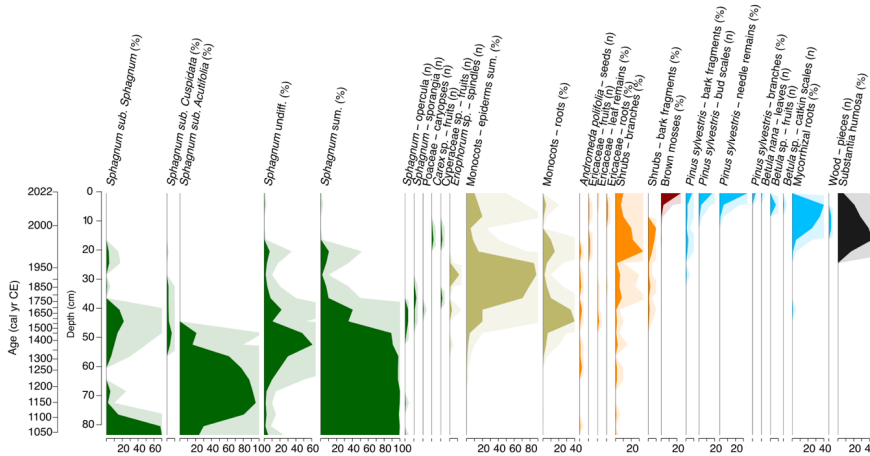


Figure 9. Plant macrofossil stratigraphic diagram illustrating the changes in the botanical composition throughout core VAR1. Note the mixed data types used, percentages and total counts.

From c. 1000 cal CE, the site is dominated by *Sphagnum* mosses, particularly those of the subgenus *Sphagnum*, although by the beginning of the 12th century, *Sphagnum sub. Acutifolia* becomes the most abundant. By c. 1450 cal CE, there was an increasing abundance of monocots, likely from *Eriophorum* species and Cyperaceae, owing to the presence of spindles and fruits identified to these species. During this period, *Sphagnum* gradually declined, eventually disappearing entirely by the start of the next phase.

The latter portion of the record, starting c. 1970 cal CE, is characterised by a shift towards more ‘woody’ vegetation. Shrub-type taxa increase initially, followed by a rise in ligneous remains, particularly those of *Pinus sylvestris*. Mycorrhizal roots, bark fragments, and pine needles become significant components of the peat's botanical composition in the upper section. *Betula* is represented in the uppermost samples by its characteristic catkin scales and fruits, along with a *Betula nana* leaf recovered from the surface sample. This late phase is also characterised by a high percentage of unidentified organic material. At the top of the core, remains of the brown moss *Tomentypnum nitens*, characteristic of calcareous fens, were identified (Hájek *et al.*, 2021).

3.3.2. Testate amoeba sub-fossil communities

Throughout core VAR1, a total of 105 distinct testate amoeba taxa were identified. The results of the testate amoeba analysis and reconstructions are presented in Figure 10. Three distinct zones were identified throughout the record by CONISS.

The first zone, extending from the base of the core to 44.5 cm (c. 1600 cal CE) is characterised by a mix of proteinaceous and mixotrophic species, including *Archerella flavum* and *Hyalosphenia papilio*. Other abundant species in this zone include *Diffflugia pulex*, *Phryganella acropodia*, *Assulina muscorum* and *Assulina seminulum*. The second zone, ending at 19.5 cm (c. 1980 cal CE), shows a replacement of the dominant species by *Galeripora discoides*, *Amphitrema stenostoma*, and xenosomic species such as *Cyclopyxis arcelloiodes*. By 35.5 cm (c. 1821 cal CE), mixotrophic species have almost disappeared, replaced by *Cyclopyxis euryostoma*, *Centropyxis aculeata*, *Diffflugia ampullula*, *Diffflugia brevicolla* and *Pseudodiffflugia gracilis*. The final zone, at the top of the core, is dominated by *Centropyxis elongata*, *Centropyxis sylvatica*, *Plagiopyxis* sp., *Euglypha rotunda* type, and *Euglypha leavis*.

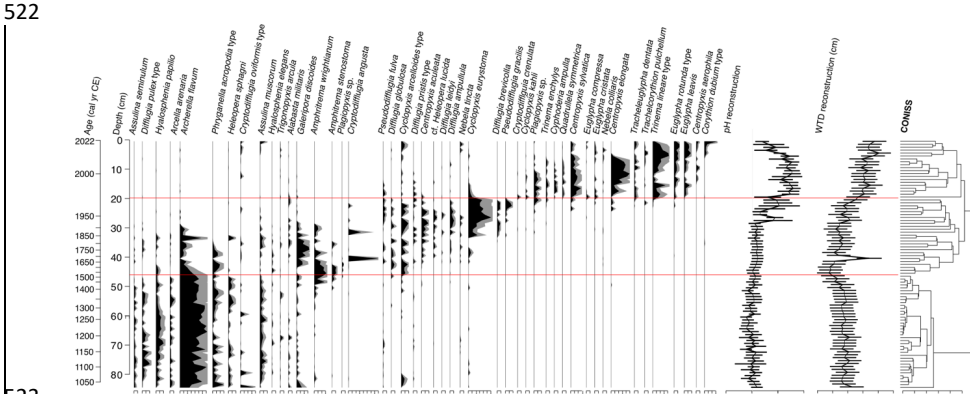
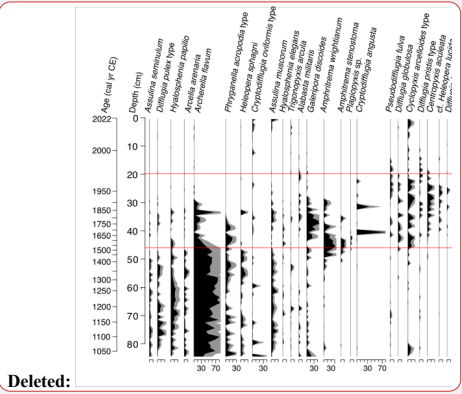


Figure 10. Stratigraphic plot showing changes in the relative abundance (%) of testate amoebae taxa identified throughout core VAR1, as well as reconstructed peat pore water pH and water table depths (WTD) and uncertainties. The results of CONISS are illustrated to the right of the figure, with horizontal red lines representing the zone boundaries defined by CONISS. Only species with maximum abundancies above 5% are illustrated. The full dataset is available in the supplementary data.

The relative abundance of testate amoeba taxa was used to reconstruct changes in water table depth and peat pore water pH over time. Both reconstructions exhibit notable trends throughout the past c. 1000 years. From the base of the core until c. 1330 cal CE, water table depth was relatively constant, averaging 7.0 ± 2.9 cm. Conditions became progressively wetter after this time, reaching a minimum of -2.5 cm by c. 1540 cal CE, possibly indicating a



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538 period of open water at the coring location. Water table depth began to decrease gradually,
539 accelerating after c. 1960 cal CE and reaching a maximum by c. 2012 cal CE.

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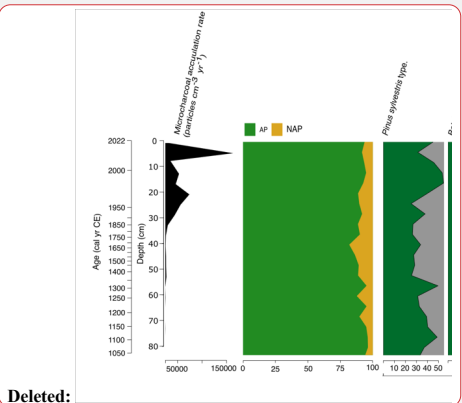
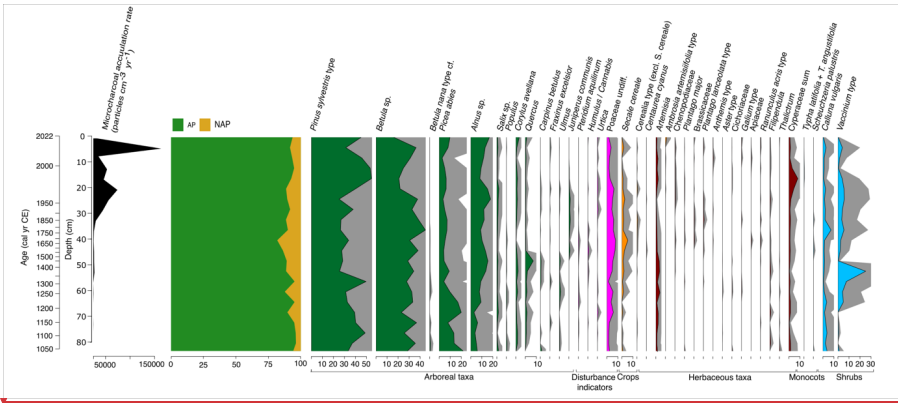
541 Reconstructed pH values were relatively stable throughout much of the record (4.7 ± 0.6).
542 Around c. 1850 cal CE, pH began to rise, reaching a maximum of 7.2 ± 0.6 by c. 1985 cal CE,
543 remaining high until the uppermost samples, where pH returned to near pre-disturbance
544 levels (4.8 ± 0.5), this change occurring after c. 1998 cal CE.

545 3.3.3. Pollen, spores, and non-pollen palynomorphs.

546
547 The results of the palynological analysis are illustrated in Figures 11 (Pollen and spores) and
548 Figure 12 (Non-Pollen Palynomorphs- NPPs). The pollen sequence from the core VAR1 is
549 dominated throughout by arboreal taxa, particularly *Pinus sylvestris* and *Betula* sp., with
550 lesser contributions from shrubs such as *Calluna vulgaris* and *Vaccinium* type. The high
551 dominance of these arboreal taxa and a low sampling resolution make detecting subtle shifts
552 in human activity in the palynological record difficult to infer (Favre *et al.*, 2008). Other
553 palynological studies from Estonia also report low variation in pollen assemblages and
554 dominance of *P. sylvestris* and *Betula* across different regions, even during periods of
555 significant land-use changes (e.g., Poska *et al.*, 2004; Veski *et al.*, 2005; Łuców *et al.*, 2022).

556
557 Figure 11. Stratigraphic plot illustrating the relative abundance of selected pollen taxa from core VAR1, as well
558 as microcharcoal (expressed as the total number of counted fragments). Relative abundances of arboreal taxa
559 are shaded green, disturbance indicators in pink, crops in orange, herbaceous taxa in brown and shrubs in teal.
560 Grey shading represents 5x exaggeration. Only taxa with a minimum abundance of 2% are shown.
561

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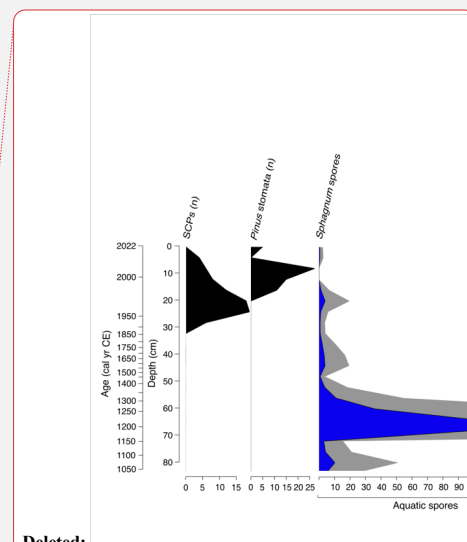
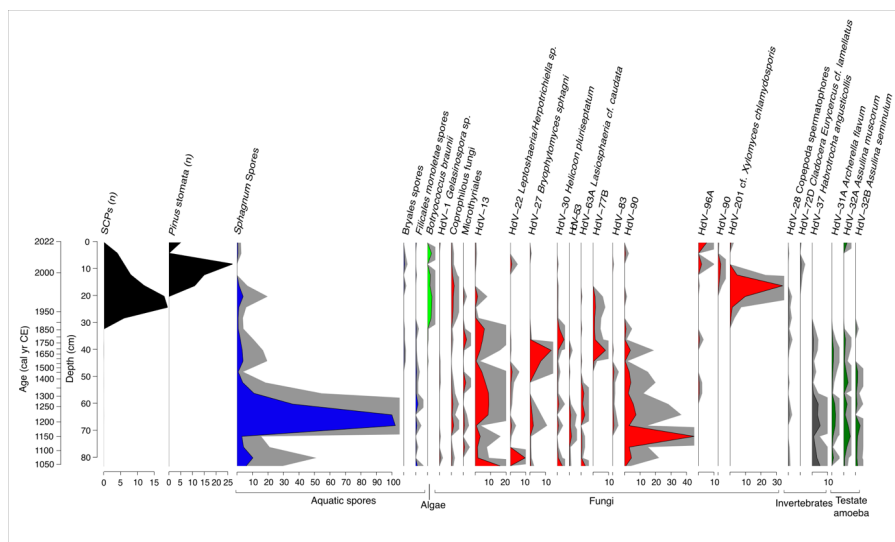


Figure 12. Stratigraphic plot illustrating the abundance of selected non-pollen palynomorphs from core VAR1, as well as spheroidal carbonaceous particles (SCPs) and *Pinus* stomata, both expressed as counts. Relative abundances of spores of aquatic taxa are shaded blue, algae: light green, fungi: red, invertebrate remains grey and testate amoebae in dark green. Grey shading represents 5x exaggeration. Only taxa with a minimum abundance of 2% are shown.

The oldest section of the core (c. 1000–1250 cal CE) provides little evidence of anthropogenic disturbance, indicating a largely forested landscape. This period is dominated by *Pinus sylvestris*, *Betula* sp. and *Picea abies*, suggesting a stable, predominantly woodland environment. After c. 1250 cal CE, there is a slight increase in pollen from grasses (Poaceae) from around 3% to c. 8% of the total assemblage until c. 1680 cal CE, as well as the appearance in low concentrations of plants indicative of disturbance, such as *Plantago lanceolata*, Chenopodiaceae and *Ranunculus acris* type and a general increase in *Secale* pollen which together may indicate agricultural activity, although wild populations of *Secale* are believed to have existed in Estonia (Veski, 1998; Poska et al., 2003). Despite this, arboreal pollen remains the dominant component of the record, with declines mainly affecting *Pinus sylvestris* (49 % at c. 1310 cal CE to 34% by c. 1680 cal CE) and *Picea abies* (16% at c. 1220 cal CE to 5% by c. 1680 cal CE). Tree species associated with early succession, such as *Betula*, remain largely unchanged, while *Alnus* increases during this period from c. 6% at the start of the record to c. 10% by c. 1680 cal CE. By c. 1680 cal CE, arboreal pollen has declined to its lowest relative abundance (around 82% of the total pollen sum). This decline is driven mainly by the changes in arboreal species, particularly declines in *P. sylvestris*, which becomes progressively less common through the core, a trend that continues until c. 1965 cal CE. By

c. 1680 cal CE a peak in cereals occurs, including *Hordeum*, although these are only present in low concentrations. The pollen record is stable after this period, with only slight (<1%) increases in *Calluna vulgaris* and *Vaccinium* type taxa occurring. These changes correspond with a rapid increase in microcharcoal accumulation rates, rising from 311 to 1027 particles $\text{cm}^{-3} \text{yr}^{-1}$ following the opening of the cement factory, peaking at 9837 particles $\text{cm}^{-3} \text{yr}^{-1}$ by c. 1970 cal CE. After this, *Pinus sylvestris* begins to recover, but the relative abundance of *Betula* sp. continues to rise. By c. 2010s cal CE, *Pinus sylvestris* becomes dominant once again, while *Betula* declines throughout the most recent samples, and microcharcoal accumulation reaches a maximum at c. 2010 cal CE, of 172237 particles $\text{cm}^{-3} \text{yr}^{-1}$.

The NPP record provides a more detailed picture of changes in the local environment than the pollen record. In the earliest portion of the record (c. 1000 - 1220 cal CE), fungi such as HdV-90 and HdV-13 (cf. *Entophlyctis lobata*) are common, and HdV-27 (*Bryophytomyces sphagni*) is present throughout. These fungi are typically associated with oligotrophic and ombrotrophic conditions, although HdV-90 can also thrive in more minerotrophic or poor-fen environments (van Geel, 1978; Kuhry, 1985).

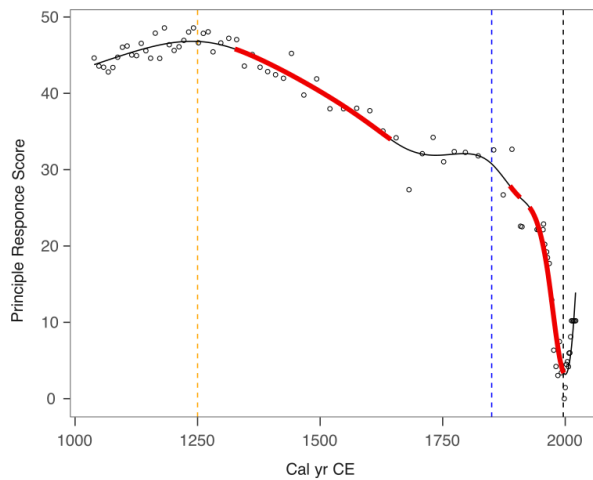
Between c. 1220 - 1460 cal CE, HdV-13 increases in abundance. Around 1850 cal CE, taxa associated with the earlier section of the record start to decline, being replaced by the microalgae *Botryococcus* taxa, indicative of aquatic conditions in addition to several fungi including HdV-55A (*Sordaria* type), HdV-112 (*Cercophora* type). The most notable species in the upper section of the core during this period is HdV-201 (cf. *Xylomyces chlamydosporis*), a wood-inhabiting fungus linked to freshwater environments or pool vegetation (Goh *et al.*, 1997; Kuhry, 1997). This species is especially abundant c. 2006 cal CE, comprising around 34% of the assemblage. After c. 1950s cal CE, the diversity of NPPs declines. A key species identified in this section is *Botryococcus braunii*, a green alga typically found in environments with high levels of inorganic phosphorus, which peaks around 1985 cal CE (Órpez *et al.*, 2009) suggesting a shift toward more nutrient-enriched conditions. Overall, the record suggests a transition from nutrient-poor, *Sphagnum*-dominated peat towards an increasingly nutrient-enriched system, followed by a change towards decomposers of ligneous material.

3.3.4. Rate of change analysis

629 The principal response curves (PrC) explained 79% of the variance in the sub-fossil testate
630 amoeba data from Varudi and are illustrated in Figure 13. The poor fit of GAM models to the
631 PrCs prompted us to use adaptive splines with a GAM, which are recommended for data
632 exhibiting abrupt changes (Burge *et al.*, 2023). However, adaptive spline GAMs cannot yet be
633 used within the GAM framework as described by Simpson (2018). One outlier was removed
634 due to the difference in testate amoebae composition owing to a high proportion of
635 *Cryptodiffugia angusta* (33.5 cm).

636 The PrC revealed two separate periods of change at Varudi, as well as highlighting periods of
637 rapid change associated with the onset of cement production in Kunda in 1871, and an earlier
638 change indicated from the palynological record to coincide with increasing human activity in
639 the landscape (Figure 11). Testate amoeba community composition was relatively consistent
640 from c. 1040 cal CE until c. 1330 cal CE and again between c. 1645 to c. 1886 cal CE. Following
641 c. 1998 cal CE, the direction of change in the PrC curve reverses, coinciding chronologically
642 with the installation of filters in the cement factory in 1996. By the end of the record, testate
643 amoeba community composition was characterised by the return of species such as
644 *Cyclopyxis arcelloides*, *Cryptodiffugia oviformis*, and *Assulina muscorum*, which were not
645 abundant since before c. 1970 cal CE.

647



648
649 Figure 13. Changes in principal response curve scores derived from sub-fossil testate amoebae data, modelled
650 using an adaptive spline generalised additive model GAM (black line). Solid red lines indicate periods of rapid
651 change, identified where the modelled confidence interval of the slope does not include zero. Vertical dashed
652 lines denote key chronological events potentially influencing environmental conditions at Varudi: The onset of
653 human activity recorded in the pollen data (c. 1250 cal CE, orange), the beginning of cement production in Kunda
654 (1871, blue) and the installation of pollution control filters at the cement factory (1996, black).

4. Discussion

4.1. Palaeoecological assessment of ecosystem and functional changes before, during, and after intensive alkaline dust pollution

The palaeoecological record of the Varudi peatland reveals three distinct phases occurring over the past 1,000 years: an ombrotrophic bog phase (c. 1000–1250 cal CE), a poor fen phase (c. 1250–1570 cal CE), and a wetland forest phase that continues to the present day. Unlike most contemporary research on peatland recovery, which has typically been conducted on historically drained, mined, or agricultural sites (Roderfeld, 1993; Graf *et al.*, 2008; Wagner *et al.*, 2008; Paal *et al.*, 2010). An advantage of our palaeoecological approach is that our study presents a unique pre-disturbance context for understanding the effects of atmospheric pollution upon Estonian peatlands.

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The different phases of development exhibited by the site in contrast with the different periods of anthropogenic activity and disturbance are summarised in Figure 14. During the early ombrotrophic phase, low palynological richness, minimal microcharcoal counts and abundant peat organic matter indicate a largely forested landscape with limited human impact near the site, consistent with other palaeoecological records throughout Estonia during this period (Veski *et al.*, 2005). The dominance of *Sphagnum* mosses and mixotrophic testate amoeba species such as *Archerella flavum* and *Hyalosphenia papilio* reflect the relatively undisturbed and productive nature of the bog during this phase (Marcisz *et al.*, 2020; Łuców *et al.*, 2022). During this period, water table depths were high, and pH levels were low, consistent with natural conditions for ombrotrophic bogs in the Northern Hemisphere (Warner and Asada, 2006).

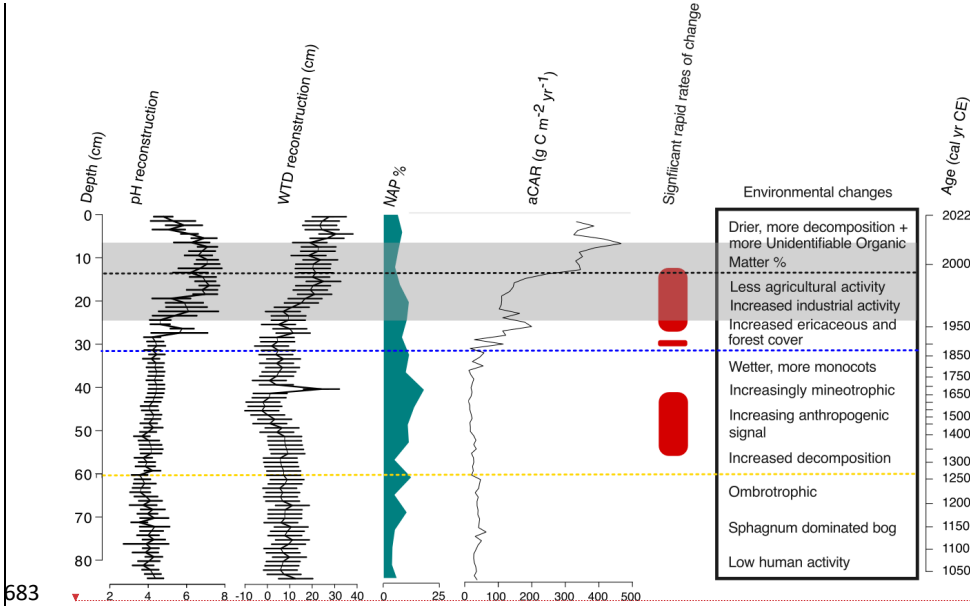
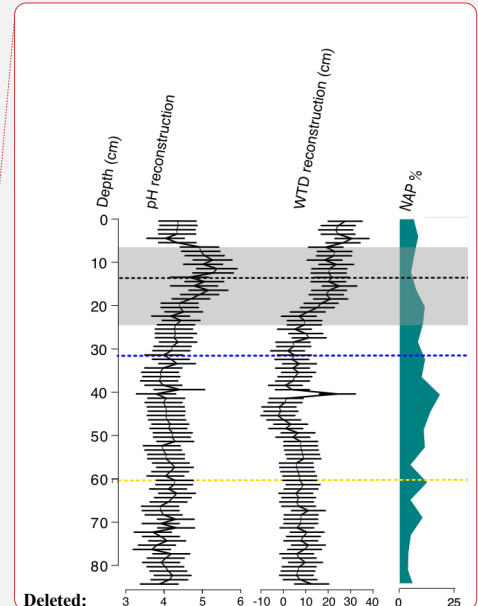


Figure 14: Summary of key trends in the Varudi peatland record. Shown are changes in reconstructed pH and water table depth, percentages of non-arboreal pollen (serving as a proxy for forest clearance related to agricultural activity), apparent carbon accumulation rates (aCAR), periods of significant change (as identified by principal response curves with Generalised Additive Models, GAMs), and a general timeline of environmental shifts. Dashed horizontal lines represent key historical events relevant to the Varudi record: the yellow line marks the approximate onset of human activity c. 1250 cal CE, possibly linked to the Livonian Crusades; the blue line indicates the establishment of the Kunda Cement Factory in 1871; the black line denotes the installation of effective filters at the Kunda Cement Factory; and the grey-shaded area highlights the section of the core with substantial cement dust, identified through LOI and μ XRF core scanning.

The beginning of agricultural activity around c. 1250 cal CE is broadly coincident with the German-Danish crusaders occupation of Estonia starting in 1227 CE (Veski *et al.*, 2005). From c. 1250 to 1570 cal CE, increased *Poaceae*, *Secale cereale*, and field weed pollen indicates a shift toward more open, meadow-like conditions (Poska and Saarse, 2002; Niinemets and Saarse, 2009), which may relate to the environmental effects resulting from economic changes resulting from the Teutonic order crusades, similar to those identified by Brown and Pluskowski (2011) in a peatland record from northern Poland.

Declines in *Pinus sylvestris* and *Picea abies*, alongside increases in early successional species such as *Betula* and *Alnus*, and minute increases in microcharcoal accumulation rates (Figure 11) suggest land clearance using slash-and-burn methods in the surrounding landscape (Jaats *et al.*, 2010). While arboreal pollen remained dominant, indicating continued forest cover around the Varudi peatland, mineral soil enrichment of the peat suggests overland flow or



707 aeolian deposition resulting from land clearance. Similar patterns of early human-induced soil
708 erosion have been recorded in peatland records from central Estonia (Heinsalu and Veski,
709 2010). This is reflected by decreasing organic matter content and increasing concentrations
710 of lithogenic elements such as Ca and Ti, patterns like those observed in Estonian lake
711 sediment records following land clearance events (Koff *et al.*, 2016; Vandel and Vaasma,
712 2018). Ti, a conservative lithogenic element, indicates soil erosion from the surrounding
713 catchment (Boyle, 2001; Boës *et al.*, 2011).

714 By c. 1570 cal CE, the site had transitioned into a wetter, more minerotrophic fen, indicated
715 by the gradual replacement of *Sphagnum* by monocots such as *Carex* sp. and *Eriophorum* ~~sp.~~
716 ~~increases~~ in *Sphagnum* ~~subsection~~ *Cuspidata* ~~and~~ subsequent declines in *Sphagnum*
717 subsections *Acutifolia* and ~~*Sphagnum* in the plant macrofossil record~~ (Figure 9). ~~This~~ shift
718 towards wetter conditions ~~is also supported~~ by the testate amoebae reconstruction, possibly
719 indicating permanent or seasonal standing water at this time (Figure 10). ~~and the presence of~~
720 ~~*Scheuchzeria palustris* pollen between c. 1500 – 1800 cal CE (Figure 11), a flood-tolerant~~
721 ~~species (Tallis and Birks, 1965)~~. This interpretation is consistent with observations from
722 contemporary bog pools, where *Sphagnum* is often rare or absent in shallow or temporary
723 water bodies on a peatland's surface, while monocots such as *Eriophorum* species are
724 abundant, as exemplified by the ~~British~~ National Vegetation Classification (NCV) bog pool
725 community classification (M3) (Rodwell, 1998). Nutrient enrichment due to soil
726 erosion resulting from forest clearance and agricultural activities in the landscape at this time
727 likely contributed to this shift (Hölzer and Hölzer, 1998).

728 The establishment of the Kunda cement factory in 1871 brought significant ecological changes
729 to the site. Substantial increases in Ti, Ca, Fe, and other elements associated with cement dust
730 fallout occurred along with ~~rapidly~~ increasing pH and declining organic matter content,
731 indicating the chemical influence of cement dust pollution upon the peat (Figures 7 and 8).
732 This pollution appears to have driven rapid changes in testate amoeba communities by c.
733 1890 cal CE, as seen in the PrC record (Figures 13 and 14). These results are consistent with
734 previous palaeoecological studies on alkaline pollution impacts in Estonian peatlands and
735 lakes (Varvas and Punning, 1993; Koff *et al.*, 2016; Vandel and Vaasma, 2018; Vellak *et al.*,
736 2014). The enrichment of the peat by cement dust is reported to have had a fertiliser-like
737 effect on ligneous and ericaceous species, increasing tree cover, size, and stand density within
738 the industrial region of Estonia at this time (Pensa *et al.*, 2004; Ots *et al.*, 2011).

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748 The concurrent hydrological changes experienced by the site, characterised by persistent
749 water table lowering after c. 1960, were likely driven by drainage and enhanced transpiration
750 rates following tree and shrub expansion onto the site (Stelling *et al.*, 2023). The simultaneous
751 compounding effect of hydrological change upon the site confounds interpretation of the
752 effects of alkaline pollution at Varudi peatland, as it is likely that the changes that occurred at
753 this time were driven by the combined effects of both disturbances.

754 From 1955 CE onward, cement production increased, reflected in peaks of Ca, Ti, and K,
755 alongside rising bromine (Br) levels, coincident with increased microcharcoal accumulation
756 rates (increasing from c. 11 to 58 particles yr⁻¹. The halogen Br is often linked to changes in
757 storminess in peat palaeoecological studies due to being commonly sourced from marine
758 aerosols (e.g., Shotyk, 1997; Turner *et al.*, 2014). Here, elevated Br levels correlate strongly
759 with cement dust indicators, suggesting that increased Br in the core corresponds with
760 enhanced plant decay and oxidation of abiotic organic matter, resulting in the formation of
761 methyl bromide (Lee-Taylor and Holland, 2000; Keppler *et al.*, 2000). The increased presence
762 of fungi in addition to microalgae species *Botryococcus braunii* in the palynological and plant
763 macrofossil records further suggests that the soil decomposition rate increased during this
764 period, coincident with periods of increased seasonal wetness (Barthelmes *et al.*, 2012;
765 Defrenne *et al.*, 2023; Buttler *et al.*, 2023; Thorman *et al.*, 2003) (Figure 12).

766 Following the restoration of Estonia's independence in 1991, more rigorous environmental
767 regulations resulted in a reduction of industrial emissions (Kask *et al.*, 2008). The effect of this
768 decline is evident in our palaeoecological record, with declining pollution markers and pH
769 beginning to return towards pre-disturbance levels after 1996 (Figures 7 and 10), as well as
770 the apparent (albeit limited) recovery of testate amoeba communities shown by the PrC curve
771 (Figure 13). Despite these improvements, the site has remained densely forested and
772 enriched, with a persistently lowered water table. We find no evidence of botanical
773 succession toward pre-disturbance conditions in the plant macrofossil record (Figure 9). The
774 presence of the calciphilous moss *Tomentypnum nitens* at the core surface likely reflects the
775 legacy of alkaline pollution at the site (Malmer *et al.*, 1992; Hájek *et al.*, 2006; Apolinarska *et*
776 *al.*, 2024). These cement deposits may still influence surface water chemistry and vegetation
777 through the upward movement of water enriched by the buried dust. Nonetheless,
778 *Sphagnum* has re-established at the site since its disappearance in the 1960s, suggesting a
779 degree of recovery. Identification and monitoring of the present-day flora could provide
780 additional insights into the extent of this recovery.

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4.2. Ecosystem Recovery After Reduction in Atmospheric Alkaline Pollution

Our study found evidence of recovery in PrC of the sub-fossil testate amoeba assemblages (Figure 13). The community composition of these environmentally sensitive proxies showed signs of turnover nearly 30 years after installing filters at the Kunda cement factory. However, the extent of recovery indicated by the testate amoebae is limited, as the community composition in the top sediment layer more closely resembles conditions from c. 1977 cal CE, the peak period of cement production at Varudi (Figures 3 and 13).

We used adaptive spline GAMs to analyse our data, which while suitable for datasets exhibiting abrupt changes such as the example from Varudi (Burge *et al.*, 2023), they cannot yet be incorporated into the Generalized Additive Mixed Model (GAMM) framework described by Simpson *et al.* (2018) and are more susceptible to issues such as temporal autocorrelation and boundary effects (Simpson, 2018; Burge *et al.*, 2023). While the model best fit our data, more evidence is needed to draw conclusive interpretations about recovery than these results alone. More recently accumulated samples may be more influenced by temporal autocorrelation (Burge *et al.*, 2023). Some testate amoeba species may infiltrate the sub-fossil community composition downcore, making near-surface communities appear more similar (Liu *et al.*, 2024). Another limitation of this study is that our results are based upon reconstructions taken from a single core and as such do not account for the spatial heterogeneity characteristic of peatlands. Although palaeoecological trends tend to be well replicated across multiple cores within a given site (e.g., Barber *et al.*, 1999; Hendon *et al.*, 2001), it may be that the trends reconstructed from VAR1 represent conditions specific to the sampling site, rather than the whole site (Bacon *et al.*, 2017). Paal *et al.* (2010) examined peatland vegetation communities in eleven bogs in northeastern Estonia at varying distances from sources of atmospheric alkaline pollution, finding evidence of *Sphagnum* re-establishment and the recovery of other bog-specific plant species at several sites as surface waters re-equilibrated to pre-pollution conditions, particularly within microforms that are less affected by contaminated groundwater. Therefore, recovery at Varudi may be spatially heterogenous, with some areas recovering more quickly depending upon their contact with polluted peat layers.

Studies investigating the recovery of vegetation communities following alkaline pollution show that ecosystem recovery may be limited due to the present, degraded condition of the

ecosystem. For example, Vellak *et al.* (2014) studied bryophyte recovery following reductions in atmospheric fly-ash pollution across Estonia and northwestern Russian sites. They found that bryophyte species growing in more heavily affected sites tended to be adapted for growing in low light conditions, due to competing with the larger and more dense vascular plants that encroached on these sites, in response to the more enriched and alkaline conditions for which they are better adapted than bog-specific vegetation (Pärtel *et al.*, 2004; Zvereva *et al.*, 2008a, 2008b). Increased nutrient and litter supply and root exudates, coupled with the faster growth of vascular plants and trees may further hinder the reestablishment of bog and fen communities, delaying or preventing recovery (Konings *et al.*, 2019). Gunnarsson *et al.* (2002) demonstrated that nutrient enrichment, often accompanied by higher pH, can give vascular plants a competitive advantage over bog-specific vegetation (Limpens *et al.*, 2003; Dieleman *et al.*, 2015).

4.3. How has the pollution impacted apparent rates of carbon accumulation at the site?

Each of the phases in ecosystem development in the palaeoecological record corresponds with substantial changes in the apparent carbon accumulation rate (aCAR) throughout the Varudi record (Figure 7). Between c. 1000 to 1250 cal CE, when the site was in a relatively pristine, undisturbed condition, aCAR rates were higher ($32.0 \pm 10.7 \text{ g C m}^{-2} \text{ yr}^{-1}$) than the average for northern hemisphere ombrotrophic peatlands ($23 \pm 2 \text{ g C m}^{-2} \text{ yr}^{-1}$) (Korhola *et al.*, 1995; Yu *et al.*, 2009). Throughout the transition from bog to poor fen, from c. 1360 to 1570 cal CE, average aCAR rates most likely initially increased ($38.5 \pm 34.3 \text{ g C m}^{-2} \text{ yr}^{-1}$). However, they were highly variable, likely reflecting the incorporation of vascular plant roots from the overlying forested phase, introducing younger carbon into deeper sediments. Between c. 1630 and 1800 cal CE, rates fell as low as $19.0 \pm 5.4 \text{ g C m}^{-2} \text{ yr}^{-1}$, aligning with reported values for Finnish fens (Korhola *et al.*, 1995). Overall, minerotrophic fens typically exhibit lower carbon accumulation rates than ombrotrophic bogs (Loisel and Bunsen, 2020; Yang *et al.*, 2023).

Following the succession from poor fen to forested fen from c. 1871 cal CE to the present, there is an apparent increase in aCAR, especially in the most recently accumulated peat ($280.7 \pm 211.3 \text{ g C m}^{-2} \text{ yr}^{-1}$). Due to the incomplete decomposition of labile organic matter at the peat surface, care must be taken when interpreting recently accumulated carbon from peat core records (Young *et al.*, 2019; 2021). This increase is likely due to high litter deposition from

865 trees and shrubs, and the rapid decline in aCAR downcore suggests that this labile material is
866 rapidly cycled back into the atmosphere rather than sequestered in the soil. Our results
867 suggest that the current vegetation composition, dominated by trees and shrubs, is less
868 effective for long-term carbon sequestration compared to earlier phases. While the water
869 table remains reduced, the site shows limited potential for recovery to its original
870 ombrotrophic condition due to ongoing drainage. At the same time, nutrient inputs and
871 evapotranspiration from the overlying trees compound this limiting factor.

872 4.4. Indicators of critical thresholds to assess peatland condition
873 and recovery due to alkaline pollution
874

875 By comparing our pH reconstruction with significant successional shifts at the site, we may
876 infer the thresholds at which transitions in the steady state of the ecosystem occurred in
877 response to changes in alkalinity at Varudi bog. Defining tipping points that may be broadly
878 applied to other sites allows for predicting ecosystem shifts in response to global change,
879 allowing for prediction of such shifts in the future (Munson *et al.*, 2018). To our knowledge,
880 our study represents the first step towards defining thresholds for tipping points in peatland
881 ecosystems in response to changes in alkalinity.

882
883 Around c. 1910 cal CE, the reconstructed pH for Varudi showed an increasing trend from the
884 previous, relatively stable conditions, reaching a maximum of 7.2 ± 0.6 by c. 1985 cal CE
885 (Figure 10). This value aligns with previous pH measurements for forest litter in the zone
886 surrounding the Kunda cement factory of 7.1 – 7.4, supporting the reliability of our
887 reconstruction (Paal and Degtjarenko, 2015). We find that the transition from an
888 ombrotrophic bog to fen-like conditions during the mid 13th century, driven by mineral soil
889 enrichment resulting from land clearance for agriculture in the surrounding landscape, was
890 associated with an increase of average pH levels from c. pH 3.9 to pH 4.1 which represents
891 (due to the logarithmic scale of pH) more than 1.5 times increase in alkalinity. In contrast, the
892 subsequent shift from fen-like to forested conditions corresponded with a small average
893 increase in pH to c. 5.7, over 40 times more alkaline than the previous state and more than
894 60 times more alkaline than during the bog phase. Our results suggest that while substantial
895 increases in pH can drive rapid and substantial ecosystem regime shifts, relatively small
896 increases in pH (increases in pH of 0.2 – 0.3) may also drive critical ecosystem transitions that
897 may be more gradual but can have equally as significant and long-lasting impacts upon

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915 peatland ecosystem functioning, such as the capacity for carbon accumulation. However, we
916 stress that the threshold values defined here remain uncertain, owing to the relatively
917 significant uncertainties associated with transfer function models in general (Amesbury *et al.*,
918 2016). Higher pH values measured from bog pools at the site in 1996 – 1997 of 7.6 - 8.5 are
919 slightly higher than our estimates for this period (6.1 - 6.6), and by 2022 pH had only fallen to
920 6.3 - 6.8 (Pärtma, 2023) although our reconstruction indicates pH had fallen to 4.8 by this
921 time, suggesting that our reconstruction, despite having a strong predictive power,
922 underestimates pH levels experienced by the site, Alternatively, this may simply reflect local
923 variation in pH within Varudi, as the pH measurements were taken from bog pools in the NW
924 margin of the bog, not adjacent to the coring location. This may warrant future re-evaluation
925 of our data using transfer functions specifically developed for reconstructing pH in polluted
926 peatlands. In addition, we cannot rule out the possibility that other impacts of cement
927 deposition (e.g., reduced photosynthesis rates due to dust deposition) and, significantly-
928 drainage; may have also played a role in driving botanical changes.

929
930 Contemporary field and lab experiments that manipulate the alkalinity of peatland soils
931 directly may provide more precise values for ecosystem thresholds in response to alkaline
932 pollution. However, to date few studies have done this for alkalinisation: one example is Kang
933 *et al.* (2018) who conducted a series of field and laboratory pH manipulation experiments
934 across seven peatlands in the UK, Japan, Indonesia and South Korea, finding that increases in
935 pH resulted in significant changes in microbial composition, resulting in increased phenol
936 oxidase activity and enhanced DOC releases. Another is the long-term (active since 2002)
937 Whim Bog experiment near Edinburg, Scotland (Sheppard *et al.*, 2011; Levy *et al.*, 2019),
938 where the effects of enrichment upon a peatland by gaseous ammonia (NH₃) and wet-
939 deposited ammonium (NH₄⁺) and nitrate (NO₃⁻) are compared. Analysis by Sutton *et al.* (2020)
940 indicates that vegetation declined three times more quickly when exposed to gaseous
941 ammonia and three times for ammonium than for a similar dose of nitrate.

942
943 A significant limitation of these studies in defining tipping points is that the long timescale
944 necessary for critical transitions to occur in some cases typically exceeds the lifespan of most
945 observational and experimental studies (Taranu *et al.*, 2018; Lamentowicz *et al.*, 2019).
946 Therefore, further palaeoecological work across a more extensive range of sites or with
947 multiple cores from within a single site may also advance our understanding of ecosystem

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Deleted: limited predictive ability of the transfer function models used for our pH reconstruction and the

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955 tipping points in response to alkalinisation. Furthermore, disentangling the effects of
956 alkalinisation from those of drainage is challenging, as both factors may influence plant
957 community composition, carbon accumulation rates, overall ecosystem functioning (Word *et*
958 *al.*, 2022) and possibly resilience to alkalinisation in peatlands. Since significant drainage and
959 cement production appear to have begun at roughly the same period in the site's history,
960 some of the ecological variability attributed here to cement dust pollution may instead be the
961 result of drainage impacts. However, it is notable that despite reconstructed pH values
962 decreasing following the cessation of significant emissions from Kunda, water table depths
963 have not recovered. This indicates that drainage is unlikely to have influenced the apparent
964 modest ecosystem recovery observed at Varudi in recent decades.

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966 5. Conclusion

967

968 This study is the first to investigate the long-term impacts of alkaline emissions on a peatland
969 over centennial timescales. It establishes the first attempt to define threshold indicator values
970 for ecosystem tipping points in response to alkalinisation. Our findings demonstrate that
971 alkaline pollution, along with later drainage, has had a profound influence upon ecosystem
972 development at Varudi for more than 750 years, with ecosystem succession following both
973 low-level, sustained mineral soil enrichment due to agricultural activities and intensive fly-ash
974 fallout sustained over 160 years. We find that an increase in pH of 0.2 to 0.3 (corresponding
975 to, approximately, a two to threefold increase in alkalinity) is sufficient to trigger a critical
976 ecosystem shift, while an increase of 1.5 pH units (a ~40 x increase in alkalinity) will cause
977 rapid ecosystem transition, resulting in the loss of *Sphagnum* mosses and changes in
978 ecosystem functioning. This can lead to a long-term decline in carbon accumulation over long
979 timescales, and such changes may be slow to recover or permanent, even if the point source
980 of pollution is eliminated. Our results, while insightful, have limitations that underscore the
981 need for additional experimental and palaeoecological research to assess peatland responses
982 and resilience to alkalinisation across a range of spatial and temporal scales. This would allow
983 a better understanding of the timescales required for peatland recovery and how these
984 ecosystem transitions influence greenhouse gas dynamics from affected sites. Alkalinisation
985 poses a growing threat to peatlands worldwide and is a developing challenge for the 21st
986 century (Sutton *et al.*, 2020). As carbon-rich ecosystems, peatlands are important for
987 regulating future atmospheric greenhouse gas concentrations. Understanding how peatlands

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991 will respond to future alkalisation is essential for predicting the role of peatlands in climate
992 change mitigation.

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994
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999 Data availability statement:

1000 All relevant data files described here are available either in the supplementary materials or
1001 via FigShare, accessible via the following link: [10.5194/egusphere-2025-1351-supplement](https://figshare.com/s/10.5194/egusphere-2025-1351-supplement),

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1002 Author contributions:

1003
1004 LOA prepared the manuscript with contributions from all authors. ML, MS and SV
1005 conceptualised the study and secured funding. LOA, ML, MS, KM, LA and NS participated in
1006 fieldwork. LA, SC, MB, ML, PK, MKK, EŁ, AC and MAL conducted formal analyses. SV and AH
1007 provided historical data and photographs.

1008 Declaration of interests:

1009 The authors declare they have no known financial interests or personal relationships that
1010 could influence the work represented in this manuscript.

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