

Urban basin deposits

the pollution history of Lille Lungegårdsvannet, Bergen

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Abbreviations

BP	Before Present (AD 1950)
GEP	Good Ecological Potential
HMWB	Heavily Modified Water Body
LL	Lille Lungegårdsvannet
NGU	Geological Survey of Norway
PAH	Polycyclic Aromatic Hydrocarbons
PAH _{sum16}	PAH, 16 US-EPA compounds
PCB	PolyChlorinated Biphenyls
SEM	Scanning Electron Microscope
TOC	Total Organic Carbon
WFD	Water Framework Directive
WWI	World War One
XRD	X-ray Diffraction

Abstract

The sediments within urban lakes will, if left undisturbed, reflect a part of the history of the city through the record of chemical compounds. The sediment chemistry in Lille Lungegårdsvannet in Bergen city is foremost anthropogenically influenced through both local point and diffuse pollution sources, but also to a large extent by physical changes, such as variation in the sediment input and the physical lake environment. Human input appears to have increased the sedimentation rate considerably as habitation and industrial activities moved closer to the lake. The surroundings of Lille Lungegårdsvannet have been characterised by varied, small-scaled crafts and industrial activities since the 17th century.

¹⁴C dating and historical information about pollution sources in Bergen provide the framework for the chronological interpretation. However, potential reworking of terrestrial macrofossils may preclude the sole use of this dating technique. By using several organic compounds, metals and cyanide in addition to the ¹⁴C dating, we provide an additional chronological constraint on the upper sediment sequence. However, dredging or erosion of the sediments has potentially created several vacuities/hiati that complicate the chronological interpretation.

Metal concentration displays a large anthropogenic influence as the metal input has been considerable at intervals. Pb, Hg, Zn, Cu and Fe show especially large fluctuating changes. Some of the concentration variability can be attributed to changes in grain size characteristics, but periods of reduced metal input, due to documented economic depression periods, can also be observed. Sediment grain size correlates negatively with concentration for all elements, except for Cd. Elements such as Fe, Ni and Cr, on the other hand, display strong associations with the geological setting around the lake.

Elevated concentrations of PAH-compounds coincide with gaswork activity, urban fires, domestic heating and, most recently, to traffic pollution. Changes in PAH compound patterns indicate changes in sources from low-temperature combustion processes towards high-temperature processes. PAH compound ratios indicate that the sources have shifted from pure wood combustion sources in pre-industrial times towards mixed and petrogenic sources in more recent times.

Data from stormwater culvert sediments strongly suggests that active sources of all investigated compounds still exist in the urban environment, including PCB - even 30 years after their usage ban. Pollutants spread into the lake through different pathways; within this study stormwater or urban runoff has been a focus of investigation and point to the presence of a large amount of suspended sediments that accompany stormwater being emitted into the lake.

Sammendrag

Sedimenter fra innsjøer i byer eller bynære områder kan, hvis de blir liggende uforstyrret, gjenspeile byens kjemiske historie ved at det utføres kjemiske analyser på daterte sedimenter. Sedimentkjemien i Lille Lungegårdsvannet i Bergen sentrum har hovedsakelig vært påvirket av mennesker via lokale forurensningskilder, men i stor grad også av fysiske endringer, for eksempel endringer i sedimenttilførsel og det fysiske miljøet. Lille Lungegårdsvannet har siden 1700-tallet vært omgitt av varierte industrielle aktiviteter. Økt menneskelig tilførsel, samt en høyere biologisk aktivitet i innsjøen økte sedimentasjonsraten betraktelig da bosetting og næringsvirksomhet flyttet til områdene i nærheten av innsjøen.

¹⁴C-dateringer og historiske opplysninger om forurensningskilder i Bergen ga rammene for den kronologiske tolkningen. De sediment som har blitt påvirket av f.eks. mudring og andre inngrep gir imidlertid usikre resultat da analyserte terrestriske makrofossiler muligens har blitt resedimentert. Ved å bruke kjemiske analyser av flere organiske miljøgifter, metaller og cyanider i tillegg til ¹⁴C-dateringer, blir en alternativ kronologisk tolkning av sedimentene i den øvre sedimentsekvensen introdusert. Imidlertid har potensielt mudring og/eller erosjon av sedimentene gitt flere hiati som vanskeliggjør den kronologiske tolkningen.

Resultatene for metallkonsentrasjonene viser for en stor del menneskelig påvirkning som kan påvises i sedimentene. Spesielt Pb, Hg, Zn, Cu og Fe viser store variasjoner. Noe av konsentrasjonsvariabiliteten kan tilskrives forandringer av kornstørrelse. Redusert metalltilførsel kan også knyttes opp mot en økonomisk nedgangsperiode. Kornstørrelsen korrelerer negativt med konsentrasjon for de fleste element, med unntak av Cd. Elementer som Fe, Ni og Cr, derimot, viser sterke assosiasjoner med den lokale geologien.

Forhøyede konsentrasjoner av PAH-forbindelser sammenfaller med gassverkets aktivitet som indikerer gassverket som en hovedkilde. Andre PAH-kilder er bybranner, boligoppvarming og trafikkforurensning. Endringer i PAH-forbindelser indikerer endringer i kilder fra lavtemperatur forbrenningsprosesser mot høytemperatur-prosesser. Forholdet mellom utvalgte PAH-forbindelser viser at kildene har skiftet fra forbrenning av tre i før-industriell tid mot blandede kilder og oljeforbindelser i nyere tid.

Sandfangsmasser viser til at aktive forurensningskilder fortsatt eksisterer i bymiljøet for alle de undersøkte forbindelsene. Miljøgifter spres i sjøen via ulike baner. Denne studien har fokusert på overvann eller urban avrenning. Resultatene peker på en stor mengde suspenderte sedimenter som følger overvann som deretter slippes ut i innsjøen.

List of publications

Andersson M, Bogen J, Ottesen R T & Bønsnes T E. Polychlorinated biphenyls (PCB) in urban lake sediments and migration potential from urban stormwater in Bergen, Norway. Submitted to Journal of Environmental Engineering.

Andersson M, Klug M, Eggen O A & Ottesen R T (2014): Polycyclic aromatic hydrocarbons (PAHs) in sediments from lake Lille Lungegårdsvannet in Bergen, western Norway; appraising pollution sources from the urban history. Science of the Total Environment 470–471: 1160–1172.

Andersson M & Eggen O A. Urban sprawl reflected in inorganic pollution in urban lake deposits, Bergen, Norway. Submitted to Applied Geochemistry.

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Note on contributions

Paper I: Polychlorinated biphenyls (PCB) in urban lake sediments and migration potential from urban stormwater in Bergen, Norway

Authors: Malin Andersson, Jim Bogen, Rolf Tore Ottesen, Truls E. Bønsnes

The candidate wrote the paper and Jim Bogen aided with the results of the stormwater drainage study. Jim Bogen, Truls Bønsnes and Rolf Tore Ottesen designed and carried out the drainage study in cooperation with Bergen City.

Paper II: Polycyclic aromatic hydrocarbons (PAH) in sediments from lake Lille Lungegårdsvannet in Bergen, western Norway; appraising pollution sources from the urban history

Authors: Malin Andersson, Martin Klug, Ola Anfin Eggen, Rolf Tore Ottesen

The candidate wrote this paper with significant input from Martin Klug, who carried out the modelling of the results from ^{14}C dating in collaboration with the first author. Ola Anfin Eggen provided statistical work. He and Rolf Tore Ottesen aided in the interpretation of the results, suggested the analytical program and reviewed the manuscript.

Paper III: Urban sprawl causing inorganic pollution in urban lake deposits in Bergen, Norway

Authors: Malin Andersson, Ola Anfin Eggen

The first author carried out all sample preparation and writing of the manuscript. Ola Anfin Eggen provided the factor analysis and reviewed the manuscript.

Paper IV: PCB contamination from sampling equipment and packaging

Authors: Malin Andersson, Rolf Tore Ottesen, Morten Jartun, Ola Eggen, Ann-Christin Enqvist

The first four authors worked together in projects where the problem of contamination came up, so all four were involved in the design of the analytical program. Ann-Christin Enqvist carried out the analyses. The first author wrote the manuscript.

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1. Introduction

Urban lakes differ from naturally occurring lakes in many aspects and are, according to Schueler and Simpson (2001) and Naselli-Flores (2008), defined by several factors: I) Urban lakes tend to be smaller and shallower than natural lakes. II) They commonly have a large drainage area ratio, meaning that their drainage areas exert a strong influence on the lake. III) The drainage area is also, to some degree, covered by impervious surfaces and thus the drainage is dominated by urban runoff. IV) Urban lakes are often man-made and managed for recreation, water supply, flood control or some other direct human use. V) They receive higher nutrient loads than natural lakes and are thus often eutrophic. Urban lakes deserve attention as they provide an important public contact to water due to a year-round accessibility (Birch and McCaskie, 1999).

Several sediment studies in urban areas have investigated trends in metal concentrations in cores from urban lakes to uncover the effects of changes within legislation, regulations and industrial practices or to determine the contribution of local pollution point sources (Long et al., 2003; Meriläinen et al., 2003; Mahler et al., 2006; Parviainen et al., 2012). Others have studied the speciation of heavy metals as they occur in urban systems and how this differs from natural systems (Baek and An, 2010; Gupta et al., 2013) or have conducted studies to verify the pollution status of the urban sediments (Von Gunten et al., 1997; Charlesworth and Foster, 1999; García-Rodríguez et al., 2007; Das et al., 2008; Hu et al., 2011). According to Schueler and Simpson (2001) it is rare to find studies that have tracked changes in lake environments that are attributed to development within the drainage area over time. Instead, the influence of the drainage area on its lake is often treated as a constant and the attention is instead often paid to factors such as trophic state or geomorphic origin within each individual lake.

Sediment studies in Norway have mostly been conducted on natural lakes (e.g. SFT 1999, 2006 and 2008) as a part of nationwide studies with pollution monitoring as the main aim. Sediment studies in urban areas, on the other hand, have been conducted on marine sediments (e.g. Lepland et al., 2010; Arp, 2011), as most towns are located along the coast.

Lille Lungegårdsvannet (LL), which falls into the urban lake category, according to the criteria by Schueler and Simpson (2001), was a topic on the City of Bergen's agenda in 2008 concerning odour as well as visual issues (colour of the lake water). The previous year the lake had again experienced a major algal bloom and the water emitted an unpleasant odour that required a determination of the quality and status of the sediments on the lake bottom. Previous studies determined that the main cause of the odour was the nutrient status of the water and the surface sediments. Hanssen (2009) estimated the amount and thickness of the sediments. However, an assessment and understanding of the chemistry of the sediment column and the amount of sediments entering the lake today was required for the City Council to be able to make decisions about the future of the lake. More information was required, irrespective of whether remediation of the lake was undertaken. The City of Bergen extracted some samples from the cores and then handed over the cores to Knut Krzywinski at the Department of Biology at the University of Bergen for cultural history studies. NGU was then approached by the municipality and asked to make a more thorough qualitative study of the lake sediments. This forms the basis for the current project.

Pollution can be defined as the introduction of elements and compounds into the environment that attain concentrations that differ from the natural, local background levels. The pollution type and severity will change as the character of the pollution sources evolve over time. The introduction of new compounds does not always come with an understanding of future effects to the environment and human health. For example, the effects of the introduction of PCB into construction materials which lead to costly remediation projects some decades later, were originally unknown.

LL is not only an urban water body. It is probably one of the most important and complete historical archives of urban Bergen. Over the past century, it has been proposed on several occasions that the lake be filled in to provide more urban space, provoking public outcry. Urban evolution around the lake and the subsequent change in use of the lake has transformed its size and input. In effect, the urban history of Bergen has therefore been recorded in the chemical history of the sediments.

1.1 Objectives of the thesis:

- This study aims to gain an understanding of the anthropogenic influences on the lake sediments and in which elements or compounds this influence is displayed. In addition, we will unravel both why and when this anthropogenic influence occurred. We will also establish which elements display an increased concentration due to human activity and which elements are due to natural sources. This information is vital when planning remediation of the lake.
- Evaluate the amount and chemical quality of the suspended sediments entering the lake via urban runoff and whether an increased influx of urban runoff results in "healthier" surface sediments. Studies have been conducted on the chemical quality of stormwater, but no long-term studies have determined the amount of suspended sediments that are released from urban, impervious surfaces.
- Understanding the connection between historical events, industrial history and the chemistry of the sediments over time. Many sediment studies have a focus on long- range transported pollution as a means of understanding pollution spread via the atmosphere. This study, however, connects the local sources as the main pollution factors.

1.2 Bergen City

Bergen was first mentioned as a town by King Magnus Berrføtt during his travels in Ireland in AD1098 and the foundation of the town has been connected to king Olav

Kyrre and the year 1070 (Helle, 1982). Palaeobotanical investigations indicate that the town started as a farming community that gradually evolved into a settlement between AD700 and 1000 (Krzywinski and Kaland, 1984; Hjelle, 2001). The settlement and its fortifications, called Bergenhus, were concentrated on the eastern side of Vågen during the early history of the town. Habitation spread further inland, but also beyond the sea front as new land was reclaimed (Figure 1). The urban centre, which acted as the capital in the 13th century and a Hanseatic town in medieval time, has always kept its status as a busy international port and trading community.

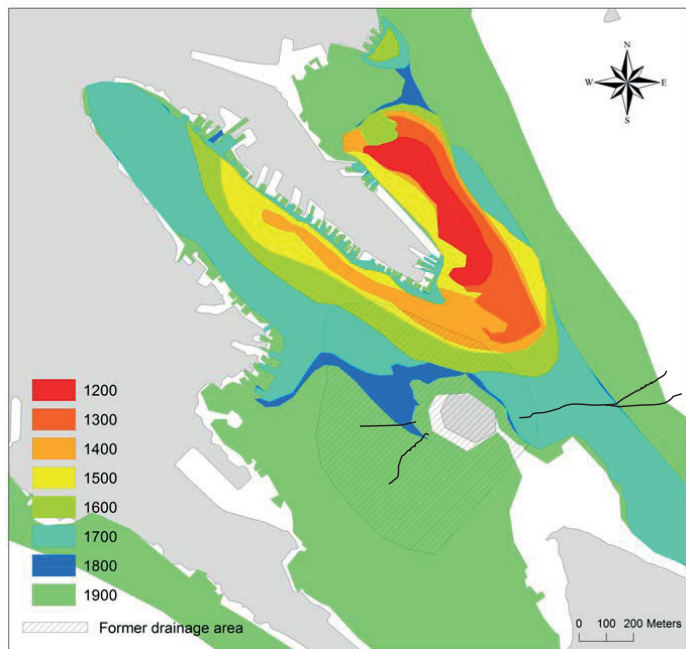


Figure 1. Evolution of habitation in Bergen from the 13th to 20th century with the approximate former drainage area (Johnsen, 2008) and streams draining into LL (Helle, 1982) indicated. LL has shrunk over the centuries into its current, manmade shape (adapted from maps constructed by the Cultural heritage Division of Bergen City and www.bergenskart.no).

The "town law" of 1276 by King Magnus Lagabøte stated that crafts in Bergen that produce large amounts of waste, such as tanneries and shoemakers, were to be located on the periphery of the town. It also forbade deposition of waste in the

harbour indicating that such activity was a public problem. The law dictated the placement of different crafts around Vågen (Figure 2), although from the 17th century these placement regulations were no longer followed and the crafts were distributed all over the town (Kjellberg and Stigum, 1936). Bergen was the biggest town in Scandinavia until the 17th century and remained Norway's biggest town until the mid 19th century, hosting a lively crafts industry.

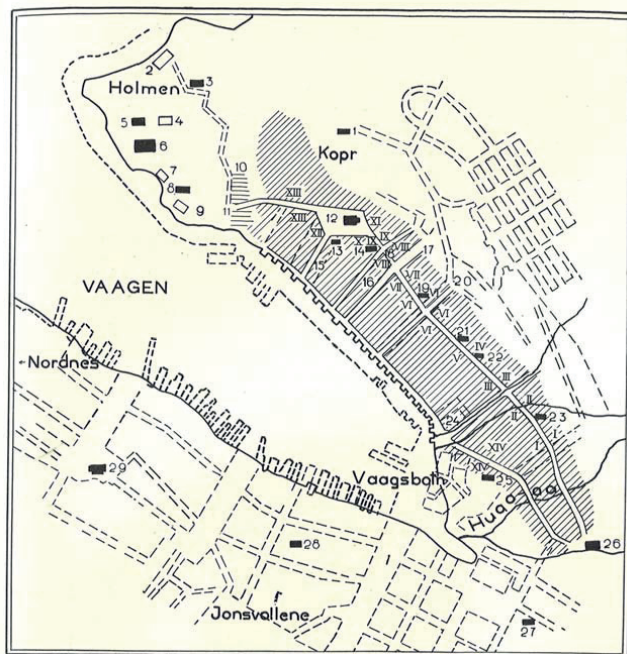


Figure 2. Map of Bergen in the Medieval period that positions the living quarters of the various craft unions: I) shoemakers, II) coopers, III) skin craftsmen, IV) goldsmiths, V) comb makers, VI) food grocers, VII) cloths salesmen, VIII) painters and saddlers, IX) tailors, X) metal workers, XI) coffin makers, XII) shield makers, XIII) food grocers, XIV) various dealers (modified from Grieg, 1936).

Sediments and deposits in central Bergen are an important part of the cultural heritage as so much of the town's history is buried deep within these. Vågen, with its much more central position, potentially contains sediments with a much earlier anthropogenic influence than LL, as the Vågen basin already received substantial amounts of waste in medieval times. These regularly had to be dredged from the

Middle Ages onwards to allow for larger vessels to moor in the harbour, thus only part of the sediments are present today. The sediments in Vågen were also partly removed as a ship exploded in the harbour in April 1944. The remaining sediments have since been eroded by propeller jet water from modern ships. Therefore, the sediment stratigraphy may have been destroyed by several processes and not been preserved intact, whereas the sediments of LL are presently considered as one of the best preserved sediment basins that contain material that can characterise the urban history of Bergen.

1.3 Lake sediments, what do they reflect?

Lakes are the receivers of material that has been eroded from, produced in, or transported to the drainage area of the lake (the allochthonous component). The lake water itself is a producer of biological material and minerals, and diagenetic processes within the sediments produce changes in the sediments (the autochthonous component). In addition to the input of terrestrial material through erosion or transport by water is the atmospheric input. The influx of anthropogenically influenced material in urban areas often overprints the signal from the natural input. The occurrence of polluting sources within urban drainage areas adds pollutants from several pathways, for example through the atmosphere and stormwater.

Lakes are considered low-energy environments, where no major streams or currents erode or re-suspend sediments or transport sediments out. Therefore lake sediments are considered a suitable sampling media that represent a historic archive. The stratigraphy of lake sediments reflects changes in biological, lithological and chemical processes over time in the drainage area and beyond. However, several assumptions are made when defining the lake sediments as accurately reflecting the chemical environment during deposition. There is a tacit understanding that elements and compounds are transported through the water column rapidly, and that supported downward transport by organic matter and complexes do not result in substantial removal of them (Van Metre and Mahler, 2010). Furthermore, it is assumed that the

metals cannot have gone through any significant post-burial remobilisation and that they accurately reflect the chronological conditions (Boyle, 2001). For example, biodegradation may affect PAH in sediments, but should be less pronounced in anoxic sediments (Gschwend and Hites, 1981). In addition, anoxic conditions do not promote bioturbation and thus chemical timelines are more accurately preserved. In consequence, lakes with high organic matter content in the water and sediment columns that contain anoxic conditions have the most potential to preserve the incoming chemistry (Gschwend and Hites, 1981; Van Metre and Mahler, 2010). Eutrophic lakes provide ideal conditions for large sedimentation rates as a considerable amount of settling algae exist in the water column and where the microbial degradation of organic carbon is insufficient (Berglund et al., 2001).

The deposition of natural sediments that contain no anthropogenic elements gives rise to background concentrations that define the local chemical concentration range. Within an urban environment these background concentrations will increase due to anthropogenic influence. A contentious issue within geochemistry is the definition of when background conditions are encountered. Sediment quality criteria in Norway, which are used when performing risk assessments, class 1 (insignificantly polluted) are defined as "background concentrations" (Bakke et al., 2010). Many sediment studies extend no more than 100 years, referring to low values after a drop in concentrations as "background values", without considering that the low values could be part of a fluctuating concentration pattern. This may in some studies correspond to sediments deposited before the mid 1800's or even later (e.g. Alve et al., 2009). Some authors define lake sediments from remote areas as background values (e.g. Förstner and Müller, 1981). In contrast, several studies have extended the core sampling well before the onset of industrialisation to obtain samples deposited considerably before the polluting influence of industry (e.g. von Gunten et al., 1997; Garbe-Schönberg et al., 1998). These studies demonstrate that low concentrations not necessarily reflect a pre-anthropogenic environment, but could represent fluctuations in concentrations that reflect changes in sediment deposition or grain size. Therefore, for most

elements, sampling must be conducted considerably far back in time from estimated anthropogenic influence in order to ensure accurate background values.

1.4 Lille Lungegårdsvannet (LL) - the study site

LL is situated in the centre of Bergen, which has expanded and spread around the water body during the past 230 years. The evolution of the water body displays a complex history. It was initially part of the fjord after the last Ice Age. As a result of isostatic uplift the surroundings emerged into islands that today form Nygårdshøyden and Sydnes. The area went through a period of marine transgression that culminated at approximately 7000 years BP (Figure 3).

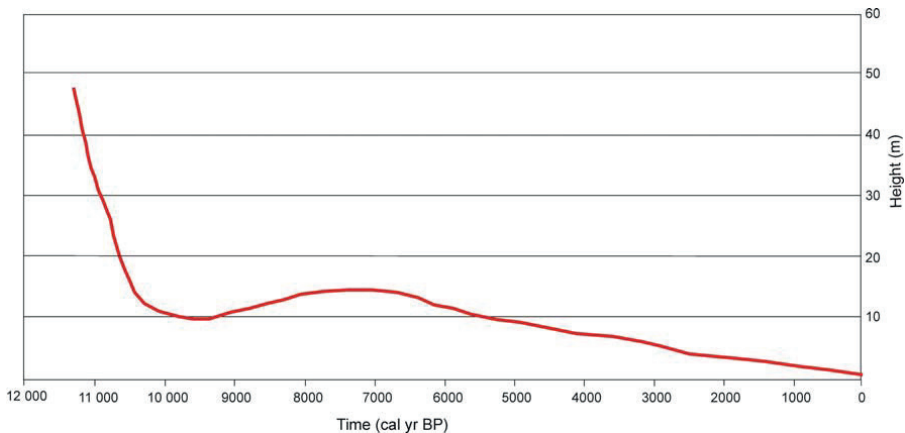


Figure 3. Calculated sea level curve for centre of Bergen (Øystein Lohne, pers. comm., 2012). The curve has been calculated on basis of sea level data from Os (Lohne et al., 2007) and Hardanger (Romundset et al., 2010).

The gradual separation from the sea through increasingly narrow connections may possibly have caused some sediment erosion by tidal currents. The separation from sea water resulted in the transformation of the water within LL from saline to brackish. The final physical separation from the sea was anthropogenic, as the narrow strait between LL and Store Lungegårdsvannet was closed in the 1920's. The final separation of LL is a well-documented part of the modern urban history of Bergen

(Harris, 1991). However, the isolation of the water body has never become fully complete as saline water still resides on the bottom of the lake. Taking these conditions into account, LL may be considered a young urban lake, as the final separation from the sea and establishment of a lake environment only became reality in the 20th century.

The environmental conditions within the water body, such as water chemistry and biological activity, have changed over time due to these physical and chemical alterations. Scanning Electron Microscope (SEM) imagery reveals abundant diatoms in the upper parts of the core, while sediments representing pre-industrial times, on the other hand, consist mainly of eroded mineral particles. We have no knowledge of how deep the freshwater front has been, before the 20th century, or how the redox boundary has moved within the water mass.

Complaints to the city regarding the odour from the lake have been recorded as early as the 18th century. The lake has been the subject of some attempts to improve the condition of the water. These include partial dredging and the filling of sand and gravel on top of the surface sediments. This was undertaken at the end of the 19th century in an attempt to either remove or cover the surface sediments (Figure 4). The issue of dredging possibly creating several vacuities are discussed further in the dating constraints chapter.

It was customary to position certain industry and craft, such as tanneries and butchers, outside the habited area to reduce noise and smell. Other crafts, such as smiths and bakers, were also placed there due to fire hazard (Kjellberg and Stigum, 1936). The tanneries were moved from Vågsbunnen to LL in 1625, along with the butchers, in order to lessen the polluting impact from the trade (Kjellberg and Stigum, 1936). The tanneries now formed a front of buildings along the shore, thereby giving access to boats (Fossen, 1985). Therefore, LL was a receiver of craft waste before the shore actually became inhabited. In addition, the lake has been extensively used as a waste disposal site, at least from the 17th century, in terms of household waste, sewage and waste from urban fires. During the start of the 18th century, a committee

was founded to find a solution to the waste problem. The solution did not involve a prohibition of waste dumping but a restriction of which side of the lake was to be used (Fossen, 1985).

The start of the 19th century was marked by a period of large expansion of the craft industry in Bergen that moved towards industrialisation in the 1840's (Ertresvaag, 1982). The main industries in Bergen during the 19th century were metal and machine, textile, food and clothing industries. However, as the historical sources chapter points out, the potential pattern of elements and compounds contained in the discharge from these industries is very similar (Paper III). It is therefore challenging to distinguish between inputs from different sources. The two first decades of the 20th century were characterised by the most rapid increase in industries, introducing many canning factories (Ertresvaag, 1982). However, as Fossen and Grønlie (1985) describe it: *"It is not the industry that formed the city; it is the city that formed the industry. Bergen did not have more industry than was needed."* Due to lack of space in the central parts of Bergen, the centre was never characterised by large industries. The industrial companies were many and varied (Paper III), but minor in size. During the 1920's, several economical setbacks slowed down the industrial growth for several decades (Fossen and Grønlie, 1985). The large number of industries as well as the gradual growth and development of the city provides many challenges when it comes to source control.

Understanding the source of pollutants within an urban lake is also made difficult, as so many polluting pathways have existed over time. The pollutants mix together in sewage effluents, in waste that is manually dumped into the lake, and in urban runoff. As the urban environment is covered with impervious areas such as roofs, stone and asphalt roads and concrete, only a small amount of the urban runoff may infiltrate, evaporate or be detained by vegetation (Mays, 2001) and particles within the runoff are flushed into the sea and lake.

1.5 Historical sources

Bergen has remained an important port that for centuries has kept a lively trade with not only other parts of Norway, but also with the rest of Europe, especially Britain and Germany (Helle, 1982; Fossen, 1979). A large number of foreign craftsmen were active in Bergen. A lively international trade meant that new products, crafts and industrial processes quickly spread to Bergen, often earlier than in other parts of the country. The following chapter points out potential, historical sources in Bergen for each element discussed in the papers. Figure 4 then includes many of these point and diffuse sources in a time frame.

As- Arsenic

Arsenic is a metalloid, which shares properties of both metals and non-metals. Arsenic has been used within medicine for 2000 years and in synthesised drugs since the 19th century (Gibaud and Jaouen, 2010). From the 19th century it was used within pigments and insecticides (Hughes et al., 2011). Arsenic pigments have been phased out, but arsenic is still in use in glass and as a means of strengthening lead and copper alloys and semi-conductors (ATSDR, 2014). Arsenic is also dispersed through coal combustion. Arsenic has extensively been used within wood preservatives, especially chromated copper arsenate (CCA) that has been used extensively in Bergen.

Cd-Cadmium

Cadmium is a toxic metal that is naturally associated with zinc and is a component in zinc and lead ores. One of the earliest known uses of cadmium was in pigments that became popular during the mid 19th century. Leather workers also used it in the tanning process. After the industrial-scale production started in 1930-40s, cadmium was used, through the electroplating process, to increase the corrosion resistance for other metals. Cadmium is also spread into the environment through coal combustion, the usage of mineral fertilisers (ATSDR, 2014) and tyre wear (Reimann and Caritat, 1998).

Cr-Chromium

During the 19th century, chromium was mainly used as a component in pigments and tanning salts. Therefore it is expected that the tanneries around LL emitted chromium into the water as the tanning process developed from using bark or alum (Grieg, 1936) to chromium salts. Chromium was also used from the 19th century within the textile industry to fix and stabilise dyes to the fibres. In later times, chromium has been used within metal alloys such as steel, as plating of metals and within wood preservatives. Chromium is also spread in the urban environment through coal and oil combustion (Reimann and Caritat, 1998).

Cu-Copper

Copper was one of the first metals to become used, due to its natural ore occurrence in a form that made it easily extractible. It has been of varied medicinal use for 4000 years. Due to its softness and malleability, copper has been widely used within construction as piping and roofing material. Copper was from the Middle Ages commonly used when producing engraving plates. Goldsmiths, which were an active group in Bergen from the 13th century, also used copper and brass to produce items such as weapons and pots (Grieg, 1936). In the 17th century, approximately 70 documented goldsmiths were active in Bergen (Kjellberg and Stigum, 1936). Copper is extensively used in various metal alloys (mainly bronze and brass), pigments, insecticides, algaecides and fungicides (Reimann and Caritat, 1998), but is also spread through the burning of fossil fuel (Mahler et al., 2006).

Fe-Iron

Iron is the main constituent of steel and is used within many pigments (Reimann and Caritat, 1998) and is present within many everyday utensils. In the middle Ages iron was refined by the smelting of bog ore. The first smiths in Bergen, which are mentioned in the 12th century, resided "up under the mountain" (Grieg, 1936). The amount of documented smiths are, however, lower than the size of town would suggest (20 smiths in the 18th century). This is according to Kjellberg and Stigum

(1936) probably due to an extensive import of metal utensils. Prahls støperi, situated in the drainage area of LL, was the first iron foundry in Bergen, which was founded in the 1840s. In addition, a number of metal workshops, smithies and scrap yards existed in the area. Iron is also spread into the aquatic environment through sewage and urban runoff.

Hg-Mercury

Mercury has been an important element used to extract gold and silver. In addition it was used within medicine, especially from the 16th century, as a cure for syphilis (Norn et al., 2008). Mercury was emitted in large quantities into Vågen due to local dentistry, as amalgam was used as a tooth filling material in Norway from the early 20th century. Mercury has been used within wood impregnation and paint in the form of pigments and fungicide (Reimann and Caritat, 1998). The use of mercury within the manufacturing of felt for hats was established during the 19th century, but later abandoned in the 20th century due to its toxicity. Bergen had during the 17th century three documented active hatters (Kjellberg and Stigum, 1936). The metal has been used in thermometers, pressure measuring instruments, switches and relays. Mercury is another metal that is spread into the air and water through coal combustion and sewage (Li et al., 2013).

Ni-Nickel

Nickel ores were mistaken for silver ores, so the historical use of nickel has not been extensively documented. However, the unintentional use of nickel in alloys can be traced back 3000 years. After its discovery in mid 18th century, it has been extensively used in various alloys, therefore any metal industry or scrap yard will have emitted nickel into the surroundings. The metal is used for plating metal objects and has been a metal component in coins. Nickel is emitted into the air via incineration, traffic and the burning of fossil fuel, and into water through sewage (Reimann and Caritat, 1998).

Pb-Lead

Atmospheric deposition of large-scale lead pollution has occurred for at least 3000 years in Europe, which had its source in metal production and smelting (Renberg et al., 2001). Since the 18th century the ductile characteristics of lead made it popular in use within building materials such as roofing, windows and paint (Drange et al., 2011). Lead-glazed ceramics for use within piping, roofing tiles and kitchen appliances (Wallace et al, 1985) became popular during the 19th century, even though unglazed varieties were still more extensively used due to a lower price. In later times, the anthropogenic sources include coal combustion, gasoline additives, and waste water discharges (Ritson et al., 1994). Additional point sources within the discharge area of LL that potentially discharge pollution with lead include metal shops, production of plastic, paint, glass, lime, graphical products and leather (Paper III).

Zn-Zinc

From the 19th century the galvanisation of steel using zinc became popular, due to its anti-corrosive properties. In addition zinc is used in many alloys, such as brass, and therefore emitted in many metal processing crafts and activities. Zinc is used in the production of rubber, glass, plastics and wood preservatives. Zinc is present in rubber tyres and is emitted through traffic (tyre wear) (Reimann and Caritat, 1998). White house paint has been very popular in Bergen, and paint containing white zinc pigments became very popular during the 19th century as zinc paint was more durable and cheap than lead paint (Drange et al., 2011).

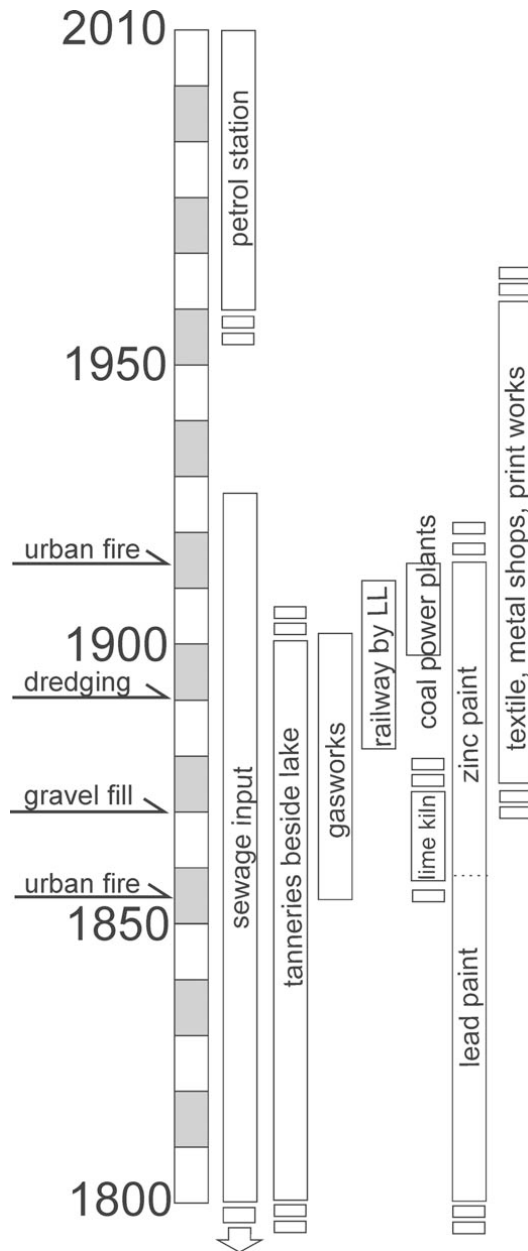


Figure 4. Timeline presenting pollution sources that were located around Lille Lungegårdsvannet during the time period reflected in the upper part of the investigated sediments.

2. Methodology

The thickness of the sediments in LL was known to be over 5 metres, particularly in the centre of the lake. What lies underneath the sediments has not been specified in any early cross-sectional maps, or later investigations in any other way than as a firm base (Hanssen, 2009). The thickness reduces toward the edges of the lake, where the sediments appear coarser (Hanssen, 2009). Thus, finer grained sediments accumulated in the centre of the lake as coarser grained material settled closer to the edges of the lake, where the sediment and water influx is situated. The sediment coring for the long core was performed in the centre of the lake using a piston corer (Nesje, 1992). Several other cores were taken during the same coring session, but the other cores were shorter than and not located as central as the analysed core. Whether the analytical results thus give rise to a concentration range that is higher, due to low grain sizes than for the lake in general has not been investigated within this study. Whether the top sediments contained in the piston corer actually were the surface of the sediments could not be verified. Therefore the long core was supplemented with undisturbed surface sediments with a Hongve type gravity corer (Boyle, 1995) at a later date. The relative displacement between the top sediments in each core was determined through the use of element concentrations of the following elements: Pb, Zn, Hg, Mn, Fe, Cu, Ni, Zr, S, P and TOC. A visual analysis used a comparison of concentration graphs at different intervals to establish the best concentration match between the long core and the shorter surface sediment core. The visual comparison study was complemented by the usage of correlation coefficients to detect the displacement (Paper III). Therefore results from both cores could be merged into a single core.

In order to quantify the amount of urban runoff from the drainage area and suspended sediment concentration within the runoff, the stormwater study was conducted (Paper I). The results, in conjunction with the analysis of the stormwater trap sediments, provide an indication of the type of material that has been eroded from building surfaces and other impervious surfaces. The sampling station for the discharge study

was located such that all urban runoff that ends up in the culvert was known to originate from a small drainage area. The study was performed in an area where no grit is spread during the winter months and the ice is removed chemically from the street surfaces by the use of magnesium salt. The monitoring and sampling study was conducted over a whole year in order to observe long term trends instead of only concentrating on storm events. The Bergen climate, which maintains a high precipitation rate throughout the year (Figure 9), is suitable for such a long-range study. The sampling of the runoff was collected using an Isco water sampler. However, the amount of water collected did not provide enough suspended material to run chemical analysis in addition to the concentration of organic matter.

The material that is not transported onwards as suspended particles, settles in the stormwater culverts. The stormwater culvert sediments have previously been studied in other parts of Bergen, so their capacity to indicate pollutant patterns was known (Jartun et al., 2008). These sediments were therefore collected to provide an indication of active pollution sources.

Previous studies in Bergen have played a part in shaping the analytical program of this study. These showed that the urban soil contains high levels of PAH (Haugland et al., 2008; Jensen et al., 2011) and PCB (Andersson et al., 2004; Jartun et al., 2008), demonstrating the importance of including these compounds in the present study. PCB has also proved to be the compound that is the most bio-available of the studied compounds and metals in sediments in Bergen (Øxnevad and Ruus, 2012). In terms of PCB, the above studies demonstrate that we still find active PCB sources in the urban environment, even though the compound was banned in Norway in 1980. However, a critical issue with PCB studies of standing building mass is that it is only possible to analyse the paint, plaster and sealants that occur on the building today. Many buildings are refurbished and are superficially "clean" and do not reflect their past polluting status. Andersson et al. (2004) demonstrated that PCB was imported and used approximately a decade earlier than anticipated by the pollution authorities. It is therefore of importance to verify this early usage of PCB in building materials

through the analysis of urban sediments as they are the "end station" of material from eroded surfaces.

3. Results and discussion

The chemical results from this study demonstrate that human activities leave permanent chemical markers in the environment. The chemical changes within the urban environment are related to industrial and other human activities as well as historical events that caused increased concentrations of metals and organic pollutants.

Throughout time, inorganic elements have been eroded from the local soil and bedrock and have thus set their fingerprint on the marine sediments and created the specific natural sediment chemistry of LL (Paper III). Streams that had their source in the surrounding hills transported eroded material into the inlet. Agricultural activity in the area, present since the Bronze Age (Hjelle, 2000), probably added to the erosion of soil. This natural sediment source became limited as habitation expanded and spread into the drainage area from the 17th century (Figure 2) and later around the shores of the inlet. Crafts that produced large amounts of refuse or produce an unpleasant smell, for example tanneries, were situated on the outside of the habited area. It may therefore be expected that this kind of craft entered the drainage area somewhat earlier. The streams were cut off and diverted due to building activity and the sediment inflow into the water body became more characterised by surface flow from impervious surfaces and sewage input. As the city of Bergen expanded in size, the sources have changed in size, character and discharge type over time. The chronology interpretation will be presented first before discussing the chemistry of the core.

3.1 Dating constraints and methods used

The sediment core was divided into two units. The interface between the two units, which is situated between 183 and 185 cm depth is characterised by a change in sediment colour and consistency. Unit 1, which is the lowermost part represents a

pre-industrial sedimentation environment, while Unit 2 represents the anthropogenically influenced part of the core.

The entire core is characterised by low amounts of terrestrial macrofossils. Therefore a large number of samples did not contain enough material to be able to be analysed for dating purposes. However, terrestrial macrofossils were considered to be the sample media within these sediments to give the most accurate results. Calibration was performed using OxCal with a 2-sigma uncertainty.

Samples from pre-industrial sediments (Unit 1) provide a consistent age progression, while results for samples taken from Unit 2 display a seemingly disordered picture where many of the results present a much higher age than the location in the sediment column would suggest (Paper II). Results from the accelerator mass spectroscopy (AMS) ^{14}C dating and historical data were entered into an OxCal model, where a pattern emerged from the periodically chaotic picture of Unit 2 (Figure 5). Reworking and deposition of terrestrial macrofossils and bioturbation may be a cause of results not matching the sediment sequence (Björck and Wohlfarth, 2001). In LL, however, bioturbation has not been considered a potential problem as the hypoxic environment has limited the bottom fauna. In addition, if the sediments had been vertically mixed the chemical concentrations would exhibit a more 'noisy' pattern. The reworking and deposition of old macrofossils is a process that would be expected in an urban environment where the lake has been used as a waste disposal location. The results for the samples within Unit 1 may also represent older, reworked material due to agricultural activity, which may have given rise to erosion. This erosion could have mobilised older terrestrial macrofossils (Krzywinski and Kaland, 1984) that subsequently deposited in LL. This might lead to the dating results within Unit 1 appearing older than their depth might suggest. This is, however, considered unlikely since all the age determinations within Unit 1 are located on a straight line within the age-depth relationship (Figure 5).

Apparent ^{14}C dating discrepancies may arise from the fact that the sediments contained very little terrestrial plant material throughout the whole core so that the

sample sizes were small. It is, as yet, unclear whether the sample sizes, the influence of reworked material or contaminated samples have contributed to the apparent older results. The ^{13}C results indicate that the material used is terrestrial (Paper II), therefore precluding the potential influence of aquatic species affecting the results.

As the AMS dating in Unit 2 provided inconsistent results, additional sources were used with the aim to provide a more consistent chronological record. Changes in concentration for many of the analysed elements provided indications of certain events or time periods that help constrain the age-depth relationship. These were subsequently inserted into the age-depth model. The use of chemical results to define age-horizons is not without risk as it stands as an example of circular arguments, where results contribute to the age definition. However, the changes in chemistry, which indicate specific historical events, are considered well defined and unlikely to have been attributed to other factors. Identification of the exact level of the historic events may be discussed, but the chosen horizons are defined to be of a high enough confidence level to be added to the age-depth interpretation. Below are the events and levels that were inserted into the age-depth relationship in addition to the AMS dating results (Figures 5 and 6). Figures 5 and 6 therefore consist of a mixture of ^{14}C dating results as well as horizons of other proxies for Unit 2 that were considered reliable enough to be included.

Depth (bss) Defined year

0 cm	2011	The year that the top core was taken.
15 cm	1960	The highest concentration of Cs in the core (Paper I), expected origin due to nuclear weapons testing.
18 cm	1940	from ^{137}Cs and ^{210}Pb age-depth relationship (Paper I)
75 cm	1930	The concentration of P decreases significantly. In 1928, Bergen City rerouted all sewage that previously had been emitted into LL to the outer fjord (Paper III).

110 cm	1870	Gravel accumulation at 110 cm depth (Paper II), the only of its kind in whole sediment sequence. Interpreted to coincide with 1870s, as gravel was placed onto surface sediments to reduce smell (U 110 cm in Figure 6).
120-125 cm	1860-1870	At this level a wide Cr-peak occurs. Many tanneries existed around LL. Using Cr to treat the hides was a process that was used from 1860-70. Before that milled bark had been used (Paper III).
130 cm	1850s	At this depth a number of coal-related elements increase in concentration more or less significantly; Pb, Ni, Hg, As and not least, PAH (Paper II). Interpreted to coincide with onset of gasworks in the 1850s (U 130 cm in Figure 6).

The horizons at 110 and 120-125 cm bss are considered to define historical events with a larger degree of uncertainty than the other horizons. The accumulation of gravel may have sunk into the surface sediments during deposition. Precise information on the onset year of the change in tanning processes from bark to chromium has not been verified. Therefore some caution must be used when using these specific horizons.

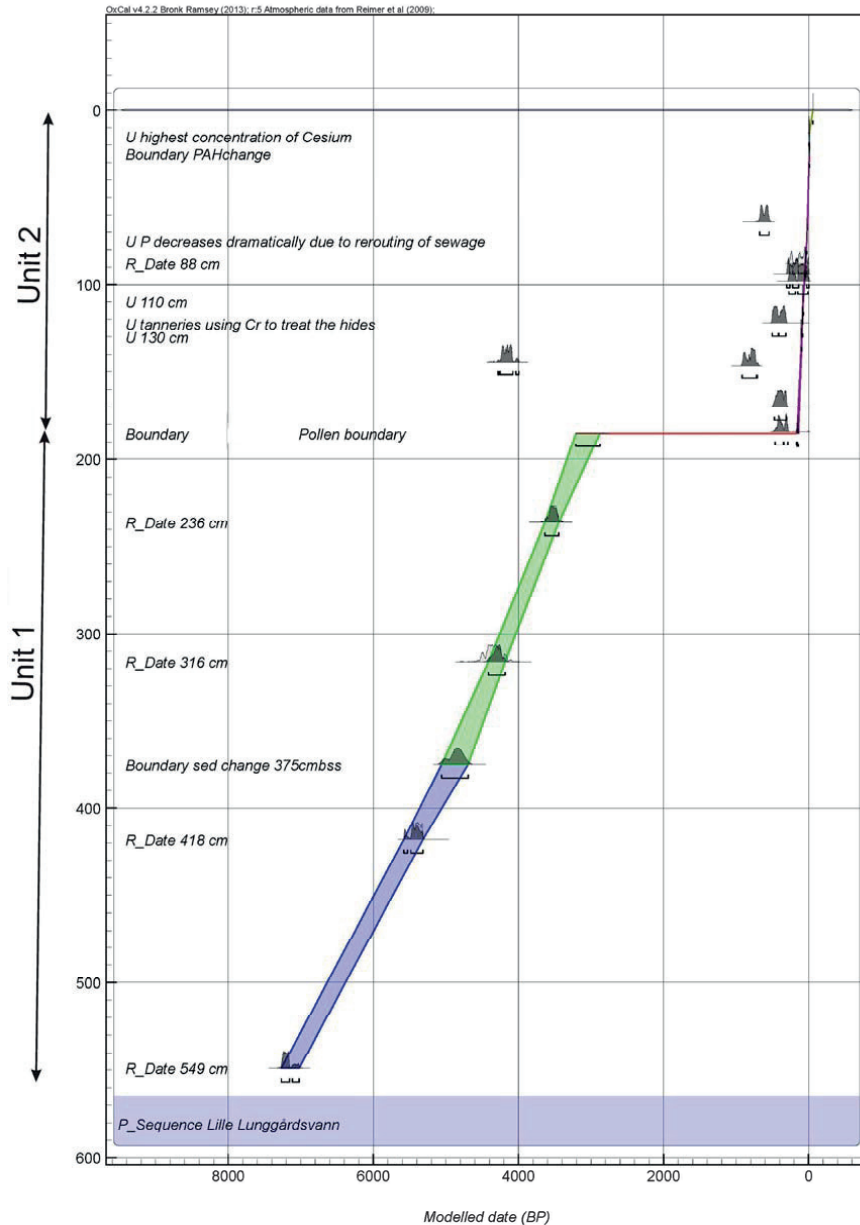


Figure 5. Age-depth relationship for the entire core based on AMS-dates and historical events identified in the chemical elements.

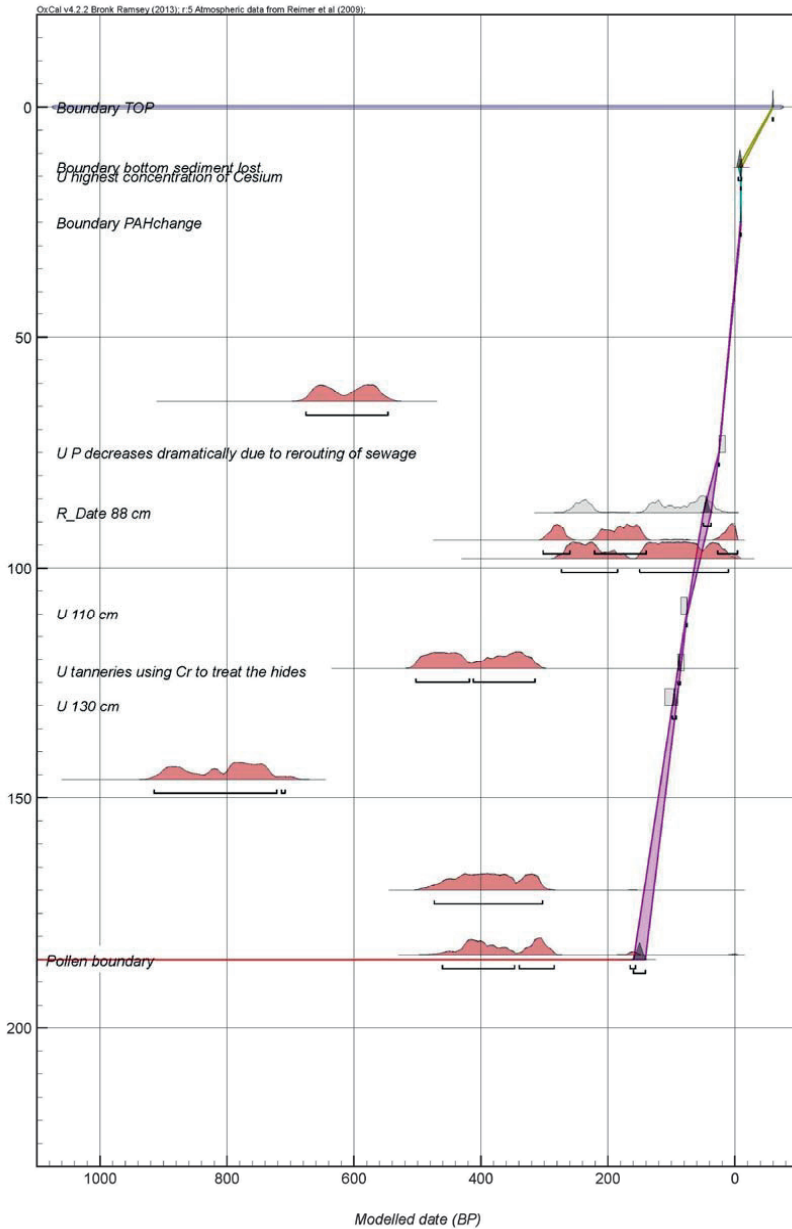


Figure 6. Age-depth relationship for Unit 2, based on the same AMS-dates and historical events as in Figure 5.

A series of closely-spaced ^{14}C dates joined through wiggle matching could also have provided an independent chronology (Blaauw et al., 2004).

Using the age-depth relationship as well as the sedimentation rate at the time and changes in chemistry for many elements during the 19th century, the bottom of Unit 2 has been interpreted to approximately represent the 1790's.

Between Units 1 and 2 a vacuity is most likely present, which represents erosion or some other sediment removal process. As discussed under the study site chapter, erosion of surface sediments as well as dredging may be possible causes for a vacuity/hiatus. Bergen City commenced dredging on a larger scale in the 1750's when dredging barges were bought, dredging equipment was built and a barge master was appointed (Fossen, 1985). Dredging in LL during the 18th century is difficult to confirm, however, as written documentation is minor and scattered. Dredging had been carried out already from the 16th century in Vågen, using large rakes from the shore and later barges, to provide an increased water depth in the harbour. Partial dredging in LL or in the narrow sound between Lille and Store Lungegårdsvann has been documented in 1825 and within LL almost annually from 1857 onwards (Bergen byarkiv, 2014), but the extent and precise location is unknown. The frequent dredging activity between 1857 and 1899 was performed to improve the sanitary conditions and coincides with the construction of quay structures around LL (Fossen, 1985).

Several factors suggest a vacuity at a depth of 183-185 cm bss; a change in the physical sediment column, a significant change in chemistry for several elements as well as for pollen. Firstly, the sediment material changed colour and consistency at the Unit 1-Unit 2 interface. Secondly, the chemistry demonstrates a significant change at this interface (Paper III). This considerable increase is only apparent with the metals that display an anthropogenic pattern, such as Pb, Zn, Cu, Hg, Fe (Figure 7) as well as P and TOC (Paper II). The substantial concentration changes at the

interface between Unit 1 and 2 are somewhat remarkable, taking into account the number and sizes of known industries at the time.

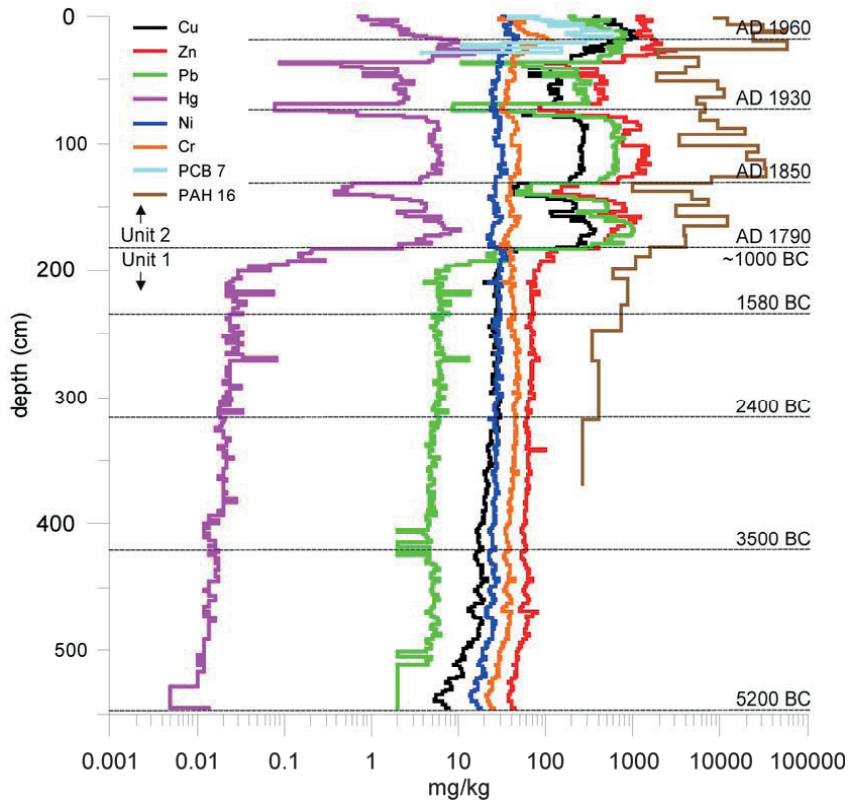


Figure 7. Summary of selected elements, compounds and chronology. Note logarithmic scale on the x-axis.

In theory, this significant increase could be due to a sudden increase in the use of metals and subsequent pollution, indicating unknown metal sources. However, this hypothesis is determined as more unlikely than missing sediments. The elements that display a geogenic pattern do not exhibit the same apparent increase, as the move into an anthropogenic era did not increase their input. Lastly, one more factor that suggests a vacuity is pollen analysis that indicates natural deposition up to 186 cm

and a major change in the relative species content from 184 cm, indicating an environmental change (Hjelle pers. com.). Therefore, the seemingly low sedimentation rate is most probably due to some sediment removal process.

Figure 5 differs from the age-depth relationship figure published in Paper II as the vacuity concept and the depth of the border between Unit 1 and 2 has become better defined during the course of the study. If the curve connecting the age determinations of Unit 1 is extrapolated up to the unit interface, it can be proposed that the sediments deposited between approximately 3000 years BP and the bottom of Unit 2 have been removed from the sediment column. The thickness of the missing sediment column may theoretically, by using Figure 5, be calculated to approximately 2 metres. However, any precise measurement is impossible to calculate as the sedimentation rate in the Medieval period cannot be verified/tested.

The surface sediment core was analysed for ^{137}Cs and ^{210}Pb at the University of Copenhagen and an age-depth relationship was constructed for the top 18 cm (Figure 6). Due to financial restrictions, the core was unfortunately not analysed further down to provide a more thorough analysis. Surprisingly, the ^{137}Cs results demonstrate that the Chernobyl disaster in 1986 cannot be detected in the sediments (Paper I). Studies conducted after the disaster provide evidence that the radioactive fallout in Norway was very dependent on precipitation patterns at that time (Lindahl and Håbrekke, 1986). Even after being transported 1800 km from the source, the radioactive fallout could vary by a factor of five over a distance of a few kilometres in central Norway. Therefore, depending on the precipitation pattern, it is possible that Bergen City escaped some of that fallout, thereby explaining the lack of increased Cs-concentrations in LL.

During the chronological interpretation of Unit 2, the possibility of sediment disruption from dredging activity also became an issue. According to written historical documents, summarised by Byrkjeland and Hammerborg (2005), surface sediment dredging was performed in 1890, removing a slice (30 cm) of the surface

sediments in an attempt to improve the water quality. Whether the entire central part of the lake where the core was taken was dredged has not been documented. Therefore there is a possibility of a vacuity also at this level of the sediment stack, but the location of this vacuity in the core is not possible to unravel from the chemical data. The presence of an additional vacuity may be suggested by looking at the general sedimentation rate in Unit 2. LL is characterised during this time by a generally high sedimentation rate (approximately 1 cm/year). The horizons that were suggested by other proxies to represent approximately 1930 and 1870, were inserted into the age-depth relationship model and provide a depth interval. If the sedimentation rate for this interval is to approximately follow the general rate within Unit 2, then the sediment height between these horizons is too small for such a high sedimentation rate. This potentially indicates that part of the sediment record is missing or that the proposed dates are inaccurate. The sedimentation rate cannot be expected to have remained constant, but the change would in that case be too large to rule out a vacuity as a result of dredging.

3.2 Sediment chemistry and pollution sources

From the mid 19th century to the present day the geographical locations of pollution sources in the drainage area of LL have been reasonably well documented. The potential pollution from these sources may be compared to the chemistry of the sediment column. Heavy metals, nutrient and sediment grain size all demonstrate horizons that concur with the historical changes within the central part of Bergen (Paper III). Unfortunately, the geographical knowledge of historical pollution sources before this time period is limited.

Metals

In general, the highest concentration peaks for the different elements occur during similar sediment depths and therefore appear to be synchronous. This is a pattern shared by all compounds and elements discussed here. At a sediment interval from

185-180 cm below the sediment surface (bss) the concentration pattern changes significantly. The sudden concentration increase of elements such as Pb, Zn, Cu, Hg, Fe, Ti, Mn, Co, Ba and Li (Figure 7) could be the outcome of sewage and craft effluent emission into the inlet at the end of the 18th century. The concentration remains high with the exception of some concentration fluctuations that are discussed below.

Scanning electron microscope (SEM) analyses of the sediment samples that contain high Pb concentration (Paper III) show that these peaks most probably are due to pollution. Within these samples, Pb is present as fine particles that consist almost solely of lead and are not tied to other elements or occurring within a mineral lattice. Pb has a high atomic number making it easy to detect on the SEM. The other heavy metals have lower atomic weight and have a lower contrast than Pb on the SEM. The possibility exists that these micron-size particles originate from eroded lead paint, as lead based paint was common in Bergen at the time.

From 130 cm bss upwards, the concentration curves for As, Pb, Hg, Zn, Cu, Mn and Co again demonstrate a large increase. This peak most likely coincides with the active period of the gasworks, which was emitting effluents and waste directly into the lake. Other coal-utilising industry was also in production by the turn of the 19th century. Raask (1985) and Lima et al. (2005) quantified the element content of coal, demonstrating that coal is a major source of heavy metals.

Between the 1920's and the late 1940's, industry suffered a substantial decline in Bergen, which is suggested to denote lower levels of pollution during that time period. This seems to be reflected as relatively lower concentrations in the sediments. In the decades after WWII a local boost in industrial activity occurred, which is reflected in the most prominent period of increased concentration for most elements. In addition to the industrial activities, the urban environment changed with an altered traffic situation in the 1960's. The Norwegian government introduced a rationing of car purchase that was repealed in 1960. This led to an enormous increase of the car

pool in the 1960's and the subsequent introduction of a new Pb source that had not been present before.

PAH

From approximately 180 cm bss an increase in PAH concentration is also observed (Figure 7/Paper II). This coincides with changes in the PAH compounds/sources. The relative occurrence of the 16 PAH compounds vary gradually over the whole core, but also more significantly at specific intervals, for example at approximately 180 and 25 cm bss. The earliest PAH sources were characterised by low-temperature processes in the bottom of the core that are replaced by high-temperature processes towards the present day.

The gasworks appears to stand out as a major PAH point source. Urban fires also contributed towards increased PAH concentrations in the sediments. Urban fires most probably contributed in a more extensive degree towards PAH pollution in Bergen than in any other city as it has been documented that fire refuse was disposed of in the water body (Asplan Viak, 2003). Six urban fires have been documented in the vicinity of the lake (Paper II). In addition, domestic heating is a known PAH source that added to the pollution throughout the centuries. Domestic heating stands as one of the prominent, current PAH sources in the urban environment (www.ssb.no).

PCB

PCB was first detected in sediments deposited in the 1930's (Paper I). PCB is a compound that was deliberately added to the environment through its use within the construction industry, polluting the environment through the subsequent decay and rehabilitation of these constructions. The detection of PCB in sediments deposited in early 20th century highlights the possibility of sample contamination. It is, as yet, unknown whether low PCB concentrations within sediments deposited in the early days of PCB production reflect a real input or are an indication of sample contamination? Designing a proper quality control program is essential, one that includes duplicates, reference material and blank samples (Boyle, 2002). The use of

blank samples proved to be crucial when investigating the contamination of packaging, as was presented in Paper IV. The use of sampling bottles contaminating samples with metals has been a longstanding issue within the field of geochemistry, but no attention has previously been directed towards paper and plastic packaging in relation to their content of PCB.

3.3 Discussion of the fluctuations in metal concentration

Boyle (2002) suggested that sediment grain size, as well as TOC (Thompson and Eglinton, 1978), can influence the content of metals in soil. This study shows that the same applies for urban sediments in Bergen, where not only the input of metals determine their concentration, but also the sediments ability to absorb the metals is important. The influence of sediment grain size and clay amount on element concentration applies not only to the metals discussed in Paper III but also for other elements such as Al, Fe, Ti, Mg, Ca, K, Mn, P, Co, Ba, Sr, Li, Sc, S and C. Elements that do not display an increasing concentration pattern with decreasing grain size, nor an increasing amount of clay are: Cd, Na, V, Mo, Zr, B, Be, Ce, La, Y. It must be noted that this applies for urban sediments, where a large part of the polluting elements are accessible and not tied up in crystal lattices.

The sharp increase in metal concentration for many of the elements at the interface between Unit 1 and Unit 2 can partly be explained by sediment grain size. The interface is characterised by a slight change in the silt-size fraction (Figure 8) that can be expected as the two units represent different times of deposition. The grain-size characteristics alone cannot explain significant concentration changes, therefore the increase in metal input from various sources is most likely present. It must be noted that only grains less than 1 mm are included in the grain size analysis. Within Unit 2, two horizons display an increase in coarser sediments, between 140-130 cm bss and 75-30 cm bss upwards (Figure 8). These horizons coincide with decreased metal concentrations, interpreted to be a combination of increased sediment grain size and increased erosion and cessation of sewage input respectively.

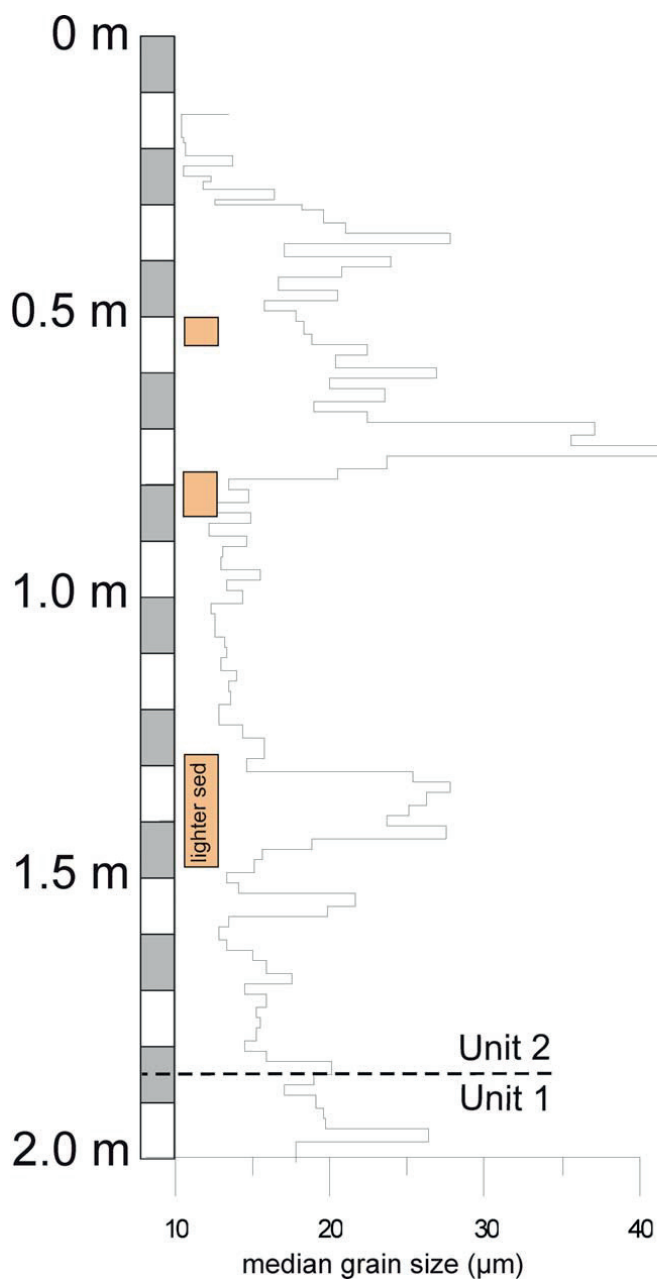


Figure 8. Sediment (<1 mm) grain-size characteristics within Unit 2 and top of Unit 1. The sediment sequences indicated by brown boxes consisted of sediments of a lighter colour.

Coulter analysis shows that the sediments display a remarkably low amount of clay-sized particles (2-8 %, median 5%) (Paper III). According to Rise and Brendryen (2013), the amount of clay-sized particles within marine and glacial marine sediments is underestimated when analysing the grain sizes using Coulter. This is due to the fact that, within the analyses, it is taken for granted that the particles are sphere-shaped. This is not necessarily the case, especially in fine-grained materials.

Rise and Brendryen (2013) suggested a formula to calculate the "true" clay contents $(4x\% < 2 \mu\text{m} + \% < 7.4 \mu\text{m}) / 2$ by using the results from Coulter laser. This would produce results (12-35%, median 21%) for clay-sized particles that differ significantly from the analytical data received from the laboratory. The increased amount of clay-sized particles could explain the large dependency of clay amount in the samples with respect to metal concentration.

3.4 Stormwater in Bergen

Stormwater studies have become more prominent as the potential polluting influence of stormwater has been established. It has been suggested that stormwater may contribute to almost half of the PCB-load in the marine environment (Gibson et al., 2003). In some stormwater studies, calculated results suggest an amount of solids that accompany the stormwater. However, to our knowledge, the present study is the first of its kind where the actual amount of stormwater and suspended sediments has been measured over a longer time period.

For centuries, urban runoff and sewage have flowed into the lake, due to impervious surfaces around LL and drainage systems that direct the runoff into LL. From 1928 onwards, the mixed sewage and urban runoff was directed away from the lake through a collective drainage system. However, even then the low capacity of the old collective drainage system has during extreme precipitation events given rise to periods when the stormwater goes through the overflow vent and straight into the lake. The City of Bergen started to rebuild the collective drainage system from the

1980's into separate systems in order to provide an increased fresh water input into the lake. The exchange of saline into fresh water has been recommended in several studies (Johansen and Skarheim, 1968, Larsson, 1994, Johnsen, 2008, Johnsen and Brekke, 2009) as a way of improving the water quality.

Bergen is characterised by a relatively large amount of precipitation. The 1961-90 annual mean in Bergen is 2250 mm, which is spread over 269 days of precipitation (Figure 9). This stands in contrast to the rest of Norway, which on average receives 760 mm/year spread over 214 days (www.met.no). Storm events put an enormous pressure on the drainage system, which in large parts of central Bergen is outdated and under-dimensioned.

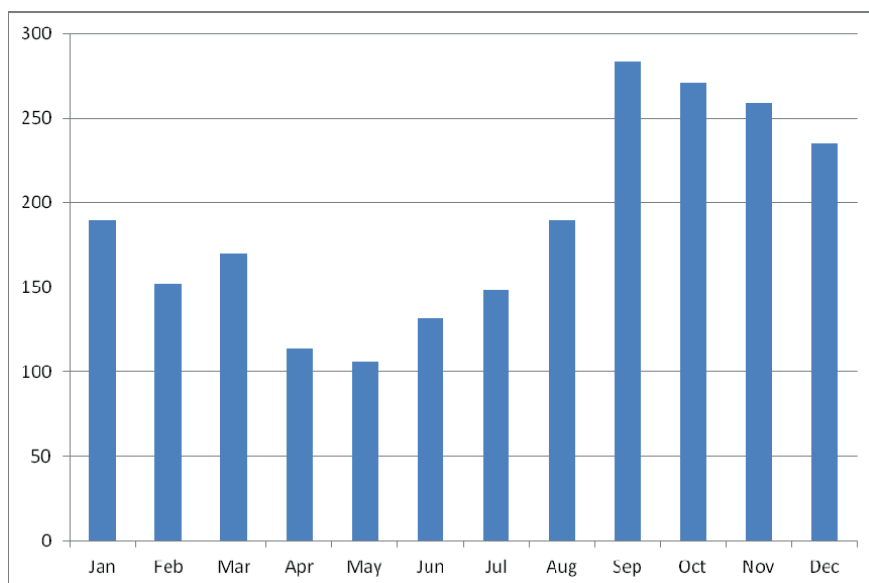


Figure 9. The 1961-90 mean monthly precipitation for Bergen. Data: met.no.

Stormwater culverts are designed to slow down the stormwater and settle the particles within the water. The amount of particles that settle in the culvert is dependent on the amount of water and how turbulent the water is, but also on particle size, specific gravity of particles, temperature and the time until next storm event (Butler et al., 1995). It has been estimated that the overflow from impervious surfaces is discharged directly into a water recipient approximately 30% of the time (Jartun et al., 2008), carrying an unknown amount of suspended material with it. The amount of urban runoff as well as the amount of suspended sediments within the runoff has now been measured within this study, to more accurately calculate for future drainage systems.

The stormwater study shows that there is an extensive, even flow of stormwater that derives from the urban catchments, 37 650 m³ for the 5 800 m² catchment (Paper I). The stormwater culvert where the measuring equipment was situated receives urban runoff from a drainage area that is 100% impervious. Therefore all flow within the culvert originates from precipitation as no other water source contributes to the runoff. The fact that there is no other water source means that the culvert system at intervals remains dry and that short intervals of precipitation will not be intercepted as measuring and sampling occurs once a day. The actual stormwater quantity is found to be three times the modelled quantity (Bingman, 1983). Therefore caution must be taken when using modelled numbers when calculating flow and sediment transport as it may be underestimated. Samples taken during periods of low drainage contain very low concentrations of suspended sediments. Generally, a low correlation exists between water discharge and sediment concentration, which is probably related to the diverse character of the sediment sources within this catchment area. Approximately 780 kg of suspended particles accompany the overflowing stormwater into the lake from the small catchment area. An approximate calculation establishes the total sediment flow for the whole current drainage area of LL (Paper I) to approximately 9,500 kg each year. Samples taken from stormwater traps suggest that the elements and compounds investigated in this study utilise urban runoff as a pathway from the urban environment to the lake sediments.

3.5 Potential and limitations of this study and potential future work

This study has shown the advantages of conducting an interdisciplinary study of urban deposits. However, some aspects of the study could have been extended or improved to provide a much fuller picture.

The number of samples dated by ^{14}C could have been extended to provide a more thorough chronology, independent from other proxies. The low amount of terrestrial macrofossils, however, put some limitations to the extraction of samples.

A more extensive analytical program would provide more information to more fully unravel the chemical history. The PCB analytical program offered by the laboratory used has been a limitation of this study, where only seven congeners were analysed. A wider analysis would have provided a fuller picture and more useful results where specific sources (such as PCB that is created in some processes outside of the production process) or degradation possibilities within the sediments could have been investigated in more detail. This applies to both paper I and IV.

The lack of nitrogen (N) results is also another analytical limitation. These analyses would have been helpful in distinguishing the origin of the organic matter, whether aquatic or terrestrial that may be determined by the C/N-ratio.

The use of XRD analyses to investigating the mineralogy of the sediments could help to unravel the occurrence of endogenic or autogenic minerals and thereby explain some of the large differences in chemistry. On the other hand, there are no differences within the natural part of the sediments, as the setting and location has remained the same, and therefore it is the anthropogenic part of the sediments that has had the most influence.

If the concentration of metals and organic compounds in the suspended material were to be included, a much more correct assessment could be made on the amounts of metals and organic compounds that come from urban stormwater.

Finally, if a more extensive pollen data analysis was to be included and combined with the geochemistry, a wider picture of the long-term environmental change, as well as more recent human impact could be provided.

4. Final remarks

Urban sediment studies demand the analyses of a higher number of proxies to provide a consistent picture. The influence of urban sediments are locally specific, therefore the results and influences of sources that are apparent in Bergen cannot directly be applied to other cities. The use of LL as a waste receiver and the closeness of sources to the lake make LL unique. Therefore in LL, the influence of sources is easily detected, even though not easily separated. This study has demonstrated a summary of techniques and analyses that may be used to provide a truthful chronology and source analysis.

5. Summary of the papers

The first three papers are concerned with different types of pollution within an urban environment, where the sources sometimes overlap. For example, the gasworks are the main source for PAH as well as metals, and the standing building mass has become a major source of PCB and metals. Paper IV is concerned with sample contamination within environmental studies.

Paper I

Concentrations of polychlorinated biphenyl (PCB) concentrations in Lille Lungegårdsvannet reflect the time period from the start of PCB production in the 1930s to the legal ban of their usage in Norway (1980). The results suggest that PCB usage either started earlier than assumed or that PCB may have been unintentionally produced through de novo synthesis in combustion processes (Kawakami et al., 1993; Boers et al., 1994; Schoonenboom et al., 1995; Kim et al., 2004; Ishikawa et al., 2007). PCB concentrations in the sediment deposited after the ban of PCB usage, as well as detected PCB in stormwater culvert sediments, strongly suggest that there are sources still active in Bergen today. Earlier studies (Jartun et al., 2008; Alston, 2011; Eidem, 2012) and results within this study imply that these sources could be erosion products from standing building mass that utilise stormwater as a pathway from source to recipient. The urban runoff study presents data collected over a whole year. Bergen is characterised by a large amount of precipitation (1961-90 annual mean: 2250 mm - see Figure 9). The large amount of precipitation causes overflow of stormwater that carries suspended particles, 0.013 kg/m³ of suspended sediments are added into the lake through urban stormwater. An impervious, small urban catchment (5800 m²) releases almost 800 kg of suspended solids per year in stormwater. This suggests that the whole drainage area of the lake potentially contributes 9.5 tonnes of mineral particles as well as organic particles via stormwater into the lake each year.

Paper II

Polycyclic aromatic hydrocarbons (PAH) are one of our earliest organic pollutants as they also form naturally and have thus existed before human activity. This study presents results that demonstrate a large concentration variability of PAH during the last 5400 years in Bergen. The PAH_{sum16} results present very low concentrations in pre-industrial times, followed by a general increase. In the past few hundred years several significant concentration peaks represent large increases in input. Gasworks, urban fires, domestic heating, railway and traffic are the main PAH sources encountered. At the same depth as the PAH concentration increased from background levels a change in compounds occurs. In the bottom of the sampled core, low-molecular-weight PAH-compounds, such as naphthalene, display high relative occurrence relative to high-molecular-weight compounds. The high occurrence of low-molecular-weight PAH-compounds reduces considerably, indicating a permanent change in sources from low-temperature towards high-temperature combustion. PAH-ratios indicate that the combustion sources have shifted from wood combustion in pre-industrial times towards mixed and petrogenic sources in the past century.

The ¹⁴C dating provides evidence that the sedimentation rate remained relatively constant for 4500 years (from 7200 to 2700 cal. yr BP). Subsequently, isostatic uplift gradually isolated the water body, leading to a decrease in sedimentation rate as well as potential erosion of the sediment column. Anthropogenic input in addition to a higher biological activity in the lake increased the sedimentation rate considerably as habitation and industrial activities encroached on the lake. The ¹⁴C dating alone, does not provide consistent data during this period, possibly due to the fact that the lake has been used as a waste site as long as habitation has existed by the lake shores. However, by using several organic compounds, metals and cyanide, the method of timing of the sedimentation becomes more robust.

Paper III

This paper presents the results for metal concentration, which displays a large anthropogenic influence. The metal input into LL has been periodically considerable. Pb, Hg, Zn, Cu and Fe show especially large concentration variability since the end of the 18th century. The anthropogenic impact for these particular elements can be seen when compared to reference sediments in Nordåsvannet. These elements are also of significance in the factor analysis as those elements with the highest loading within the anthropogenic factor. Some of the concentration variability can be attributed to changes in grain size characteristics. However, periods of reduced metal input, due to documented economic depression periods, can also be seen in the results.

Sediment grain size correlates negatively with concentration for all elements discussed, with the exception of Cd. The amount of clay fraction particles also influences the concentration for the same elements, unlike TOC that does not display the same correlation. The interface between natural (Unit 1) and anthropogenically influenced sediments (Unit 2) is accompanied by a reduction in Cd. As and Ni concentration are interpreted to be the result of hypoxia, changes in pH or reduced erosional input. As with PCB, metal analyses of stormwater culvert sediments confirm that urban runoff contributes to the metal pollution load at present. Urban soil is also believed to be a pollution source of stormwater sediments.

Paper IV

Paper and plastic packaging have the potential to contaminate both soil and water samples. This paper focuses on analytical quality control and demonstrates the problems when laboratory results are utilised without such quality control. Several examples of PCB results, where an unusual and unexpected PCB congener distribution is presented, lead to further analyses to investigate the source. This demonstrated a high predominance of light congeners. These light congeners are found within the packaging material. Some of the paper packaging analysed had been

manufactured more than 20 years ago, whilst the analysed plastic packaging was produced in 1997 and 2008, i.e. during a time when the use of PCB had already been prohibited. Re-sealable plastic bags were found to contain the highest PCB concentrations. These are used by researchers worldwide as sampling bags for soils and sediments. This paper raises the issue that sample packaging is a potential source of contamination in many previous studies and that analysis of the packaging should be an integral part of the sample analysis prior to sample collection. These important results were taken into consideration when subsequently designing the entire study of LL, the results standing as a cautionary note regarding the suitability of packaging during sediment studies.

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