Holocene pollen and spores variability derived from marine sediment analysis from the Adriatic Sea. Roman Climate Optimum, Industrial Revolution and present day under scope

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Bremen, den 25. August 2019

Salvador Ruiz Soto

"I hated every minute of training, but I said, 'Don't quit. Suffer now and live the rest of your life as a champion.'"

Muhammad Ali

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Abstract

In our days, one of the main topics that concern science and society is climate change. Due to the rising of greenhouse gases concentration in the atmosphere and the increase of land alteration by humans, the global climate has changed dramatically within the last 200 years. This is reflected by recurrent episodes of extreme weather phenomenon, like torrential rain, floods or melting ice sheets. In order to predict future scenarios, it is necessary to understand the current state and the development from past environmental conditions to the ones we have in these days. This can be, among other methods, achieved by reconstructing the paleoenvironment in marine sediments with the use of different proxies that are affected by climatic changes.

The climate has an influence on the vegetation and determines the distribution of the vegetation zones including characteristic plant species. So, a change in the climate can lead to shifts of these zones. Human activities can also affect plant dispersion by e.g. deforestation, monoculture farming and sealing of soils due to the extension of settlement and traffic areas. To differentiate between natural and human induced changes, a comparison between the vegetation in times of less disturbance and strongly anthropogenic influenced environmental conditions is needed.

The goal of this study is to reconstruct the vegetation patterns of the Italian Peninsula and their shifts over time by analysing pollen in marine sediments of the Adriatic Sea and the Gulf of Taranto. This Mediterranean region provides high quality sedimentary records, fulfilling requirements to undertake paleoenvironmental studies with the extra value of having an extensive record of instrumental data of key parameters, like temperature, precipitation and river discharge for centuries. And why pollen? Pollen is produced by all seed plants, having species specific characteristics that enable to directly identify the producing plant species. Found in marine sediments, they provide an integrated and quite reliable information about the current vegetation patterns on land as well as their changes over time, when marine sediment cores covering past time periods are analysed. Besides, pollen is an excellent tool to date sediments. Across the history, there are known episodes of abrupt change in the species composition, which can be seen in the sediments, enabling the correct dating. This is especially useful when other dating techniques are not sensible enough like in very recent sediments from the Holocene. Therefore, pollen studies provide a good understanding of the shifts occurred in vegetation over long time periods as well as the past hydrological conditions and are an excellent tracer for human activities.

As a first step towards a comparison between natural and human-induced vegetation changes in the Italian Peninsula, an overview over the current pollen distribution, and dispersion in the Adriatic Sea and the Gulf of Taranto is obtained by investigating the pollen found in river and sea surface sediments. Marine sediments close to river mouths that are distributed along the coast reflect the signal of the vegetation from the river discharge areas. The finding of typical pollen species shows pollen transport

in suspension within the discharge waters from the north of Italy in sediments of the Gulf of Taranto. Moreover, clear distribution patterns and trends for certain species are found and the intervention in nature by humans as well as the selective use of species is shown. The human marker species Poaceae and Cerealia have their highest occurrence near the Po River discharge point, meanwhile *Olea europaea* reaches its highest abundance in the southern sediments reflecting the major species cultivated in the respective areas. Therefore, in this study the role played by the Po River plume waters for pollen transport in the Adriatic Sea and the Gulf of Taranto is proved.

An insight into vegetation patterns and potential changes during times with less disturbed conditions is gained with the analysis of a well-dated marine sediment core from the Gulf of Taranto covering the time interval 50 BC - 186 AD that can be assigned to the Roman Climate Optimum. During this time period, vegetation is mainly dominated by arboreal species. As an exception, herbaceous species are predominant for a short time period after the year 79 AD. Around this year, a volcanic eruption for the Vesuvius is described. The change in the vegetation is seen with a notable decrease in *Quercus robur* type pollen within the assemblage. Pollen types related to human activity have still low abundance within the pollen spectrum suggesting that the intervention in nature caused by the Romans was more local than regional.

In order to compare the vegetation patterns and changes during the Roman Times to a period stronger influenced by human activities, the pollen from another well-dated sediment core also retrieved in the Gulf of Taranto were analysed. In this case the studied time interval covers 1837 - 2006 AD and so the era during and after the Industrial Revolution. Even if the pollen species not differ from the ones occurring during the Roman Times, the overall pollen concentration is lower during this time period, probably due to constructional measures in the Po River drainage network since the XVII century. High discharge episodes recorded for this time period for the Po River are reflected by an increased occurrence of pollen, spores and dinoflagellates. The time period of the Post-Industrial Revolution also shows a dominance for arboreal pollen over herbaceous taxa. The intense land modification occurred in the Po Valley is not reflected in the marine sediments from the Gulf of Taranto as the Po River signal must be buffered by the discharge waters from other eastern Italian rivers draining in the Adriatic Sea.

This thesis shows the current pollen and spores distribution in the Adriatic Sea and the Gulf of Taranto, the development of the vegetation of the Italian Peninsula during two different past periods in the history, as well as the influence of Po River discharge and alterations caused by humans in the Italian region. It also complements previous studies on dinoflagellates conducted on the same sediments. The results here presented add an important and valuable piece of information for palynology studies in Italy. Such broad overview is usually rarely considered as most studies are focused on local changes without regarding the whole system. The results are in line with the findings from other authors but controversy arises from archaeological results and studies from the Po River area. Therefore, this study is not intended as a final work but a complementary information to the current state of the art. Many uncertainties are not solved, like the specific contribution of the particular eastern Italian rivers, the Aeolian input or the pollen transport and translocation once in the marine realm. That leaves an open door for more detailed studies in the region and comparisons with surrounding areas with the intention to shed light on the unknown aspects that still remain unanswered.

Kurzfassung

Eines der bedeutendsten Themen unserer Zeit, welches die Wissenschaft und die Gesellschaft beschäftigt, ist der Klimawandel. Aufgrund des Anstiegs der Konzentrationen der Treibhausgase in der Atmosphäre und der zunehmenden Veränderung des Festlandes durch den Menschen, hat sich das globale Klima innerhalb der letzten 200 Jahre dramatisch verändert. Dies wird durch wiederkehrende Episoden von extremen Wetterphänomenen wie Starkregen, sowie durch Fluten oder das Abschmelzen der Eisschilde deutlich. Um zukünftige Szenarien vorherzusagen, ist es notwendig, den aktuellen Zustand und die Entwicklung der Umweltbedingungen von der Vergangenheit bis heute zu verstehen. Dies kann, neben anderen Methoden, durch die Rekonstruktion der Paläoumwelt mit Hilfe von verschiedenen Proxies in marinen Sedimenten erreicht werden, die von klimatischen Veränderungen beeinflusst werden.

Das Klima hat Einfluss auf die Vegetation und bestimmt die Ausbreitung der Vegetationszonen mit ihren charakteristischen Pflanzenarten. Aus diesem Grund kann eine Veränderung des Klimas eine Verschiebung dieser Zonen zur Folge haben. Daneben können auch menschliche Aktivitäten die Ausbreitung der Pflanzen beeinflussen, u. a. durch Abholzung, Anbau in Monokulturen und Flächenversiegelung auf Grund der Ausbreitung von Siedlungs- und Verkehrsflächen. Um zwischen dem natürlichen und dem vom Menschen bedingten Wandel zu unterscheiden, ist es nötig, die Vegetation zu Zeiten mit ursprünglichen Umweltbedingungen mit denen zu vergleichen, die stark anthropogen beeinflusst sind.

Ziel dieser Arbeit ist es, die Vegetationsmuster der Italienischen Halbinsel und deren Veränderungen über die Zeit zu rekonstruieren, indem Pollen in marinen Sedimenten des Adriatischen Meeres und des Golfs von Tarent analysiert werden. Diese Region des Mittelmeeres besitzt qualitativ hochwertige Sedimente, die Anforderungen für Paläoumweltstudien erfüllen, mit dem zusätzlichen Nutzen eines großen Datensatzes von jahrhundertelangen instrumentellen Aufzeichnungen wichtiger Parameter wie Temperatur, Niederschlag und Wasserabfluss. Und warum Pollen? Pollen werden von allen Samenpflanzen produziert und besitzen artspezifische Merkmale, die direkt auf die produzierende Pflanzenart rückschließen lassen. Gefunden in marinen Sedimenten liefern sie integrierte und recht zuverlässige Informationen über die derzeitigen Vegetationsmuster an Land sowie deren Veränderungen im Laufe der Zeit, wenn marine Sedimentkerne analysiert werden, die vergangene Zeitperioden umfassen. Des Weiteren sind Pollen ein exzellentes Werkzeug, um Sedimente zu datieren. Aus dem

Verlauf der Geschichte sind Episoden bekannt, in denen es zu abrupten Änderungen in der Artzusammensetzung kam, die in den Sedimenten abgebildet werden und so eine korrekte Datierung ermöglichen. Dies ist besonders hilfreich, wenn andere Datierungsmethoden nicht empfindlich genug sind, wie in sehr jungen Sedimenten des Holozäns. Aus diesem Grund helfen Pollenstudien, die Veränderungen der Vegetation über lange Zeiträume sowie vergangene hydrologische Bedingungen zu verstehen und sind zudem ein exzellenter Marker für menschliche Aktivitäten.

Im ersten Schritt hin zu einem Vergleich zwischen natürlichen und vom Menschen verursachten Veränderungen der Vegetation auf der Italienischen Halbinsel, wird ein Überblick über die derzeitige Pollenverteilung und -ausbreitung im Adriatischen Meer und dem Golf von Tarent erstellt, indem die Pollen in Oberflächensedimenten des Meeres und der Flüsse analysiert werden. Meeressedimente in der Nähe von Flussmündungen, die entlang der Küste verteilt sind, spiegeln die Vegetation der Einzugsgebiete wider. Da für Norditalien typische Pollenarten in den Sedimenten des Golfs von Tarent gefunden wurden, kann der Transport von Pollen in Suspension des Abflusswassers abgeleitet werden. Darüber hinaus werden klare Verteilungsmuster und Trends bestimmter Arten deutlich und der Eingriff des Menschen in die Natur sowie die selektive Nutzung von Arten gezeigt. Die anthropogenen Markerspezies Poaceae und Cerealia kommen am häufigsten nahe der Po Flussmündung vor, während *Olea europaea* am verbreitetsten in den südlichen Sedimenten ist, was die in dieser Region am stärksten angebaute Art reflektiert. Aus diesem Grund wird in dieser Studie die Rolle der Abflussfahne des Po Flusses für den Pollentransport in das Adriatische Meer und den Golf von Tarent nachgewiesen.

Einen Einblick in die Vegetationsmuster und deren potentiellen Veränderungen in Zeiten mit noch ursprünglichen Bedingungen wird durch die Analyse eines gut datierten marinen Sedimentkerns aus dem Golf von Tarent gewonnen, der den Zeitraum 50 v. Chr. - 186 n. Chr. abdeckt und somit dem Klimaoptimum der Römerzeit zugeordnet werden kann. In dieser Zeitperiode wird die Vegetation hauptsächlich von Baumarten dominiert. Eine Ausnahme bildet ein kurzer Zeitraum nach dem Jahr 79 n. Chr., in dem krautige Arten vorherrschen. Für dieses Jahr ist eine Eruption des Vulkans Vesuv beschrieben. Die Veränderung in der Vegetation wird durch die auffällige Abnahme von *Quercus robur* Typ Pollen deutlich. Die Pollenarten, die mit menschlicher Aktivität zusammenhängen, zeigen nur eine geringe Häufigkeit innerhalb des Pollenspektrums, was darauf schließen lässt, dass der Eingriff der Römer in die Natur eher lokal als regional war.

Um die Vegetationsmuster und deren Änderungen während der Römerzeit mit einem Zeitabschnitt zu vergleichen, der stärker von menschlichen Aktivitäten geprägt ist, werden die Pollen eines weiteren gut datierten Sedimentkerns analysiert, der ebenfalls aus dem Golf von Tarent stammt. In diesem Fall umfasst der untersuchte Zeitraum die Jahre 1837 - 2006 n. Chr. und somit die Zeit während und nach der Industriellen Revolution. Auch wenn sich die Pollenarten nicht von denen unterscheiden, die während der Römerzeit auftreten, ist die Gesamtkonzentration, wahrscheinlich auf Grund baulicher Maßnahmen innerhalb des Po Abflusssystems seit dem 17. Jahrhundert, geringer.

Episoden mit starkem Abfluss, die für diesen Zeitabschnitt für den Po aufgezeichnet wurden, werden durch ein verstärktes Auftreten von Pollen, Sporen und Dinoflagellaten angezeigt. Der Zeitraum der postindustriellen Revolution zeigt ebenfalls eine Dominanz von Baumarten über krautige Arten. Die intensiven Modifikationen in der Po-Ebene spiegeln sich nicht in den marinen Sedimenten des Adriatischen Meeres und des Golfs von Tarent wider, da das Signal des Po Flusses durch die Abflusswässer der ostitalienischen Flüsse, die in das Adriatische Meer fließen, gepuffert wird.

Diese Doktorarbeit zeigt die aktuelle Verteilung von Pollen und Sporen im Adriatischen Meer und dem Golf von Tarent, die Entwicklung der Vegetation der Italienischen Halbinsel innerhalb zwei verschiedener vergangener Zeiträume sowie den Einfluss des Po Abflusses und der Veränderungen durch den Menschen in Italien. Sie ergänzt zudem vorangegangene Studien zu Dinoflagellaten, die an denselben Sedimenten durchgeführt wurden. Die hier gezeigten Ergebnisse liefern wichtige und wertvolle Informationen für palynologische Studien in Italien. Solch breiter Überblick wird normalerweise nur selten beachtet, da sich die meisten Studien auf lokale Veränderungen konzentrieren und nicht das Gesamtsystem betrachtet wird. Die hier präsentierten Ergebnisse stimmen mit denen anderer Autoren überein, doch es zeigen sich Kontroversen zu archäologischen Erkenntnissen und zu Studien im Gebiet des Po Flusses. Deshalb ist dieses Werk nicht als abschließende Arbeit anzusehen, sondern als Ergänzung zu dem aktuellen Stand der Forschung. Viele Unklarheiten, wie der spezifische Beitrag der einzelnen ostitalienischen Flüsse, der äolische Eintrag oder der Transport und die Verlagerung von Pollen innerhalb des Meeres werden nicht gelöst. Dies lässt Spielraum für weitere detaillierte Studien in der Region und für Vergleiche mit den umgebenden Gebieten, um die unbekannten und noch unbeantworteten Aspekte aufzuklären.

Chapter 1

Introduction

1.1 General motivation and unique features of the project

The importance of marine coastal environments from an economic and social point of view is extraordinary. Concern on environmental deterioration, especially during the last decades, is growing due to the relationship between anthropogenic activities, such as eutrophication and pollution of coastal waters, and human induced climate change (e.g. Smith & Schindler, 2009). Such human impact on the environment, especially after the Industrial Revolution has lead experts to name this time period also as a new era in the geological time, "Anthropocene" (Crutzen & Stoermer, 2000). The contribution of human activities to climate change is also gaining traction in political debates as governments are weighing up how to minimize the effects. This is seen by the growing number of initiatives and policies made from governments all around the world, at both national and international levels, to mitigate the influence on the environment caused by anthropogenic activities and to provide a future without compromising a healthy environment. Such initiatives and policies need a solid scientific background, which can assess the current situation, the most likely future scenario and the strategies for a sustainable development with the minimum climate deterioration.

Understanding and differentiating between natural and anthropogenic processes is not an easy task, because despite detailed and growing amounts of information it is difficult to understand all the processes happening in the environment. This problem gets more tangled and more difficult to tackle when attempting to assess human impact before the 1950s, because of fewer records or unreliable instrumental data. Instrumental data and more recent monitoring represent another challenge because the measurements are done in a period that is already influenced by humans. Not only the anthropogenic bias challenges the correct interpretation of the data and the correct measurement of the natural processes, the short temporal record of high-quality data cannot be readily used as a model to explain temporal and spatial variability in the long term (e.g. Lutherbacher et al., 2012, 2016). However, certain techniques like the establishment of climate models or the study or sedimentary archives allow gaining insight on longer time intervals with the extra value of discerning between natural and human induced climate change. None of these methods provides a full answer to climatic studies but these are complementary. Models studies can produce high temporal and spatial resolution data but this data needs to be tested and validated against real and measured data. The model itself does not have any insight in the steering mechanisms controlling the dynamic of the system. This insight is usually gained by the study of sedimentary archives, which also provide a high temporal resolution, and so complement the information lacking in the model. Considerable progress has been made during the last years by combining proxy data and climate model simulations (e.g. Lutherbacher et al., 2016; Christiansen & Ljungqvist, 2017; Zhang et al., 2018). Nevertheless, not all the sedimentary archives meet the requirements to be used for environmental reconstructions. For this purpose, a sedimentary archive requires to have very high quality in terms of extraordinary time control, preservation of signals that can be used as proxies, allow the record of data in a high temporal resolution in a relative continuous and long time intervals and be located in areas where even small climate variations are recorded. Due to the strict list of requirements to be fulfilled, high quality sedimentary records are rather uncommon (Dupont, 1999). So, an exceptional case is a sedimentary record, which fulfil all the requirements described and is located where extra information of instrumental data of key parameters covering pre- and post-industrial time is available.

Such extraordinary conditions are found in the Adriatic Sea and the Gulf of Taranto, being a region that provides high quality sedimentary archives. Moreover, there is minimal disturbance in the sediment composition due to anaerobic conditions. In this region, not only high quality sedimentary records are found but also the unique benefit of having available the longest and oldest environmental records registered by calibrated instruments. The instrumental record for air temperature covers the periods between 1654-1670, 1716-2007 (see Camuffo & Bertolin, 2012a,b), and 1725-1998 (Camuffo, 2002), while the rainfall record covers the last 300 years (see Camuffo et al., 2012c) and Po River discharge record the last 200 years (Zanchettin et al., 2008). All these measurements are done in a very high temporal resolution allowing a fine, detailed analysis and understanding of the steering mechanisms. The whole region is highly influenced by discharge waters from riverine system. From all the rivers draining water into the Adriatic Sea, the Po River contributes with the greatest budget (Goudeau et al., 2013). The confluence of the discharge waters result in a freshwater plume being transported in the marine realm southwards, reaching the Gulf of Taranto. This plume contains fine particles in suspension which sediment within the gulf with a sedimentation rate ranging between 0.49-0.92 mm/yr (Goudeau et al., 2014). In addition, sediment records from the Gulf of Taranto have a special feature, which makes them ideal to establish an age model for recent material. Due to the existence of volcanoes in the regions and the information of their eruptions, an exact age model can be calculated by mean of volcanic glass analysis.

Perhaps the biggest challenge of environmental science is to try to understand the climatic changes that occurred historically and to make future predictions that do not only answer general public interest but also give a more clear statement about the measures to mitigate the worst effects of human induced climate change. The interest and concern from the public domain about how climate will be in the future and how it will affect humanity is increasing because of current events of abrupt and constant rise in the global temperatures (IPCC, 2013). The knowledge, interest, and opinion on this topic may be reinforced by the opportunities that television, social media and internet offer (Eurobarometer, 2011; Anderson, 2017). The world is no longer restricted to our street, neighbourhood or city but we live in an interconnected world where assessing information and news from remote locations around the planet is easier than ever in history. This increases the chances of finding information but it is not exempt from leading the readers to misinformation (Vosoughi et al., 2018). This fact also leads to a loss of trust in institutions that are responsible for research due to the lack of rigor and contradictions that people can find through the different sources of information. It creates confusion and to some extent, a negative effect on people who do not see it clearly and the uncertainty and noise created makes it difficult to accept the evidence. Media has certain impact on society's awareness of climate change and even on the behaviour humans may adopt for everyday activities, although there is no simple explanation for that (Arlt et al., 2011).

Therefore, we, as researchers, have the responsibility not only to publish new data, new interesting discoveries, and new processes but practising a better way of science communication (Greenwood, 2001; Leshner, 2003). Communicating the findings, the advances, and the improvements in a more understandable and accessible language to everyone is a must for the scientific community (Somerville & Hassol, 2011). The Quaternary, and more specifically the Holocene, is probably not the best scenario to start with because of the several and acute climatic variations that happen during this period (Pillans & Gibbard, 2012). The Holocene is characterized by constant climatic changes in short periods of time, with no fixed patterns and duration but usually in a millennial-scale as many different proxies show (Guiot, 1987; Adams et al., 1999; Bradley, 1999). These sudden and random changes affect a wide range of environmental parameters like temperature, ocean circulation, atmospheric gases or ice cover but whose in-depth trigger mechanism remains yet unknown. According to the position from the International Union for Quaternary Research (INQUA) found in Pillans & Naish (2004), the Quaternary is the most important time period of the Earth from an anthropological point of view due to the frequent and abrupt climatic, oceanographic and biotic changes whose major event include the debut and evolution of the Human species. The human implication in this play set is of major importance to evaluate future scenarios bearing in mind many of these changes occurred on the very short time scale, even comparable to human lifespan (Adams et al., 1999).

For this reason, a deep understanding and knowledge of the climate fluctuations and the role of the humans on these climate fluctuations and so on the vegetation change can shed some light in such an interesting and important topic and provide to certain extent answers to the impact caused in the region by anthropogenic activities.

1.2 Brief introduction to proxies and anthropogenic traces in Italy

Mediterranean regions have always been a hot spot to study climate change as many studies show. These studies are based on a wide variety of proxies to conduct paleoenvironmental reconstructions. Proxy data can belong to analysis in marine sediment cores (e.g. Chen et al., 2011, 2013; Piva et al., 2008; Schilman et al., 2001; Sangiorgi & Donders, 2004; Sangiorgi et al., 2005; Taricco et al., 2009; Versteegh et al., 2007; Zonneveld et al., 1997, 2000, 2001, 2008, 2009, 2010a, 2010b, 2012), lake sediments (e.g. Wick et al., 2003; Jones et al., 2006; Roberts et al., 2008; Woodbridge & Roberts, 2011), speleothems (e.g. Bar-Matthews et al., 1997, 1999, 2000, 2003; Frisia et al., 2005, 2008, 2012, 2015; Mangini et al., 2005; Orland et al., 2008, 2009, 2012), corals (e.g. Felis et al., 2000, 2003, 2004; Rimbu et al., 2006) or even from studies started in the 1990's where tree rings were used (e.g. Galli et al., 1992; Touchan et al., 2005).

This situation of having such a high number of studies in the region is not random. The Italian Peninsula, more specifically in the Pirro Nord site (Apulia, southern Italy), presents evidence of activities performed by species from the Homo genus already in the Early Pleistocene between 1.3 and 1.7 Ma BP (Arzarello et al., 2007). These findings are not evenly spread across the Italian Peninsula suggesting a still quite primitive and local population but already set the basis and show the long history of occupation by Homo species. Until 7500-4000 BP signals of local farming are recorded in the sediments in central and southern Italy through pollen analysis that show anthropogenic activities and hence, some kind of vegetation distortion and land management (e.g. Vannière et al., 2008; Colombaroli et al., 2009; Tinner et al., 2009; Pittau et al., 2012). This land management and vegetation disturbance is not limited to the central and southern parts. Studies from the Po River catchment area, covering the southern slopes from the Alps, the Po Valley and the northern Apennines, date back to around 7000 years BP with signals of anthropogenic activities (Tinner et al., 1999; de Wit & Bendoricchio. 2001; Mercuri et al., 2006; Wick & Möhl, 2006). Even though this long record of human occupation, population expansion and regional traceable anthropogenic signals do not appear until a few millennia later. According to diverse studies (e.g. Allen et al., 2002; Oldfield et al., 2003; Di Rita & Magri, 2009), distortion of the land do not reach a regional scale until around 4000 BP for the northern part of the Peninsula and until around 2200 BP for the central and southern parts of the Peninsula. The first anthropogenic effects registered in sedimentary records throughout the whole Italian Peninsula are found at the time in which the Roman Empire was expanding. This rather fast change in the population and the effects it caused on the environments, boom of the agriculture, deforestation to use wood as resource and the land for crops and river embankment to benefit from permanent water supply, lead to an increase in the surface of the Po River Delta as a result of an increase of erosion in the upper and medium sector of the river (Ciabatti et al., 1967; Marchetti, 2002). This increase in sediment load transported by the Po River during this time period cannot be explained, as demonstrated by Chen et al. (2011), by cyclical component of the climate and is attributed to human effect on the environment. Information extracted from pollen analysis and marine records points out a rather complete recuperation of the natural to its less disturbed conditions as the Roman Empire collapsed (Oldfield et al., 2003). To see such enormous land change caused by humans in this region in the recent history, time has to pass until 800 BP, then 500-400 BP and finally since the beginning of the 17th century up to present (Stefani & Vincenzi, 2005).

Up to this moment, all the disturbances, alterations and modifications of the environment were related to changes in the vegetation coverage including deforestation and agricultural activities, mining and building activities. After the Industrial Revolution, due to an increase in agricultural activities, the population growth and the introduction of fertilizers as productivity enhancers lead to an increase in the eutrophication of river waters, like in the Po River, and so of the coastal waters (e.g. Marchetti et al., 1989; Sangiorgi & Donders, 2004; Zonneveld et al., 2012). Such eutrophication of the environment resulted in an increase of harmful algal blooms, more acute in the northern Adriatic Sea due to the intense use of fertilizers in the Po plain region (Justič et al., 1987; de Wit & Bendoricchio, 2001). This effect was not obviated by the governments and after forbidding the use of specific components in the fertilizers, the occurrence of these algal blooms has decreased since then (Mozetic et al., 2010).

1.3 Pollen

According to Trigo et al. (2008), pollen, as derived from Latin *pollen-inis*, means very fine dust or flour, and was initially used by Linnaeus. A pollen grain is the male gametophyte formed by pollen sacs in angiosperms and gymnosperms. Pollen grain size is very variable ranging from about 5 to 200µm. Even though pollen grains can have different shapes, spherical and oval are the most common ones (Pacini, 2015). They are generated by meiosis of stem cells in the pollen sacs, resulting in microspores, which lead to pollen grains after development. In each pollen grain, two different parts can be distinguished:

1. Inner part, containing three cells. One cell is in charge of forming the pollen tube once the pollen grain is on the stigma surface. One generative cell, which will allow the reproductive cycle, and one vegetative cell which will degenerate, also called vegetative.
2. Outer part, forming the shell and protecting the living cells, can also be divided in two different layers with different chemical composition. The inner layer, intine, consists mostly of cellulose, hemicellulose and pectines (Kovacik et al., 2009). The outer layer, exine, is extremely resistant, made of sporopollenin (Rowley & Skavarla, 2000) acting as a defence against environmental threads like corrosion, ultraviolet light, high temperatures or desiccation, and allowing the viability of the inner cells when they reach the female organ (Scott, 1994; Frenguelli, 2003).

Once the pollen grains are formed, they can be liberated as single grains (monads), coupled-grains (dyads), in groups of four grains (tetrads) or more (polyads). Following Erdtman (1969), the unique architecture of the exine makes it possible to establish a relationship between pollen type and species of plant producing this pollen type. Pollen can also have different types of apertures, which vary in the shape and the specific position around the pollen grain. Such is the variability in shape and number, position, and type of the apertures that Erdtman (1952) proposed a system (NPC), which summarizes the different options one can find when attempting to identify pollen.

Pollen can be transported through animals, air currents and less commonly, water. This is of particular interest because it will have an effect on their distribution pattern. Each means of transport has its particular efficiency, specificity and theoretical distance that pollen can travel (Pacini, 2015). The study of pollen can provide quite valuable information to fields like medical science, agriculture, ecology, meteorology, and criminology but also allows conducting paleoclimatic and paleoenvironmental reconstructions based on fossil pollen. Pollen records from the Italian Peninsula have been studied numerously (Oldfield, 2003; Sangiorgi & Donders, 2004; Mercuri et al., 2006; Vannière et al., 2008; Colombaroli et al., 2009; Di Rita & Magri, 2009; Tinner et al., 2009; Mercuri et al., 2012; Mercuri & Sadori, 2012; Pittau et al., 2012; Mercuri et al., 2013).

In this particular case, we will focus on pollen found in marine sediments transported by wind and riverine systems after being displaced in the majority of the cases long distances. This general occurrence of pollen found in marine sediments from all around the world makes it especially suitable for our purpose.

It is well known, pollen found in marine archives provide an integrated and to some extent, a quite fidelity information about changes occurred in vegetation, and hence in the climate conditions. The covered time in some cases spans over long and continuous periods, even reflecting the change between different eras. According to Dupont (1999), paleoreconstructions based on pollen studies provide a good understanding in the shifts occurred in vegetation during long time periods accompanied by the hydrological conditions. In other studies (e.g. Sangiorgi & Donders, 2004; Hooghiemstra et al., 2006; Mercuri et al., 2012b), the suitability of the combination of studies based on pollen to undertake direct land-sea correlation is shown. With this, it is not meant to say that vegetation and climate reconstruction based on pollen analysis do not require some special care, and key information must be taken into consideration. In Dupont (1999), a defined list can be found with key aspect to consider, like the huge variation in pollen production among the different species, the transport along the water column in the ocean, the possible resuspension and further transport conducted by ocean currents, sedimentation, an early diagenesis and the fossilisation process within the sediment. Due to the long distances pollen travel from the source to the sedimentary material in which they are found, having a precise and detailed knowledge about the play role of each transport vectors is of vital importance prior to any interpretation of the paleorecord.

1.4 Dinoflagellate cysts

Dinoflagellates, protist microorganisms belonging to Dinoflagellata division, are unicellular, eukaryotic and ecological diverse occurring in almost all aquatic environments and latitude ranges (Taylor, 1987; Dale & Dale, 1992; Matthiesen et al., 2005; Bravo & Figueroa, 2014). Size varies between 20 and 200 µm for the majority of the species. Dinoflagellates reach their maximum species diversity in regions with warm waters (Stover et al., 1996) but they also form complex communities in polar regions. As one of the major groups of the marine phytoplankton, they are important primary producers (e.g. Parsons et al., 1984; Taylor, 1987; Dale & Dale, 1992; Marret & Zonneveld, 2003). Despite the phototrophic characteristic shown by many dinoflagellates, they can also be heterotrophic or mixotrophic, obtaining the energy from other organisms such as diatoms or dissolved organic matter or as a combination of autotrophic and heterotrophic feeding strategy respectively (Jacobson & Anderson, 1986).

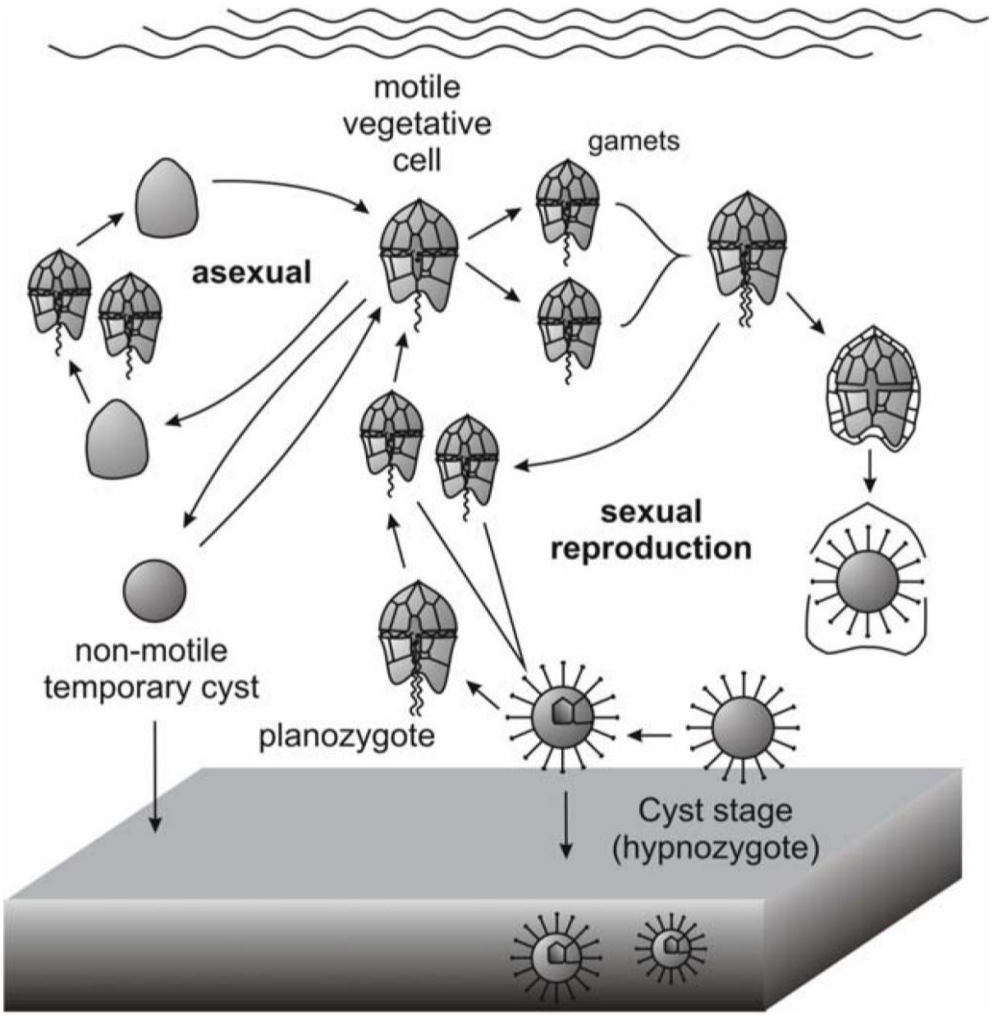

Figure 1-1. Simplified life cycle of cyst-producing dinoflagellates (in Bockelmann, 2007, after Dale, 1986).

In general, dinoflagellates have different strategies related to the motility of the cell, changing from non-motile (cyst) to motile stages. The motile stage is characterized by the presence of two flagella; one embracing the cell and the other one in a longitudinal position confers free movement across the vertical gradient in a so-called whirling motion (Fensome et al., 1993). Dinoflagellates can be athecate, also known as naked, or thecate with an “armour” of cellulose plates. Dinoflagellates’ life cycle is usually described as complex due to the high changes occurring during the reproductive stage, which can be sexual or asexual (Figure 1-1). According to Taylor (1987), the sexual stage produces the majority of the dinoflagellates. Moreover, during the sexual stage, dinoflagellates produce gametes, which fuse and in some species form a hypnozygote (Dale, 1986). The hypnozygote, the so-called resting cyst, is protected by an outer cyst wall allowing the organism to survive longer during dormancy periods (e.g. Wall & Dale, 1967; Fensome et al., 1993). The most common chemical component of the cyst wall is dinosporin (Kokinos et al., 1998), which leads to a high resistance against unfavourable environmental conditions. The resistance is comparable to sporopollenin found in pollen grains. Some other species present calcite or silica walls (Head, 1996). Organic-walled dinoflagellate cysts (dinocysts) are formed during the biological encystment process, which may be linked to environmental parameters like solar irradiance, day length, temperature or inner encystment periods (Anderson & Keafer, 1987). This cyst period of dormancy is reversible and is called excystment. During this process, the protoplast hatches through a small aperture in the wall, the archeopyle. Like the process of encystment, the excystment is also controlled by environmental factors like temperature, nutrients availability and oxygen concentration (Dale, 1983).

Dinocysts have become an increasingly important tool to undertake paleoceanographic/ paleoenvironmental reconstructions, especially in regions where dissolution hampers the preservation of mineralized microfossils (e.g. Dale & Fjellså, 1994). The number of studies involving dinocysts from sediment records, after the pioneer ones conducted by Wall & Dale (1967) and Wall et al. (1977), has experienced a rapid growth covering a wide range of environments and habitats, ranging from tropical to polar regions, including both marine and fresh waters (e.g. Bouimetarhan et al., 2009a,b, 2012, 2013; Chen et al., 2011, 2013; Dale et al., 1999, 2002; Dale, 1996, 2009; de Vernal et al., 1994, 1997, 2001, 2007, 2018; Esper & Zonneveld, 2007; Marret & Zonneveld, 2003; Marret et al., 2001, 2008; Marret, 1994; Pospelova et al., 2002, 2005, 2006, 2008, 2010, 2015, 2018; Radi et al., 2001, 2007; Sangiorgi & Donders, 2004; Sangiorgi et al., 2005; Versteegh, 1997; Zonneveld & Brummer, 2000; Zonneveld & Pospelova, 2015; Zonneveld et al., 1997, 2000, 2001, 2007, 2009, 2010a,b, 2012; Zonneveld, 1997). Dinocysts present some features, which makes them an excellent tool to perform studies on past climate conditions. Their habitat is not restricted to a specific area, as they are found in almost all aquatic environments. They are very sensitive organisms, reacting to small variations in environmental conditions, which is “recorded” in the dinocysts association for the studied region, allowing to reconstruct palaeoenvironmental oceanic conditions like sea surface temperature, salinity, nutrient availability and/or upwelling (e.g. Bouimetarhan et al., 2009b; de Vernal et al., 1997, 2001; de Vernal & Marret, 2007; Dale et al., 2002; Marret et al., 2001; Sangiorgi et al., 2005; Pospelova et al., 2006, 2008). Studies on modern sediments, or in the water column can shed some light on the extent of human impact on marine ecosystems in a defined location by monitoring freshwater discharge, which helps to trace pollution due to industrial and agricultural activities (Dale et al., 1999; Pospelova et al., 2002, 2005; Sangiorgi & Donders, 2004; Radi et al., 2007; Pospelova & Kim, 2010; Zonneveld et al., 2012), being even more important in estuarine and fjords systems because of their particular geographical features (Radi et al., 2007; Dale, 2009; Zonneveld et al., 2009). Moreover, the reaction to nutrients, trace elements and pollutants seems to be nutrient/pollutant and region specific, allowing a better understanding of each specific region (e.g. Matsuoka, 1999; Pospelova et al., 2002, 2005; Krepakevich & Pospelova, 2010; Shin et al., 2010).

Based on the dinoflagellate's species-specific cyst morphology, their sedimentary distribution reflects the distribution of the particular motile stage in the upper water column at a low taxonomic level (Zonneveld et al., 2008) and their occurrence is controlled by i.e. nutrient availability, turbulence, temperature and salinity (Taylor, 1987). For a reliable interpretation of the sedimentary record, it is important to know about the possible post-depositional influences on dinocyst preservation. Studies on sediment traps at different water depths at one mooring site indicate rapid transport through the water column without species-specific degradation (Zonneveld & Brummer, 2000; Susek et al., 2005; Zonneveld et al., 2010b). Besides bioturbation and relocation of the sediments, especially aerobic degradation can have a major impact on the settled cysts (Zonneveld et al., 1997; Versteegh & Zonneveld, 2002; Zonneveld et al., 2008).

The use of dinoflagellates also has its limitations. It is important to know that some species are catalogued as resistant and others as non-resistant, when the dinocyst association is altered after the sedimentation process due to aerobic organic matter (OM) degradation (Versteegh & Zonneveld, 2002; Zonneveld et al., 2007; Kodrans-Nsiah et al., 2008). It was found that several dinocyst species are vulnerable to aerobic degradation while other species rank among the most resistant parts of the marine organic matter (Versteegh & Zonneveld, 2002). Field and laboratory studies revealed that heterotrophic dinocyst species are more sensitive than autotrophic species (e.g. Zonneveld et al., 1997; Hopkins & McCarthy, 2002; Versteegh & Zonneveld, 2002; Zonneveld et al., 2008; Bogus et al., 2014). The difference in sensitivity is thought to depend on different cyst wall compositions that is determined by the nutritional strategy of the organism (Bogus et al., 2014). This can lead to misinterpretation of marine sediments due to the degradation of selected species, altering the signal recorded. The fact, that marine sediments represent a compilation of, usually, many years makes it impossible to retrieve accurate seasonal information about the dinoflagellates associations but also about the environmental conditions (Zonneveld et al., 1997).

In this study, dinocyst data of published data sets is used to shed some light on the land-sea correlation in the studied region.

1.5 Marine palynology

The information provided by marine palynology data complements the data acquired by other disciplines and studies including isotopic, radiogenic, geomorphic, correlation, biochemical or geochemical analysis. It is widely used to reconstruct paleoceanographic and paleoclimatic scenarios, with a special importance when the precision of the other dating techniques does not allow establishing an accurate age model (e.g. Newnham et al., 2018; Boehnert et al., submitted 2019 - see Appendix A-5).

Pollen found in marine sediments represent an excellent tool to reconstruct inland vegetation and human impact on the vegetation (see Montade et al., 2019 and references therein). This is supported by studies conducted by Heusser & Morley (1985), Hooghiemstra et al. (2006), Mercuri et al. (2012a) or Combourieu-Nebout et al. (2013, 2015) in which they state that pollen found in marine sediments and hence, its interpretation reflect the regional vegetation. It also has its limitations due to environmental changes in short periods of time. Having a precise set of samples, which are entirely comparable among them, is difficult due to the nature of the marine sediments. Surface samples in general, do not represent a single year but a sequence of years depending on the sedimentation rate. Moreover, to conduct a more realistic analysis, detailed information about the relationship between the numerical values and the vegetation, in terms of pollen producers, is needed. The lack of this valuable information can lead to wrong analysis and interpretation caused by over- or underrepresentation of certain species (Erdtman, 1969). Moreover, straight correlation between pollen found in the sediments and its source usually cannot be done, because there is no information about pollen production and specific dispersion (Behre, 1981). Even with this information, it is not a simple task. A large proportion of the pollen fraction is lost due to filtering in the forest or adhesion at physical structures (Whitehead, 1983).

There is also a species-specific selection of the pollen found in the marine sediments. Aeolian transport of wind pollinated pollen types is predominant in areas with low to none river discharge (Heusser & Morley, 1985; Hooghiemstra & Agwu, 1986). Although this transport can carry pollen far away from the source, the pollen concentration and the distance from the source follow an inverse relationship - the greater the distance, the smaller the concentration in the air (Tampieri et al., 1977; Mandrioli et al., 1984; Spieksma, 1992). Pollen size also affects the transport capabilities. Long transport is reported to be extremely effective for small pollen types (Cabezudo et al., 1997; Cecchi et al., 2006; Fernández-Rodríguez et al., 2014), like for *Betula* or *Olea* (Siljamo et al., 2008; Hernández-Ceballos et al., 2011; Rojo & Pérez-Badia, 2015). The shape also plays a role, which is the case for pollen of *Abies* and *Pinus*, which can be transported long distances too (Whitehead, 1983; Rousseau et al., 2008). Other pollen types, even from the same family, like *Picea* are supposed to stay more locally (Hick, 2001). *Quercus* pollen type is not well defined according to what extent it can be transported. Some studies (e.g. Mandrioli et al., 1982; Recio et al., 1999) conclude its transportation capabilities are more restricted but in Rousseau et al. (2008) it is considered as long transported pollen. Nevertheless, a potential Neves-effect can also alter the pollen spectrum (Chaloner & Muir, 1968). Sediments from coastal areas and river mouths vicinities have a more diverse pollen spectrum (Ruiz Soto et al., to be resubmitted; See Chapter 4.) than sediments from abyssal or deep-sea sites. Pollen and spores preservation also plays a central role. Badly preserved pollen grains challenge the identification with corrosion as the most important effect altering the shape and the recognizable characteristics of the grains (Andersen, 1970).

1.6 Scientific objectives

The main objective of this project is to provide information about the exact relationship between anthropogenic activities and changes occurring in the environment using as resource marine sediments retrieved from the Adriatic and Ionian Sea, including the Gulf of Taranto. The southeastern Italian coastal ecosystem in the Gulf of Taranto represents, in these days, a hot spot to study environmental degradation due to human activities, especially due to the increase eutrophication in the region.

This study combines the information of the current pollen distribution across the region and a paleoclimatic and paleoenvironmental reconstruction of two time periods, Roman Climate Optimum (50 BC – 186 AD) and Post-Industrial Revolution (1837 AD – 2006 AD), to improve the understanding of the vegetation dynamics in the region and to assess anthropogenic activities and environmental parameters.

The questions, which came up and guided this study, are:

- What mechanisms and processes control the modern pollen and spore distribution of terrestrial origin in the western Adriatic Sea, the northern Ionian Sea and the Gulf of Taranto?
- How can the present palynomorph distribution be related to anthropogenic activities and current climate scenarios?
- What is the characteristic provenance of the different river discharge plumes in the region in terms of palynological imprint?
- In a sub-decadal period, what is the relationship between climate, anthropological activities and change in the vegetation of the Italian Peninsula during the Roman Climate Optimum (50 BC – 186 AD)?
- In a sub-decadal period, what is the relationship between climate, anthropological activities and change in the vegetation of the Italian Peninsula during the Post-Industrial Revolution (1837 AD – 2006 AD)?

The approach trying to answer this question is based mainly on terrestrial palynomorphs (pollen and spores) with the aim to assess the changes occurred in the vegetation of the Italian Peninsula. Another proxy, dinocysts, is used in combination with the terrestrial signal to gain insight in the relationship between land-ocean, which can allow correlating different environmental parameters like temperature and rainfall.

1.7 Outline

The thesis is written and organised in a cumulative form. Chapter 1 represents a detailed introduction on the topic and scientific background involving the different projects. In chapter 2, the environmental setting of the study area is described in terms of climate, oceanography and vegetation. Chapter 3 covers the general methodology and materials used in a condensed way. Chapters 4, 5 and 6 comprise the three different projects conducted within the frame of the doctoral thesis. A brief description for each chapter (4-6) is presented below. Chapter 7 consists of summary and conclusions achieved throughout the projects and chapter 8 a brief outlook with possible future studies in the region to broaden the knowledge of the processes taking place there. The references of chapters 1, 2, 3 and 7 are listed in chapter 8.

Manuscript 1 (Chapter 4):

"Pollen and spores distribution patterns in the Adriatic and Ionian Sea and derived riverine fingerprint"

Salvador Ruiz Soto, Karin A.F. Zonneveld, Francesca Sangiorgi, Ilham Bouimetarhan, Timme H. Donders

To be resubmitted, *Review of Paleobotany and Palynology*

This manuscript aims to examine the current pollen and spore distribution in the Adriatic and Ionian Sea and its possible provenance by also analysing selected rivers. A set of 63 modern top core samples belonging to 55 marine sediments core (Adriatic Sea, Ionian Sea and Gulf of Taranto), 7 rivers and 1 lagoon have been analysed. All these samples enable to set the modern palynomorphs distribution in the region, the specific contribution of each river to the system and the possible influence of the dominating wind systems blowing in the region. Results obtained from this paper show the current pollen and spores species distribution within the Adriatic Sea and the Gulf of Taranto. It was not possible to determine the individual contribution for the main two blowing system (Bora and Sirocco) but it was possible to set pollen provenance according to riverine fingerprints.

The study was designed by K.A.F. Zonneveld, T.H. Donders and S. Ruiz Soto. Sample material was provided by K.A.F. Zonneveld and a collection of already prepared slides was supplied by F. Sangiorgi. Sediment processing, samples preparation and analysis under the microscope was performed by S. Ruiz Soto. Data interpretation and manuscript writing was done by S. Ruiz Soto in collaboration with all the co-authors.

Manuscript 2 (Chapter 5):

"Reconstructing Italian vegetation development in the Roman Climate Optimum and Roman Transitional Period (50 BC – 186 AD) using pollen and spores found in marine sediment from the Gulf of Taranto"

Salvador Ruiz Soto, Karin A.F. Zonneveld, Timme H. Donders

In preparation. Target journal: *Estuarine, Coastal and Shelf Science*

In this study, we analyse the palynological content of a well-dated sediment core (~3.5 years temporal resolution) retrieved from the Gulf of Taranto covering the time period 50 BC - 186 AD. During this time the general vegetation pattern remains quite constant as so do the climate. Arboreal pollen are generally more abundant than non-arboreal pollen. A major pollen disturbance is seen around the year 79 AD related to a Vesuvius eruption. After this eruption, arboreal pollen decrease notably, especially *Quercus robur* type, in favour of herbs pollen. Remarkable is the appearance of *Pistacia* at the upper end of the core section compared to scattered appearances before 767.5mm. *Olea europaea* or *Castanea sativa* have low values in general. The presence of typical alpine pollen types, like *Picea*, in the sediments of the southern Gulf of Taranto supports the hypothesis that pollen are transported suspended in marine waters tracing back to the Po River. The combined analysis of pollen and dinocysts concentration shows a striking correlation for both trends along the studied section.

The study was designed by S. Ruiz Soto and K.A.F. Zonneveld. Sample material and slides were provided by K.A.F. Zonneveld. Sediment processing, samples preparation was previously done by former colleagues of the group. Microscopic analysis was performed by S. Ruiz Soto. Data interpretation and manuscript writing was done by S. Ruiz Soto in collaboration with all the co-authors.

Manuscript 3 (Chapter 6):

"Vegetation trend reconstruction during the Post-industrial Revolution based on pollen and spores using a well-dated marine sediment record from the Gulf of Taranto (South Italy)"

Salvador Ruiz Soto, Karin A.F. Zonneveld

In preparation

In the frame of this study, we reconstruct the pollen and spores trend of a well-dated sediment core (~3.5 years temporal resolution) retrieved from the Gulf of Taranto covering the time interval 1838 - 2006 AD. Within this time period, trends for arboreal and non-arboreal pollen remain practically constant, with a dominance of arboreal pollen. Pollen types related to the north Italian peninsula are found in the sediments as they are transported in suspension to the Gulf of Taranto in plume waters discharged by the Po River. No big human intervention is found in the pollen assemblage, due to low percentages for pollen related to human activity. Pollen and dinocyst concentrations present a positive linear correlation. Years in which high Po River discharge or floods are described are reflected as peaks in the palynomorph concentration. These results demonstrate the tight relationship between Po River discharge and palynomorphs found in the marine sediments in the Gulf of Taranto. However, no specific vegetation reconstruction for the Po Valley region can be derived from the dataset as a progressive dilution of the Po River signal occurs as the discharge waters mix with other small rivers draining into the Adriatic Sea along the east coast.

The study was designed by S. Ruiz Soto and K.A.F. Zonneveld. Sample material and slides were provided by K.A.F. Zonneveld. Sediment processing, samples preparation was previously done by former colleagues of the group. Analysis under the microscope was performed by S. Ruiz Soto. Data interpretation and manuscript writing was done by S. Ruiz Soto with the supervision of K.A.F. Zonneveld.

Chapter 2

Environmental setting

2.1 Regional climate

The Mediterranean region is located between the northern part of the African continent, the southern part of the European continent and western Asia. This location, extending from warm to temperate latitudes, extends from 30°- 46° N (Luterbacher et al., 2012) strongly conditions the particular regional climate characteristics. There is a large seasonal contrast between summer and winter for temperature, rainfall and dominating wind regimes. As described in Saliot (2005), the climate in the Mediterranean region has a dipole-like character, ranging from dry summers to cool and wet winters. This is the due to the air circulation controlled by the confluence of three main systems, the Scandinavian pattern, the East Atlantic (EA) pattern and the North Atlantic Oscillation (NAO) (Hurrell & Van Loon, 1997; Cassou et al., 2004; Fil & Dubus, 2005). However, the Mediterranean climate can vary year to year and the net NAO index has even a quite intra-annual variation (Ramdani et al., 2009).

From these air circulation systems, the NAO has the most effect because the region lays in its transition zone, and changes in NAO index provoke high changes in the climate of the region affecting especially temperature and rainfall across Europe, and hence over the Mediterranean region and Adriatic Sea. The NAO index depends on the surface sea-level pressure difference between the Azores and Iceland, with high and low pressure, respectively (Hurrell, 1995). When the NAO index is in positive-phase, an excess of rainfall occurs in northern Europe and stable and dry conditions in southern Europe, including the Mediterranean. A negative NAO index reverts the previous situation, with relatively dry conditions in central northern Europe but it results in an excess of rainfall and higher temperatures in southern Europe, including the Mediterranean and the Adriatic Sea (Lionello & Sanna, 2005). This dependence of the system on the NAO index is observed in the resulting river discharge of Mediterranean rivers. An increase in rainfall will result in higher water discharge from the Po River and, in general, all of the other Italian rivers, which increase the amount of surface waters in the Adriatic Sea (Chen et al., 2011).

Regionally, the two principal wind systems control the atmospheric circulation in the region, the Bora and the Sirocco (Kourafalou, 1999; Artegiani et al., 1993). The Bora is a dry, cold and continental wind blowing from the northeast (Artegiani et al., 1993; Orlić et al., 1994; Palinkas & Nittrouer, 2006), playing a key role on the evaporation and heat loss (Artegiani et al., 1993), but it also triggers the formation of deep water in the north Adriatic Sea (Hendershott & Rizzoli, 1976; Kourafalou, 1999). The Sirocco is a moist wind blowing from the southeast transporting humid and warm air into the Adriatic region as it travels over the Mediterranean Sea incorporating moisture caused by the evaporation (Artegiani et al., 1993, Cavaleri et al., 1997; Pasaric et al., 2007). Establishing a clear seasonality for the occurrence of each wind regime is not possible as both can co-occur any time of the year. Nevertheless, it is more common to have strong Sirocco winds in spring (Sivall, 1957). The fact that Bora and Sirocco have different characteristics and origins results in differing effects, that each one has, on the plume formed by Po River discharged waters as they enter the Adriatic Sea. Moreover, they influence regionally the sea level and oceanic circulation, even though they do not have an extensive duration (Orlić et al., 1994). The modification of these parameters can be directly correlated to changes in water temperature and hence, to the amount of dissolved oxygen (DO), which can trigger oxidative reaction resulting in different preservation rates of pollen and spores (Versteegh & Zonneveld, 2002).

2.2 Oceanic circulation

Oceanic circulation is highly influenced by the climate, the size and the shape of the basin. The Mediterranean Sea has a semi-closed shape, which acts as a concentration basin. Evaporation rate within the basin is larger than the freshwater input. The only point where the Mediterranean Sea has a considerable exchange of water is through the Strait of Gibraltar. Through this Strait, warm and fresh (15.4 °C, 36.2 PSU) Atlantic water enters the basin at surface level and increases its salinity as it moves eastwards. The cold and salty (15 °C, 38.4 PSU) Mediterranean water, on the contrary, exits the basin as a current of dense water both at intermediate and deep depths (Laurent et al., 2012). The Mediterranean Sea itself is subdivided in smaller seas and sub-basins connected to the main system through narrow straits. Each one will have its particular characteristics regarding sea bottom orography, salinity, sea surface temperature or freshwater input, but they all share, in general, the same principles applying to the regional climate of the Mediterranean region.

One of the sub-basins of the Mediterranean Sea is the Adriatic Sea. This large, elongated and semi-closed basin is located between the Italian Peninsula, and the Balkan Peninsula oriented in an NW-SE direction, with a unique water exchange point with the Mediterranean Sea through the Strait of Otranto. Water depth ranges from 35 m in the north to 1200 m in a depression in the south before reaching a more shallow depth again at the Strait (Artegiani et al., 1993). This variation in the water depth is the reason for some authors to establish a subdivision of the Adriatic Sea according to the bathymetry (northern, central, southern), increasing in depth from north to south (Orlić et al., 1992; Zavatarelli et al., 1998; Bensi et al., 2013).

This latitudinal gradient, differential in water temperature and bathymetry affects the amount of Dissolved Oxygen (DO) too. Hence, it is possible to find zones with different DO, and zones of hypoxia. The occurrence of these hypoxia zones is very variable. For some regions, hypoxia zones can occur in an episodic manner or seasonally but there is also a lot of uncertainty for other zones. It is studied and defined that most hypoxia zones are concentrated in the northern part of the basin (European Environment Agency, 2016). The occurrence of hypoxia zones in the north has been increasing as a response to an increase in eutrophication (Justič, 1991). In the central and the southern Adriatic, below

100m, a clear difference in DO values lead to the hypothesis of a weak exchange connection between the two water masses that meet at the depth (Lipizer et al., 2014).

The oceanic surface waters within the Adriatic Sea are dominated mainly by two water masses, the Adriatic Surface Waters (ASW) and Ionian Sea Surface Waters (ISW) responsible for the particular and regional surface circulation. Each water mass has its own physical fingerprint. The ASW is formed from discharged fresh water, drained into the system by the rivers, like the Po River among others, found on the eastern Italian margin by relatively cool, suspended matter, high chlorophyll-a and low salinity waters (Degobbis et al., 1986; Kourafalou, 1999; Boldrin et al., 2005). These discharged waters do not spread homogeneously across the basin. They flow southwards, usually confined to the Italian coast because of the cyclonic surface currents induced by Coriolis forcing, (Artegiani et al., 1997b; Lee et al., 2007; Colombaroli et al., 2009).

Po River discharge is the major net contributor to ASW. This fresh water supply takes place in the northeast of the basin through a vast and complex deltaic system. The second surface water mass of the Adriatic Sea, the ISW enters the basin through the Strait of Otranto and has completely opposite physical characteristics compared to the ASW. ISW is characterized by relatively high temperature and salinity but low suspended matter and nutrient concentration (Artegiani et al., 1997a, b). As it enters the basin from the south, it flows northward along the Balkan coast, on the eastern margin of the southern Adriatic Sea. Both water masses, ASW and ISW start mixing as they meet already in the north Adriatic Sea but the completion of the mixing occurs outside the basin, within the Gulf of Taranto (Socal et al., 1999; Boldrin et al., 2005; Caroppo et al., 2006; Lee et al., 2007).

Besides ASW and ISW, another important water mass can be found in the Adriatic Sea but also in the Gulf of Taranto, is the Levantine Intermediate Water (LIW). This water mass presents high temperature, salinity, and relatively high nutrient content and can be observed at intermediate water depths ranging from 150 - 440 m (Nittis & Lascastaros, 1999; Sellschopp & Alvarez, 2003). Immediately underlying this ILW, Modified LIW is found. It represents a transition zone to the Adriatic Deep Water, with characteristics from both water masses (Sellschopp & Alvarez, 2003). Adriatic Deep Water (ADW) is a deeper water mass formed by much cooler and dense water than the overlying water masses previously described (Sellschopp & Alvarez, 2003; Hainbucher et al., 2006; Rubino & Hainbucher 2007).

2.3 Po River

According to the Po River Basin Authority (Autorità di bacino distrettuale del fiume Po), the Po River the longest river with a length of 652 km and has the highest discharge in Italy. The total Po River basin is around 74,000 km^2 wide, of which 71,000 km^2 are situated in Italian territory and the remaining area in Switzerland and France. It flows eastwards from the western Italian Alps and drains its water in the North Adriatic Sea through a complex deltaic system, with a spatial extension of 380 km^2. The Po

River acts as an aggregator, for 141 smaller rivers and affluences, for the southern part of the Alps and the northern part of the Apennines forming an extended drainage basin (Figure 2-1). This vast drainage basin is the Po valley, covering the regions of Piedmont, Valle d´Aosta, Liguria, Lombardy, Veneto, Emilia-Romagna, Tuscany and Autonomous Province of Trento, concentrate almost 16 Mio. inhabitants acting as the most important resource region for the country. Economic activity represents 40 % of the national Gross Domestic Product (GDP), aggregating 37 % of the national industry and 35 % of the national agricultural activities (Po River Basin Authorithy, accessed 2019). This high population density and anthropogenic activities generate also a great impact on the hydrological fluxes. Total water withdrawal is estimated to be around 20.5 billion m³/year, of which 6.0 billion m³/year come from groundwater and 15.5 billion m³/year from surface waters. Irrigation is the most water demanding activity, requiring 16.5 billion m³/year (~80 %), meanwhile water withdrawal for potable and industrial use is around 4 billion m³/year (~20 %). In Montanari (2012), a hydrological flux summary is presented (Figure 2-2) The temperature in the Po River basin has a continental character with temperatures in winter below 0 °C and in summer above 25 °C but with an average temperature above 10 °C. Precipitation is on average less than 1400 mm per year (Blasi et al., 2014).

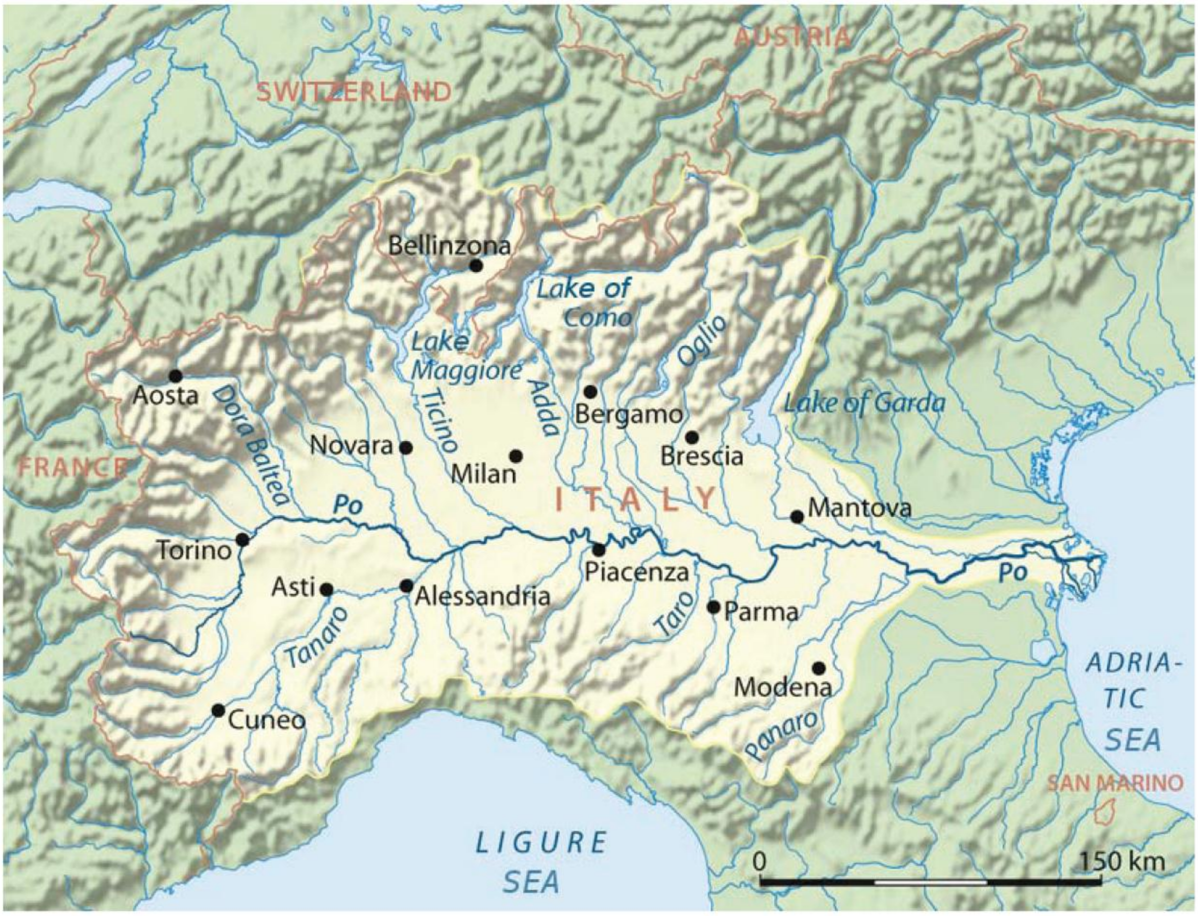

Figure 2-1. Map of the Po River basin (from Wikipedia, in Montanari, 2012).

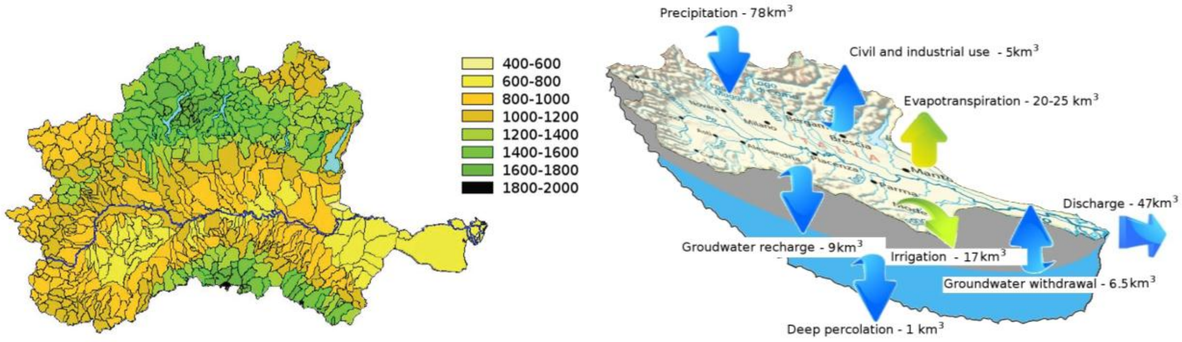

Figure 2-2.Left: Mean annual rainfall over the Po Basin (from Po River Basin Authority, 2006, in Montanari, 2012); right: Mean annual main hydrological fluxes for the Po River basin (in Montanari, 2012).

Following Nelson (1970), sediment load of the Po River belonging to the Alpine mountain range and the Apennine mountain range have an equal proportion. A bifurcation of the sediments provided by the Po River takes place after exit the Po Delta, and some sediments are transported to the northeast by the northern Adriatic gyre (Wang & Pinardi, 2002). The fine-grained sediments are transported southwards too. In addition, resuspension of the material also occurs in the marine realm (Wang & Pinardi, 2002). Sediments transported by the Po River can settle on the Po shelf or remain in suspension and flow southwards (Figure 2-3); being one half for each option. Sedimentation rate is exceptionally great at the Po shelf, with ranging between 1 and 4 cm/yr. (Palinkas & Nittrouer, 2007).

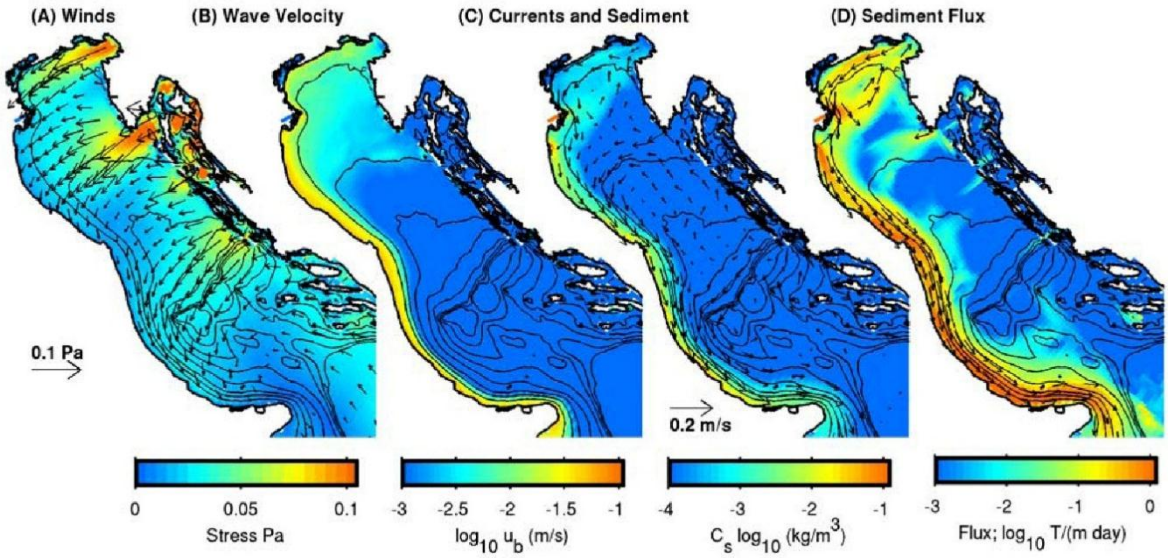

Figure 2-3. "Time-averaged (A) wind stress and (B) wave orbital velocity, (C) depth-averaged suspended sediment concentration (shading) and current velocity (arrows), (D) depth-integrated daily averaged sediment flux (t/m · d) in the northern and central Adriatic Sea." (in Harris et al., 2008)

This sediment supply and variation through the time sets the extension of the Po Delta under constant fluctuation. Progradation of the delta during the first half of the 20th century was slower than in the previous 200 years, but the maximum extension was recorded between the 1930s and 1940s. Since then and even with the constant and voluminous supply of sediments from the Po, the delta has been retreating (Syvitski & Kettner, 2007). In addition, the Po Delta shows high alterations caused by humans, by filling most of the wetlands with sediments, reclaiming land for agriculture or changing the river runoff through canals (Cencini, 1998)

Boldrin et al. (2005) estimate that the amount of freshwater drained from the Po River is on a long-term average about 1511 m^3/s. However, the hydrology of the Po River can vary drastically through the year. Sporadically high discharge events can occur but high discharge seasons are usually spring and autumn, due to snow melting in the mountains and seasonal rain respectively, conditions which can rise up the discharge up to around 2000 m^3 (Kourafalou, 1999; Boldrin et al., 2005). Meanwhile, it has a lower water level in summer and winter. Water discharge by the Po River has a significant influence in the characteristics and the dynamics of the Adriatic Sea, affecting parameters like temperature, salinity, sedimentation and nutrient availability but also the water circulation (Giordani et al., 1992; Tankéré et al., 2000). Despite the great number of river draining in the Adriatic Sea, Po River supply one third of the total amount of fresh water entering the basin (Kourafalou, 1999).

The discharge and the development in the marine system of the waters discharged by the Po and Apennine rivers is strongly controlled by atmospheric circulation. Po discharge waters are pressed and confined to the eastern Italian coast when Bora winds blow but it will also move the water plume southwards along the coast. This situation is completely different when Sirocco winds blow. In this case, Po plume will spread eastwards in the northern Adriatic (Orlić et al., 1994; Kourafalou, 1999; Palinkas & Nittrouer, 2007). Flooding events occur, due to water accumulation in the shallow northern Adriatic, in the Po mouth when high tides and strong Sirocco winds blow (Orlić et al., 1994; Pirazzoli & Tomasin, 2002; Ferrarese et al., 2008; Jeromel et al., 2009). Even though episodic floods seem to follow a 5 years-cycle, long-term predictions of caudal are still far from being modelled or established (Montanari, 2012)

The contribution, in terms of sediment and water, for other smaller rivers from the region is significantly low compared to the Po River discharge (Penna et al., 2004; Milligan & Cattaneo., 2007). However, a mix of sediments from the Po River and Apennine rivers characterizes the sediments from the northern part of the Adriatic Sea (Cattaneo et al., 2003). An estimation of the final deposition Po River and Apennine rivers sediments is illustrated in Figure 2-4. This counts also for the discharged waters. Water discharged by the Po River and constituting the Po plume is loaded with sediments, nutrients and freshwater elements from other local eastern Italian rivers too (Penna et al., 2004; Milligan & Cattaneo, 2007).

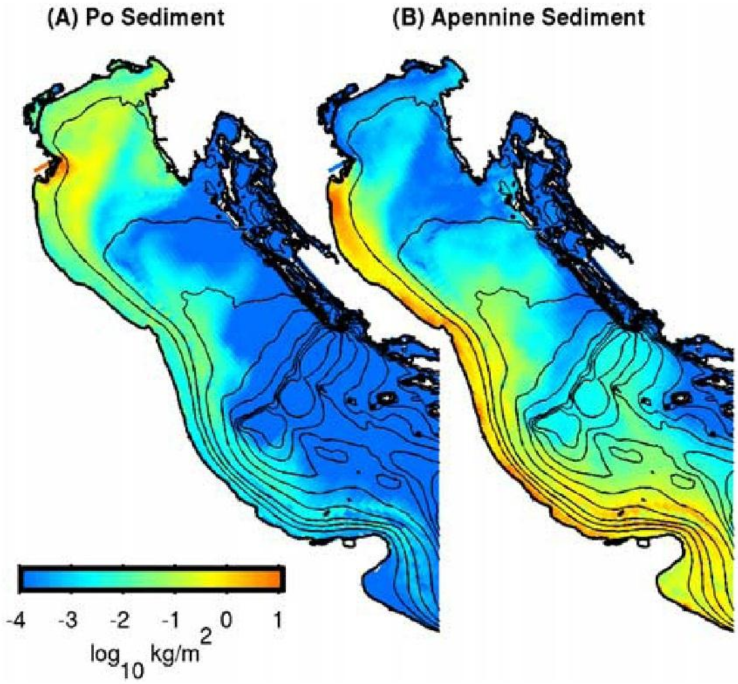

Figure 2-4. "Final deposition of fluvial sediment estimated for the (A) Po River and (B) Apennine rivers. Depth contours at 25 m up to 200-m water depth." (in Harris et al., 2008)

2.4 Mediterranean Vegetation and its characteristic in the Italian Peninsula

The Mediterranean basin has one of the highest biodiversity catalogue of the world, for total number of species but also endemism. Vegetation diversity is speculated to be around 25.000 species, comprising flowering plants and ferns, for which 50 % are listed as endemic (Allen, 2001).

In Lang (1994, and references therein), a comprehensive description and summary of the vegetation across Europe and the Italian Peninsula can be found. Many of the following information was extracted and synthetized from his work, especially from an overview over the recent natural European vegetation. Vegetation in the European region is in principle controlled by climatic factors such as temperature or precipitation. In relation to these parameters and their values, it is possible to distinguish five different vegetation zones across Europe, with each having particular climatic conditions and so, specific dominating groups of species. Vegetation zones are (after Lang, 1994):

- Artic and Alpine
- Boreal
- Temperate
- Mediterranean
- Pannonica-Pontica-Anatolian

Though the Italian Peninsula is under the influence of the Mediterranean climate, an altitudinal gradient controls the vegetation distribution. That leads to a wider climatic condition, due to different levels of temperature, rainfall and/or solar radiation. It can also be seen in the different vegetation belts in the Peninsula, making it possible to find typical Mediterranean forest but also regions with a temperate and even boreal forest. Two Mediterranean, three temperate and one boreal zones are described by Lang (1994) for the region.

The Mediterranean zones, thermo-Mediterranean and meso-Mediterranean, cover the coastal Peninsula from the border to France to a bit more northern of Gargano on the Adriatic Sea coast, where they are already substituted by the temperate vegetation. It is characterized by an evergreen and sclerophylle forest, rich in bushes and herbs. According to Lang (1994), the thermo-Mediterranean forest, found at coastal and low altitude, is constituted mainly by a mixture of bushes, olive trees (*Olea europaea* var. *sylvestris*), evergreen *Quercus* (*Quercus suber*), Carob trees (*Ceratonia siliqua*), Mastic trees (*Pistacia lentiscus*) and *Myrtus communis*. The meso-Mediterranean forest transition into the thermo-Mediterranean forest as altitude increases. This still keeps a lot of similarity with the thermo-Mediterranean forest, but there is a wider spectrum of *Quercus* trees (*Q. suber*, *Q. ilex* type, *Q. coccifera*) and from the oleaceae family species like *Phillyrea latifolia* gain more weight in the association.

Temperate vegetation zone described and detailed in Lang (1994) with its three sub-modalities, Middle-European forest, sub-Mediterranean and supra-Mediterranean thermophile mixed forest, covers the central part of the peninsula, the Apennines, and the Po River Valley. The Middle-European mixed

Quercus forest is characterized by species like *Q. robur* type, *Carpinus betulus*, *Tilia cordata,* and *Acer platanoides*. Sub-Mediterranean and supra-Mediterranean thermophile forests, with a rich variety in *Quercus* (*Q. pubescens*, *Q. petrae*, *Q. cerris*, *Q. pedunculiflora*, etc), have also species like *Fraxinus ornus*, *Ostrya carpinifolia*, *Carpinus orientalis, Castanea sativa* among others. The last of the temperate zones is found at a higher altitude as the two previous ones, is a forest characterized by the dominance of species like *Abies alba* (silver fir) and *Fagus sylvatica* (beech) (Lang, 1994).

Boreal vegetation zone is restricted to the upper part of the Alps. It comprises a needle-leaved forest rich in *Picea abies*, *Abies alba*, a variety of *Pinus* species (e.g. *Pinus cembra*, *Pinus sylvestris*, *Pinus mugo*), *Larix decidua* (Lang, 1994). Similar species are described for this location in Blasi et al., (2010).

The fact that the temperate zones are so widely distributed and cover even more surface than the Mediterranean zones, underline the transitional zone in which the whole peninsula finds itself. When compared to other Mediterranean regions, like the Iberian Peninsula, though being at the same latitude, Mediterranean vegetation zones in the Italian Peninsula cover less surface as shown in Lang (1994). From a phytogeographic point of view, particular interest attracts the Po Valley region. It represents the boundary between Central European and Mediterranean regions, having a mix of species from both phytogeographic regions (Cencini, 1998). Herbs represent a numerous and well-distributed group of plants found in almost the total extension of the peninsula. Especially successful are herbs from Chenopodiaceae, *Plantago*, *Urtica*, *Artemisia*, Asteraceae, and Poaceae associated to a variety of substrates and land uses (Pignatti, 1982) and some of them are related to a dry land and human effect (Sangiorgi & Donders, 2004). Herbaceous taxa like *Typha*, *Ephedra* and Cyperaceae have a wide distribution linked to pits, lakeshores, and riverbanks. *Ephedra* is more related to coastal and dry lands (Pignatti, 1982).

Cultivated species with culinary uses *Corylus avellana*, *Castanea sativa*, and *Olea europaea* spread across the whole peninsula (Pignatti, 1982). Cultivation of *Olea europaea* has caused an expansion of territory in which this Mediterranean tree can be found, trespassing the bioclimatic limits, growing at higher altitudes and latitudes what makes its present distribution do not reflect that of the wild variety (Carrion et al., 2010).

After the broad overview of the general vegetation characteristics dominating the Italian Peninsula, a more in detail vegetation distribution of selected species in the Po Valley, Alpine and Apennine regions follows. Po River valley vegetation shows an altitudinal zonation (Combourieu-Nebout et al., 2015) and can be divided in three different vegetation zones:

- Alpine and subalpine forest covering the sides of the Alps.
- Pre-alpine vegetation, temperate forest.
- Low alpine and Po plain.

In the alpine/subalpine forest corresponding to the boreal zone previous described by Lang (1994) and Blasi et al. (2010), the dominant species are *Picea abies, Abies alba* and *Larix decidua*, forming an evergreen coniferous forest in the Alps and the northern part of the Apennines. *A. alba* is distributed across the lower Alps and the Apennines but can be found in the south of the peninsula and in forest with a different vegetation zone character (Parducci et al., 2004; Kehlet et al., 2004).

Pre-alpine vegetation is characterized by a temperate deciduous forest. As described before, this vegetation belt is mainly rich in *Fagus sylvatica* and *Abies alba* but also include other species like *Acer, Alnus, Salix* and *Ulmus*, a cultivated conifer forests and some open pastureland (Pignatti, 1979; Ferrari, 1997; Blasi et al., 2010, 2014).

The low alpine and Po Valley region support an intensive agricultural activity. They are mainly covered by a mix of evergreen and deciduous mesophilous forest rich in *Quercus robur* type, *Carpinus orientalis*, *Carpinus betulus*, *Ostrya*, *Fraxinus* (Cencini, 1998; Blasi et al., 2014), in conjunction with a vast agricultural use of the land (Giglio, 2006). These species of trees previous mentioned are described as typical central European species and within the Po River Delta they are mixed with Mediterranean species like *Quercus ilex* type or *Phillyrea angustifolia* (Cencini, 1998). Some of the goods cultivated in this region are maize, corn, sugar beet, bairley, soybean, rice, and horticulture in general (Giglio, 2006).

Dunes, recreational areas, and coastal woods have also experienced a degradation of their natural vegetation coverage and vegetation communities, being local species devastated in favour of mainly wood species (Cencini, 1998). The genus *Pinus* deserves a special mention due to its broad and widespread distribution across the whole peninsula. The numerous representatives (e.g. *P. sylvestris*, *P. maritimus*, *P. pinaster*, *P. cembra*, *P. mugo*, etc.) can be found high in the mountain but also in coastal habitats (Pignatti, 1982; Lang, 1994), and associated to anthropological activities (Blasi et al., 2010). The fact that it has such a wide soil, temperature and water requirements makes it a genus commonly used in reforestation activities (Pignatti, 1982) and protection against soil erosion (Calama et al., 2007). A similar case is described for *Juniperus communis*. This species is also well distributed across the Italian Peninsula even though some distribution restrictions may apply due to its preference for arid land (Pignatti, 1982).

The central part of Italy is dominated by the Apennine mountain range. Regarding the available information about vegetation coverage in this region by e.g. Lang (1994), Pignatti (2011), Blasi et al., (2010, 2014) and Combourieu-Nebout et al. (2015), the vegetation consists of a typical temperate deciduous species in the lower part, flanks and the southern Apennines. The low Apennines and the coastal domain are characterized by a Mediterranean forest, resulting in an overlapping zone of both vegetation belts on the slope of the Apennines, which leads to a very diffuse transition between the zones. Here, almost all the arboreal species mentioned for the Po Basin can be found, including also *Alnus glutinosa*, *Populus alba* and *Salix*. This species require rich and moist soils but their spatial distribution is not restricted to a particular location and can grow broadly in the peninsula (Pignatti, 1982). Different is the case for *Fraxinus excelsior* and *Betula pendula*. These two species have stricter soil requirements, growing only in wet forest and so are confined to the middle-north Apennines and the Alps (Pignatti, 1982).

Sclerophylle vegetation type dominates the coastal line (from Liguria to the Gargano Peninsula) and low altitude slopes in the Apennines. Typical Mediterranean species dominate the vegetation spectrum, with *Olea europaea* of exceptional importance, in combination with *Ceratonia*, *Thymus*, *Juniperus* and a broad variety of *Quercus* species (Lang, 1994; Blasi et al., 2014). In addition, some species of *Pinus* are also quite widespread in this sclerophylle forest (Pignatti, 1982).

Chapter 3

Material and Methods

3.1 Marine sediments - Core description

A set of marine surface samples have been collected by multicoring during cruises P339-CAPPUCCINO (2006), P411-CARPACCIO (2011), P488-CAPRICCIO (2013), PRISMA (1998) and the river samples collected in a field expedition (Zonneveld et al., 2008, 2013). Marine samples were retrieved using multicorer, which provides undisturbed water/sediment interface, being the best option for studies involving modern sediment deposition and the establishment of data set (Gersonde & Seidenkrantz, 2013).

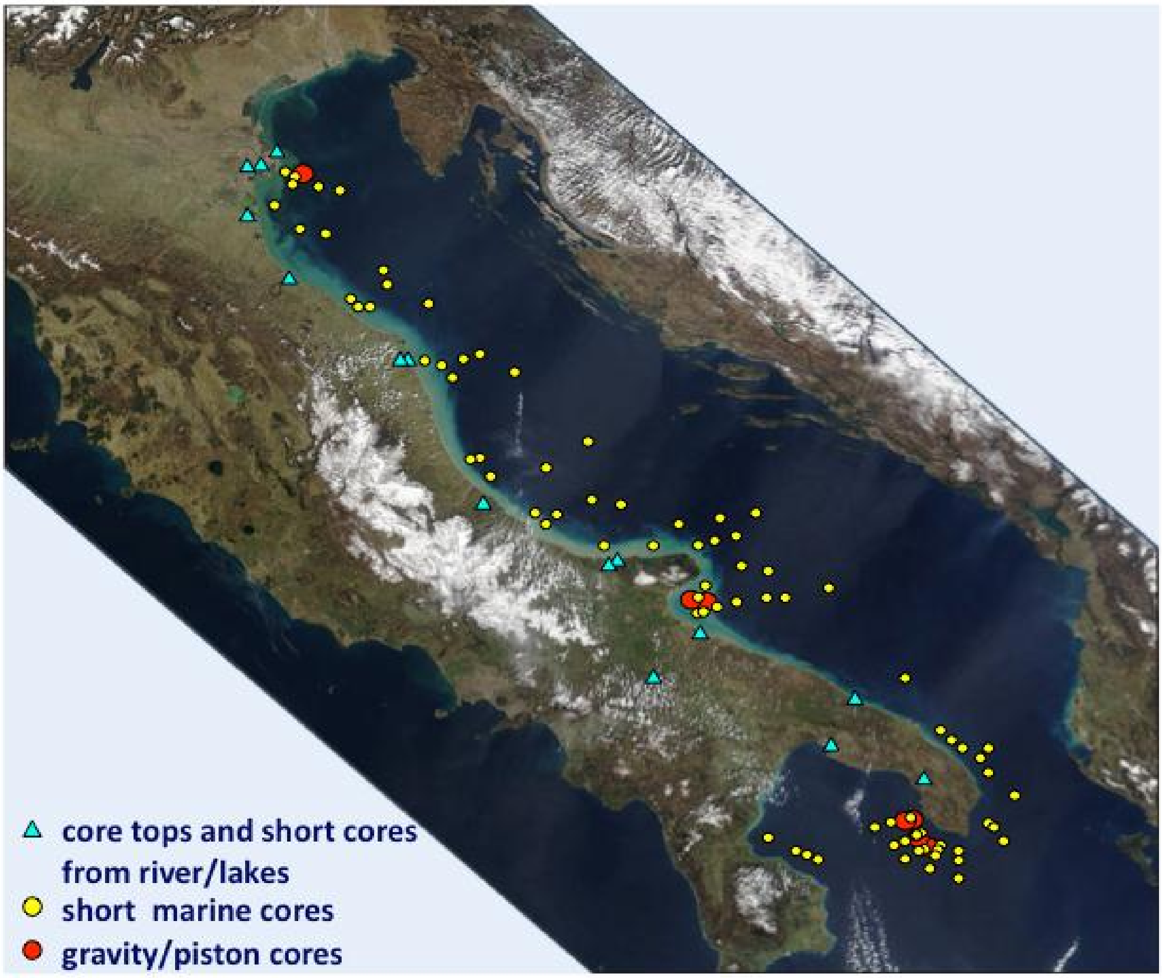

Figure 3-1. Satellite image of the study area with sample locations (modified from NASA, provided by Prof. Zonneveld).

The first project consists of a 63 samples, 55 marine and 8 inland. 55 marine surface samples were retrieved, using multicores, from the Adriatic Sea and Gulf of Taranto (Figure 3-1). They cover a wide extension, from 45.10° to 39.64°N, and water depth ranges between 18 and 1587m. 7 river and 1 lido samples were retrieved by push coring.

Marine Core DP30PC, used for the second study, retrieved during the RV Pelagia (DOPPIO cruise), 2008, located at the ocean margin (39° 50.070 N,17° 48.050 E) and at a water depth of 270 m (Figure 3-1) is composed of homogeneous olive-grey silt. Subsampling of the core was done at every 2.5 mm providing, according to the age model, around 3.725 years resolution (Chen et al., 2011).

Marine Core GeoB-10709-5, used for the third study, retrieved during the RV Poseidon (Cappuccino cruise, 2006; Zonneveld et al., 2008), located at the ocean margin (39° 45.390 N,17° 53.570 E) and at a water depth of 172 m (Figure 3-1) is composed of homogeneous greenish-grey soft mud. Subsampling of the core was done at every 2.5 mm providing, according to the age model, around 2.13 years resolution (Zonneveld et al., 2012).

3.2 Samples processing

Samples were prepared for palynological analysis using standard laboratory procedures without the acetolysis step (Faegri & Iversen, 1989). Wet sediment (1 cm^3) was dried overnight (24h), reweighed and decalcified with diluted HCl (10 %). One tablet of exotic *Lycopodium* spores (18.583 ± 1.708 spores per tablet – Batch N° 483216) was added to the samples before decalcification in order to allow calculation of palynomorphs concentrations. The dissolved carbonates were removed by 3 consecutive steps of decanting and refilling the samples with water with 24h waiting time between the decanting steps to minimize sediment lost. The decalcified sediment was treated with HF (40 %) to remove silicates. Samples with HF were agitated for 2 hours and left with HF solution for 2 days without extra agitation. The supernatant solution was neutralized with demineralized water and decanting steps. The residues generated were neutralized in an external container with KOH (40 %). After chemical treatment, samples were wet sieved over a 10 µm nylon mesh screen using an ultrasonic bath to disaggregate organic matter. Down-core samples are sieved over a 20 µm metal sieve. The residue was concentrated in Eppendorf vials with 1.5 ml water. An aliquot (50 µl) was mounted on a permanent glass slide using glycerine jelly and the slide sealed with paraffin wax.

Counts were set to a minimum 200 pollen grains per site, when possible. The counting process was performed with a light microscope at 400-x, and 1000-x magnification for grains, which were unclear. Pollen grains were identified following Moore et al. (1991), Trigo et al. (2008), Beug (2015) and the reference collection of the Department of Palynology and Paleoecology at the University of Utrecht (The Netherlands). Both aliquot method and *Lycopodium* spore counting methods were used depending on the characteristics of the samples. Palynomorph concentration for surface samples was calculated by mean of *Lycopodium* method, except for a defined small set of samples. *Lycopodium* method is based on an extrapolation. Each *Lycopodium* tablet contains a defined number of spores. According to Stockmarr (1971), the calculation of the concentration of pollen and spores is extrapolated from the number of *Lycopodium* spores counted (see equation below). For the down-core analysis, all calculations were according to the aliquot method by Zonneveld et al. (2009).

Palynomorphs results are expressed as percentage, concentration, and accumulation rate (palynomorph flux). Percentage of pollen and spores is expressed as the percentage of each count compared to the total sum. The analysis was performed in two stages. First including *Pinus* data for the calculation and a second time without considering *Pinus*. In this way, the overrepresentation for *Pinus* in the dataset is avoided and the contribution and association among the other species is clearer to establish (Erdtman, 1969). Palynomorph concentration per cubic centimetre and per gram, were calculated following the equations:

$$\text{Palynomorph} / cm^3 = (P \times L) / (l \times V)$$
$$\text{Palynomorph} / g = (P \times L) / (l \times N.W.)$$

P: Palynomorph counts.
L: Lycopodium spores added.
l: Lycopodium spores counted.
V: Volume of sediment processed in cm^3.
N.W.: Net weight processed in grams.

Accumulation rate was calculated for the palynomorphs analysed down-core in conjunction with the sedimentation rate data derived from the age model. The result is expressed as palynomorphs per cm^2 per year (palynomorph / cm^2 / year) and it is calculated following the equation:

$$AR = \text{Palynomorph} / cm^3 \times SR$$

AR: Accumulation rate or palynomorph flux (palynomorph /cm^2 /year).
SR: Sedimentation rate (cm / year)

Samples preparation for projects 2 and 3 with material from cores DP30PC and GeoB-10709 were already available in the samples archive of the Marine micropalaeontology department (MARUM). For more information about the methodology followed to prepare the samples see Chen et al. (2011) and Zonneveld et al. (2012) respectively.

3.3 Statistical analysis

Standard multivariate ordination techniques were performed on the relative abundance data of pollen and spores in order to find association trends within the species found in the assemblage. The analysis was performed using CANOCO v.5. (Ter Braak & Smilauer, 2012). The linear (unimodal) or nonlinear (bimodal) character of the dataset is tested. Principal Component Analysis (PCA) was performed, within the dataset. In any case, the results obtained from the analysis allow establishing groups of species with similar environmental requirements and potentially same source. In the study involving the surface sediment dataset from the Adriatic and Ionian Sea, the PCA also allowed establishing the provenance of the different pollen species according to the distribution. The position of any point, represents its optimum abundance within the dataset when taking the perpendicular projection of the point over the axis.

For species richness, two different methods have been performed, the Shannon-Wiener biodiversity index and rarefaction analysis. Rarefaction analysis was performed for the period covering the Roman Climate Optimum (manuscript 2, chapter 5) Shannon-Wiener biodiversity index was calculated for two different projects, the surface sediment project and the Post-Industrial Revolution period. The purpose, for the first project (surface sediments), was to identify locations with a higher diversity of pollen types and to assess the effect that distance to the coast or river mouth has over the different pollen types found in the sediments. For the third project (Post-Industrial Revolution), the aim was to spot changes in species richness during the studied period. Calculations of the Shannon-Wiener biodiversity index (H′) were done on the raw counting data and following the equation:

$$H' = -\sum_{i=1}^{R} p_i \ln p_i$$

p_i = proportion of individuals belonging to the *i*th species within the sample.

Chapter 4 - Manuscript 1

"Pollen and spores distribution patterns in the Adriatic and Ionian Sea and derived riverine fingerprint."

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Keywords: Po River; pollen distribution; surface sediment; spores; Adriatic Sea; Ionian Sea

Highlights:

- 63 surface sediments samples have been investigated, 55 marine - 8 riverine.
- Po River discharge waters can still be recognized in the Gulf of Taranto.
- Arboreal pollen dominates the assemblages, high % of monocots in Po River mouth.
- On-shore sites have a higher diversity than those offshore.
- Rivers signals are reflected in marine sediments close to discharge site.

Abstract

Terrestrial palynomorphs (pollen and spores) retrieved in marine sediment cores are often used to investigate hydrological cycle and freshwater input to the marine realm in the past. Studying terrestrial palynomorphs in marine sediments provide us with the unique opportunity to investigate terrestrial and marine signals of climate and environmental changes simultaneously. However, little is known about the present-day distribution patterns and provenance of pollen and spores, which deposit in marine sediments after being transported by river waters. This information is key to perform reliable paleoreconstructions.

To obtain insight into the transport ways, distribution, provenance, and associations of pollen and spores in the Adriatic Sea and Gulf of Taranto we investigated the palynomorph content of surface sediments from 7 eastern Italian river mouths and 55 marine sites. We show that there is a clear difference between pollen/spore associations that are deposited in coastal sites where surface water masses consist of river discharge waters, the so-called Adriatic Surface Waters (ASW), and offshore

sites, which surface water masses, consist of Ionian Sea Waters (ISW). We observed a striking coherence between the pollen/spore association of the river mouth sediments and those of the marine samples in close vicinity, indicating that fluvial transport is the major transport way of the ASW-sample palynomorphs. Palynomorphs of sites outside the river plume (ASW) most likely have undergone aeolian transport. The palynomorph associations of marine ASW samples can be divided into 4 major associations:

1.) "Po River association" dominated by Monocot pollen and containing high relative abundances of *Picea*, *Larix* and Chenopodiaceae.

2.) "Central Italian river association" characterized by high proportions of pollen from trees that are specific for mesophilous forest including *Carpinu*s, *Corylus*, *Fagus* as well as herbs such as *Rumex*, *Plantago* and *Artemisia.*

3.) "mixed association" characterized by relatively high relative abundances of cosmopolitan species such as those from the Asteraceae, Chenopodiaceae, and *Ephedra.*

4.) "southern association" characterized by a Mediterranean sclerophyll forest pollen/spore assemblage with high relative abundances of *Olea*, other Oleaceae, *Pistacia*, *Phillyrea* and *Quercus ilex* type.

Between the region off Ancona and the Gargano Peninsula, the "Po River association" signal is observed in samples recovered from sites that underlie the most offshore part of ASW waters whereas more coastal ASW samples contain a "central river association". South of the Gargano Peninsula and in the Gulf of Taranto the signals are mixed or formed by a "southern association". We also show for the first time, that pollen brought into the marine realm by the Po-river can be observed throughout the entire plume, is still traceable in the Gulf of Taranto.

1. Introduction

Over the last decades, there is a growing concern about the effects of the current climate chance on marine ecosystems. Model studies such as used by the IPCC2013, suggest that Mediterranean ecosystems might increasingly experience important changes during this century in response to drier summers, soil moisture loss, heat stress increase and risk of low agricultural productivity (Collins et al., 2013). For accurate model projections, it is extremely important to get insights into detailed information about past climatic and environmental variability and their impact on different ecosystems before the influence of human activities. Marine palynological records are particularly useful to obtain this information since sedimentary well-preserved pollen/spore assemblages reflect vegetation composition, which in turn is strongly influenced by climate change and human activities (Prentice, 1988; Montade et al., 2019 and references therein). However, an adequate reconstruction of past vegetation change needs a clear understanding about the source area of the sedimentary palynomorphs. In turn, this requires detailed information about the transport paths from the source of pollen/spore production to their deposition in the marine realm. A method to obtain insight into these pathways is to study pollen/spore distribution in modern marine sediment samples. Given we know the present-day vegetation, the pollen/spore distribution patterns can allow to establish a source-sink relationship which in turn that can elucidate different modes of transport (e.g. van der Kaars, 2003; Dupont & Wyputta, 2003; Beaudouin et al., 2007; Montade et al., 2011; Yang et al., 2016; Luo et al., 2016a, 2016b, 2018a, 2018b). When these transport modes are known, the study of pollen/spore associations from marine sedimentary archives of pre- and post-industrial times allow to achieve knowledge about past patterns of vegetation while integrating simultaneously the marine climate and environmental signal.

It is well known that pollen/spores are transported into the marine realm by both Aeolian and fluvial processes. Most of pollen and spores found in deep-sea environments are transported by wind from regions with no or small river influence (Heusser & Morley, 1985; Hooghiemstra et al., 1986, 2006). Fluvial transport appears to be the major transport mechanism in riverine, river mouths and delta environments, where sediments have a high pollen and spore concentration and where the assemblages generally reflect an integrated overview on the vegetation of the catchment area in detail (Muller, 1959; Davey & Rogers, 1975; Heusser & Balsam, 1977; Dupont & Wyputta, 2003; Sangiorgi & Donders, 2004; Luo et al., 2016a, 2016b, 2018a, 2018b; Gu et al., 2017; Bouimetarhan et al., 2018). However, studies on modern pollen and spore transport show that transportation pathways and dispersion mechanisms vary between regions and the provenance of pollen and spore associations of every region needs to be studied in detail before adequate reconstructions can be established (e.g. Davey & Rogers, 1975).

In this study, we contribute to this discussion by investigating both fluvial and marine pollen and spores associations and concentrations in the Adriatic Sea and the Gulf of Taranto. In this latter region, exceptionally high quality sedimentary records covering the last few millennia have been retrieved and investigated at a three-year resolution (e.g. Chen et al., 2011; 2013; Goudeau et al., 2014; Zonneveld et al., 2016). These sedimentary archives are located at the most distal end of a river system plume. This plume has its origin in the northern Atlantic Sea, where the Po-river discharges into the northern Adriatic Sea, and it will flow southwards close to the Italian coast due to the compression effect produced by the cyclonic circulation of the water mass. The plume waters extend along the entire north-eastern Italian Peninsula and are additionally spiced by discharge waters from Apennine Rivers on its way south (Bortoluzzi et al., 1984; Degobbis & Gilmartin, 1990). Many rivers drain their waters into the Adriatic Sea, providing also particulate matter in suspension but the share associated to the Po River and northern Apennine rivers is greater than for the rest and their remarkable imprint is still recognizable in the sediments around Gargano (Trincardi et al., 1994; Syvitski & Kettner, 2007). River sediment supply decrease to the south of the Peninsula, being Ofanto River the southest river, whose sediment supply can be of importance in terms of net input (Cattaneo et al., 2003). Ofanto River also represents the southest river sample covered in this study. Additionally, sediments are also transported into the marine realm by two wind systems; the Bora and the Sirocco. The Sirocco is a moist wind blowing from the southeast transporting humid and warm air into the Adriatic region (Artegiani et al., 1993, Cavaleri et al., 1997; Pasaric et al., 2007) and the Bora is a dry, cold and continental wind blowing from the northeast (Artegiani et al., 1993; Orlić et al., 1994; Palinkas et al., 2006).

According to Sangiorgi & Donders (2004), the Po catchment area is the major source of the pollen and spores found in sediments of the northern Adriatic Sea. However, until now no detailed information is present whether this holds for the complete Adriatic Sea mud belt. Pollen and spores that originate in the Po-catchment area might be transported as far as the most distal part of the plume system located in the Gulf of Taranto or, alternatively, are settled in a region closer to the river mouth. Furthermore, no information is available to what extent the pollen and spore assemblages in the surface sediments of the Adriatic Sea and Gulf of Taranto originate in the Po- and Apennine rivers catchment areas or are transported by wind. Different taxa also have variable transport properties depending on settling velocities and preservation potential. In this paper, we aim to obtain insights into these factors by providing information about the pollen and spore content (composition and concentration) of the sediment load of the 7 major rivers draining into the Adriatic Sea and comparing these with the pollen and spore composition of 55 marine surface sediments from the Adriatic Sea and Gulf of Taranto (Figure 4-1).

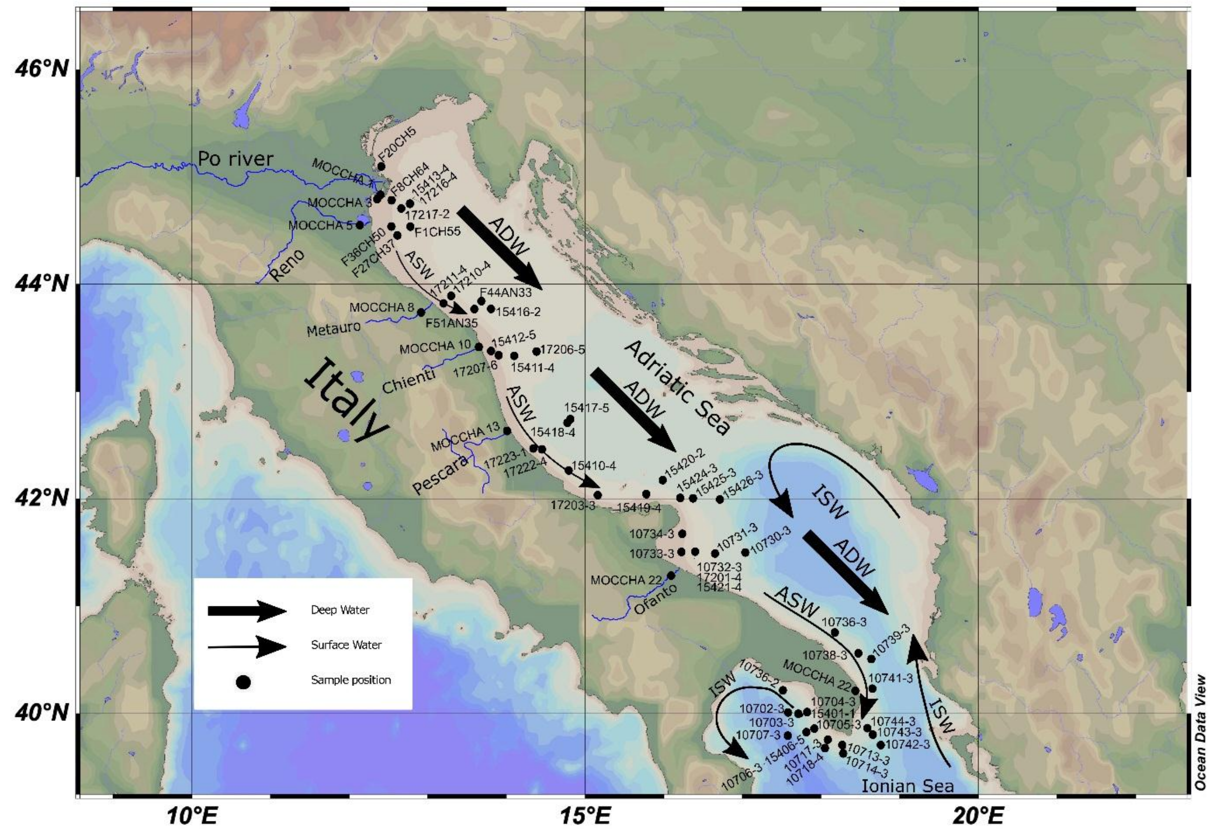

Figure 4-1. Map of the research area depicting sample positions and main upper ocean surface current systems.

1.1. Oceanographic setting

The Adriatic Sea, located between the Italian Peninsula and the Balkans, is one of the sub-basins of the Mediterranean Sea. It is oriented in NW-SE direction, is semi-enclosed and has an elongated shape (Artegiani et al., 1993). It can be divided into three different parts according to the bathymetry (northern, centre, southern), increasing in depth from north to south (Orlić et al., 1992; Zavatarelli et al., 1998; Bensi et al., 2013).

Two major water masses, the Adriatic Surface Waters (ASW) and Ionian Surface Waters (ISW) characterize surface circulation (Figure 4-1). The ASW is formed by relatively cool, suspended matter rich and low salinity waters that are discharged into the basin by the Po River and eastern Italian rivers. Its waters flow along the western margin of the basin due to the cyclonic surface currents induced by Coriolis forcing (Artegiani et al., 1997b; Lee et al., 2007; Colombaroli et al., 2009). The major source of the ASW is formed by discharged waters from the Po River in the northeast of the basin.

The Po River is the largest river in Italy draining the southern and western Italian Alps as well as the northern part of the Apennines. It flows eastward through the so-called Po Valley to drain its water in the North Adriatic Sea through a deltaic system. On its way south, the ASW is additionally supported by discharge waters of rivers that drain the Apennines. The freshwater entering the marine realm is rich in nutrients and suspended matter (Degobbis et al., 1986; Kourafalou, 1999; Boldrin et al., 2005) with the Po River being responsible for around one-third of the riverine input into the Adriatic Sea (Orlić et al., 1992; Kourafalou, 1999). Fresh water discharge and sediment load of the smaller rivers south of the Po River is significantly smaller compared to the Po River discharge (Penna et al., 2004; Milligan & Cattaneo, 2007). It is estimated that both Alpine and Apennine mountain ranges have approximately an equal contribution to the sediment load of the Po River (Nelson, 1970). About half of the sediments transported by the Po River settle on the Po shelf whereas the rest remains in suspension and flows southwards, notably the fine-grained fraction (Palinkas & Nittrouer, 2007). In addition, resuspension of the material occurs (Wang & Pinardi, 2002). A mix of sediments from the Po River and Apennine rivers characterize the sediments south of the Po River pro-delta and north of the Gargano promontory (Cattaneo et al., 2003).

The discharge of the Po and Apennine rivers are strongly influenced by the seasonal variations of wind regimes. The Po River experiences two seasonal increases in discharge of around 2000 m^3/s due to melting of snow and ice in the mountain regions in spring and enhanced rainfall in autumn (Kourafalou, 1999; Boldrin et al., 2005; Syvitski & Kettner, 2007). Bora winds tend to confine and press the Po plume against the coast, meanwhile, Sirocco winds spread the plume eastwards across the northern part of the region (Orlić et al., 1994; Kourafalou, 1999; Palinkas & Nittrouer, 2007). Sirocco winds can occasionally cause flooding events due to the accumulation of water in the shallow North Adriatic (Orlić et al., 1994; Pirazzoli & Tomasin, 2002; Ferrarese et al., 2008; Jeromel et al., 2009). It is more common to have strong Sirocco winds in spring (Sivall, 1957) and Bora winds, more common in winter, participate in the formation of dense and deep water in the northern Adriatic (Hendershott & Rizzoli, 1976, Kourafalou, 1999). This is directly related to the temperature of the water and hence, to the amount of dissolved oxygen (DO), which could alter the preservation of pollen and spores.

The second surface water mass of the Adriatic Sea, the ISW, enters the basin in the south through the Strait of Otranto. This water mass is characterized by high temperatures, high salinity but low suspended matter and nutrient concentration (Artegiani et al., 1997a, b). It spreads northward along the eastern side of the southern Adriatic Sea. The mixing of ASW and ISW terminates outside the basin, within the Gulf of Taranto (Socal et al., 1999; Boldrin et al., 2005; Caroppo et al., 2006; Lee et al., 2007). Intermediate waters that underlie the ASW and ISW are formed by the high salinity, high temperature and relatively eutrophic Levantine Intermediate Water (LIW) that can be observed between 150 – 440 m water depths, both in the Adriatic Sea and the Gulf of Taranto (Nittis & Lascataros, 1999; Sellschopp & Alvarez, 2003). Below this LIW, a transition zone formed by Modified LIW is found, with the particularity of representing an intermediate state between the LIW and the Adriatic Deep Water (Sellshopp & Alvarez, 2003). Below these water masses much cooler and dense waters of the Adriatic Deep Water (ADW) can be found (Sellschopp & Alvarez, 2003; Hainbucher et al., 2006; Rubino & Hainbucher, 2007).

1.2. The Mediterranean climate-driven factors

Due to the combination of three main air circulation systems, the Scandinavian pattern, the East Atlantic pattern and the North Atlantic Oscillation (NAO), the climate in the Mediterranean region is characterized by hot and dry summers and cool wet winters (Hurrell & Van Loon, 1997; Cassou et al., 2004; Fil & Dubus, 2005). In winter, the most influencing climate mode is the NAO reflecting the pressure gradient between the Azores and Iceland; with high and low pressure, respectively (Hurrell, 1995). Due to this dipole-like relationship, changes in the NAO index have a continental effect over the climate, especially on rainfall and temperatures, in Europe including the Mediterranean region and the Adriatic Sea. A positive NAO index brings excess rainfall in northern Europe meanwhile a negative NAO index brings excess rainfall and higher temperatures in southern Europe, including the Mediterranean and the Adriatic Sea (Lionello & Sanna, 2005). For this reason, an increase in rainfall will result in higher discharge not only of the Po River but also of other Italian rivers, increasing the amount of ASW (Chen et al., 2011).

1.3. Vegetation and land management

Due to the orographic characteristics of the Italian Peninsula, the vegetation boundaries are well defined (Figure 4-2). The Po Delta extension is variable; during the first half of the 20th century progradation was slower than in the previous 200 years and the delta reached its maximum extension between the 1930s and 1940s. Despite the continuous supply of sediments from the Po River, the delta has been retreating steadily since then (Syvitski & Kettner, 2007). The Po Delta has been highly altered by humans, by filling most of the wetlands with sediments, reclaiming land for agriculture or changing the river runoff through canals (Cencini, 1998). The vegetation of the Po catchment area and the Adriatic borderland is strongly influenced by altitudinal zonation (Sangiorgi & Donders, 2004; Combourieu-Nebout et al., 2015) and it is a very important region from a phytogeography point of view. The vegetation of Italy constitutes the boundary between Central European and Mediterranean regions, so the plant species here found represent a mixture from both phytogeography regions (Cencini, 1998).

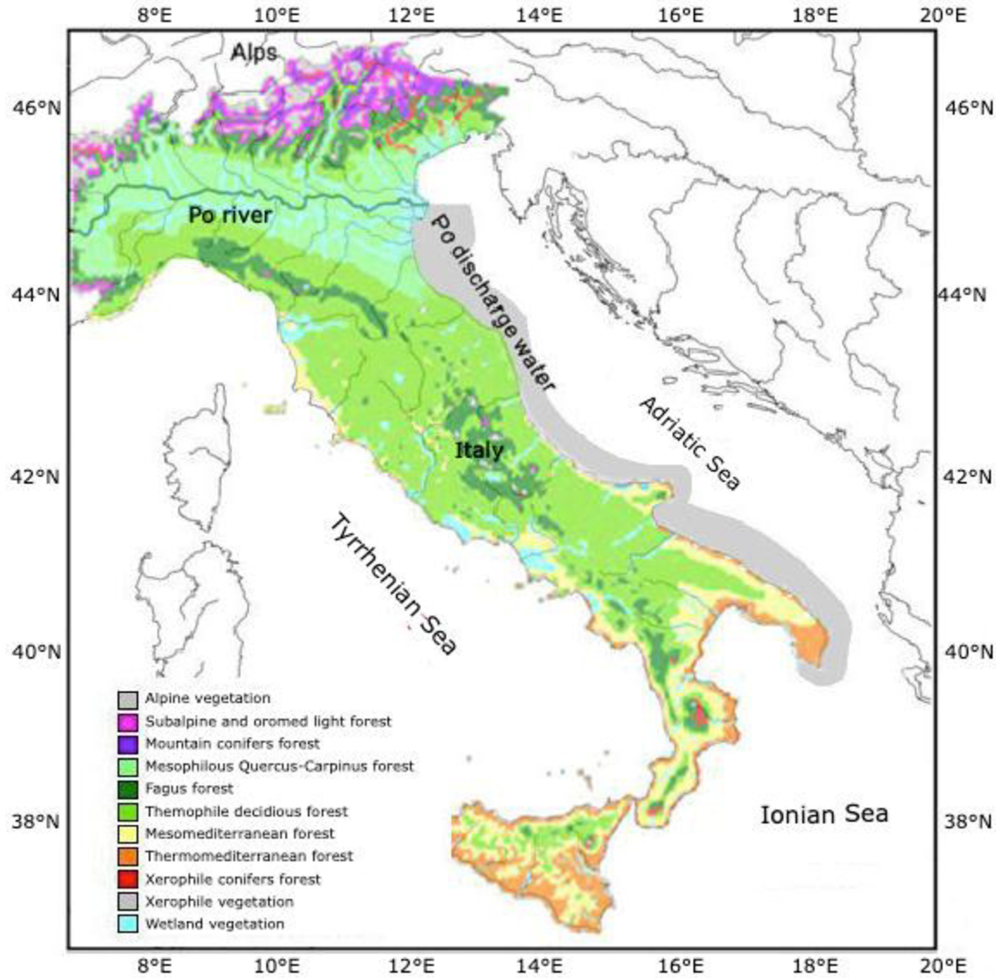

Figure 4-2. Vegetation map of the Italian Peninsula (modified after Pignatti, 2011, originally from Bohn et al., 2000/ 2003).

In the catchment area of the Po River, three vegetation zones can be distinguished; (a) Alpine and Subalpine forest covering the sides of the Alps, (b) Pre-alpine vegetation, temperate forest and (c) Low alpine and Po plain. (ad. A.) In the alpine and subalpine forest, the dominant species are *Picea abies, Abies alba* and *Larix decidua* that establish a coniferous forest (Lang, 1994; Blasi et al., 2010). The distribution of these species is limited to the Alps and the northern part of the Apennines. Species from the genus *Abies*, in particular *Abies alba*, are distributed across the lower Alps and the Apennines. Their distribution is not restricted to the northern part of Italy but can reach the south of the peninsula as well (Parducci et al., 2004; Kehlet et al., 2004). (ad. B.) The Pre-alpine vegetation is characterized by a homogeneous mesophilous forest rich in beech (*Fagus sylvatica*), silver fir (*Abies alba*), *Acer, Salix* and *Ulmus* in combination with open pasturelands and cultivated conifer woods (Pignatti, 1979; Ferrari, 1997; Blasi et al., 2010, 2014). (ad.C.) The low alpine and Po Valley regions are mainly covered by a mesophilous forest rich in *Quercus*, *Carpinus orientalis* and *Carpinus betulus*, *Ostrya*, *Fraxinus excelsior* (Cencini, 1998; Blasi et al., 2014) in combination with an intensive agricultural use of the land which reveals a great human impact on the land. The cultivated land consists mainly of maize, wheat, sugar beet, barley, soybean, rice, and horticulture in general (Giglio, 2006). Typically, Mediterranean species are holm oak (*Quercus ilex* type), southern ash (*Fraxinus angustifolia*) or *Phillyrea angustifolia* (Cencini, 1998). These species and black alder (*Alnus glutinosa*), white poplar (*Populus alba*) and willow (*Salix*), also represent the typical thermophile and mesophile forests, at moist and rich soils and with a distribution covering potentially the whole territory (Pignatti, 1982). *Fraxinus excelsior* and *Betula pendula* are found in the wet forest too but their extension is restricted to the middle of the Apennines and north to the Alps (Pignatti, 1982).

The genus *Pinus* is widespread throughout the Italian Peninsula from mountain to coastal habitats (Lang, 1994). Different species of the genus are commonly used for reforestation (Pignatti, 1982) and protection against soil erosion (Calama et al., 2007). The *"Pinus"* forests are mixed with *Juniperus communis* that can also be observed all around the peninsula, especially in the arid regions (Pignatti, 1982; Blasi et al., 2014). In the Po plain, dunes, recreational areas on the Po Delta, different species of *Pinus* dominate in detriment to the natural vegetation communities (Cencini, 1998). Other common cultivated species are *Corylus avellana*, *Castanea sativa*, and *Olea europaea*. The vegetation in the central part of Italy is strongly linked to the presence of the Apennine mountain range. According to different studies (e.g. Pignatti, 2011; Blasi et al., 2010, 2014; Combourieu-Nebout et al., 2015) the vegetation consists mainly of *Fagus, Corylus* and *Fraxinus* in the higher parts, a homogeneous mesophilous deciduous forest rich in *Quercus robur* type*, Q. cerris, Castanea sativa, Alnus, Fagus* and *Carpinus* mixed with typical Mediterranean forest species such as *Olea europaea, Quercus ilex* type and *Pistacia* in the lower parts, along the flanks and in the coastal domain (Pignatti, 1982).

A sclerophyll forest rich in *Olea europaea*, *Quercus ilex* type, *Ceratonia*, *Thymus*, *Pistacia* and diverse species of *Pinus* characterizes the vegetation of the Mediterranean forest in Italy (Lang, 1994; Blasi et al., 2014). Herbs are also common throughout Italy. Chenopodiaceae, Plantaginaceae, Urticaceae, *Artemisia*, Asteraceae, and Poaceae are found in a great variety of substrates and different land use (Pignatti, 1982). Some are representative of a dry land and human impact (Sangiorgi & Donders, 2004). Other herbaceous taxa like *Typha*, and Cyperaceae have a wide distribution linked to pits, lakeshores, and riverbanks, while *Ephedra* is more common in coastal and dry habitats (Pignatti, 1982).

2. Material and Methods

A set of 7 river sediment samples, 1 lagoon sample and 55 marine surface samples have been collected by push-coring, multicoring and box coring respectively during a field expedition in 2009 and research cruises P339-CAPPUCCINO (2006), P411-CARPACCIO (2011), P488-CAPRICCIO (2013), PRISMA (1998) (Zonneveld et al., 2008, 2013).

The surface sediments (0-1 cm) of both marine and river cores were prepared for palynological analysis using standard laboratory procedures without acetolysis (Faegri and Iversen, 1989). Sediment (1 cm^3) was first treated with diluted HCl (10 %) and then with HF (40 %) to remove carbonates and silicates, respectively. To calculate palynomorph concentrations, one tablet of *Lycopodium* spores (18.583 ± 1.708 spores per tablet) was added to each sample before the chemical treatment (Stockmarr, 1971). A subset of sample has been processed according to the aliquot method described by Marret & Zonneveld (2003), for which a correction factor was calculated. Following the chemical treatment, samples were physically treated using an ultrasonic bath to disaggregate organic matter and then wet sieved over a 10 µm filter mesh. A fraction (50 µL) was embedded in glycerine gelatine, mounted on a glass slide and sealed with paraffin wax. One to four slides per sample were counted using a light microscope with 400-x and 1000-x magnification. Pollen grains were identified according to Moore et al., (1991), Trigo et al. (2008), and Beug (2015) with help of the reference collection of the Paleoecology group, Department of Physical geography at the University of Utrecht (The Netherlands). The genus *Plantago* includes *Plantago lanceolata, Plantago major* and *Plantago maritima*. *Ephedra* includes pollen from *Ephedra fragilis* and *Ephedra distachya*. Oleaceae include *Olea europaea, Fraxinus* and *Phillyrea*. Relative abundances have been calculated both with and without *Pinus* in the pollen sum to avoid a potential Neves-effect (Chaloner & Muir, 1968). Shannon-Wiener index has been calculated to spot sediment core with higher palynomorph (pollen and spores) diversity. Maps showing relative abundances of species (Figures 4-7, 4-8 and Appendix A-1) and the pollen/spore diagram (Figure 4-6) calculated without considering *Pinus* for the calculation.

2.1. Statistical analysis

The multivariate ordination technique Principal Component Analysis was performed using CANOCO v.5. (Ter Braak & Smilauer, 2012). Before the analysis, the linear character of the dataset was tested by executing a Detrended Corresponding Analysis (DCA). Two statistical analyses (PCA) were performed. The first on the relative abundance dataset where *Pinus* was included in the pollen sum. A second analysis was performed on the dataset where *Pinus* was excluded from the pollen sum. Both analysis were conducted with the shown pollen types, whose proportions were previously calculated in relation to the complete dataset. Interpolated distribution and concentration maps were established with Ocean Data View (Schlitzer, 2018) and the interpolation method is DIVA gridding, with an interpolation set to 20 miles for X and Y axes.

3. Results

A total of 64 pollen taxa were recognized in the examined material (Appendix A-3). The number of pollen and spores per site found varies between 37 (GeoB 10726-3) and 614 (MOCCHA 25). The pollen/spore concentrations and relative abundances of the investigated surface samples allow the recognition of an onshore-offshore as well as a north-south trend in the system that are reflected both in pollen concentrations as well as in relative abundance data. Data will be stored on www.pangaea.de.

3.1. Absolute abundance

Pollen/spore (p) concentrations are higher in river samples and marine samples from coastal regions where surface waters are formed by ASW compared to the offshore samples. Lowest concentrations are observed offshore in the central Adriatic Sea. The highest concentrations are observed at the Cevaro river (Moccha 19; 58985 p/g), and lowest concentrations are observed in the central Adriatic open waters (GeoB 10726-3; 357 p/g; Figure. 4-3).

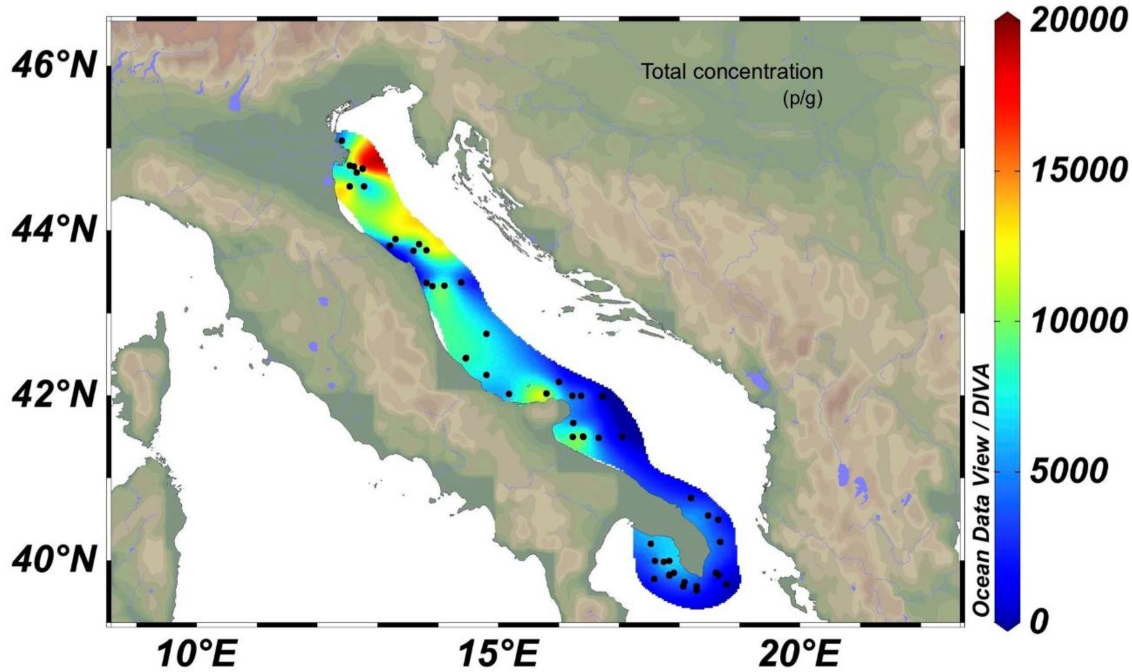

Figure 4-3. Distribution map depicting the concentration (palynomorphs/gram) of the sum of palynomorphs.

Beside an onshore offshore trend, an approximate north-south trend can be observed in marine samples recovered from ASW influenced regions. Concentrations are highest in the northern samples recovered from the Po River mouth region with maximal abundances registered for the samples off the Po River delta (F1CH55; 18175 p/g) and off the Po River mouth (F8CH64; 7583 p/g). Lowest concentrations are observed in the most distal part of Puglia, and "open sea" samples. However, this overall north-south trend is interrupted by areas with enhanced pollen/spore concentrations that correspond to the known sediment depocenters off the Po River mouth (samples F1CH55, GeoB 17216-4, GeoB 17217-2, GeoB 15413-4), off Ancona (samples GeoB 15416-2, GeoB 17210-4, GeoB 17206-

5), in the northern part of Gargano (samples GeoB 15419-4, GeoB 15421-4) and in the Gulf of Taranto (samples GeoB 10703-3, GeoB 10704-3) (e.g. Frignani et al., 2004, 2005; Palinkas & Nittrouer, 2006, 2007; Petrinec et al., 2012).

3.2. Relative abundance data

The pollen/spore associations in both the marine and river-mouth surface samples are dominated by arboreal pollen (Figures 4-5; 4-6). *Pinus* is the most dominant species across the entire studied region, with a minimum percentage of ~6 % in the Po River delta (Sample Moccha 3), and highest relative abundance of 75 % in sample GeoB 10742-5, southern Puglia (Figure 4-4b).

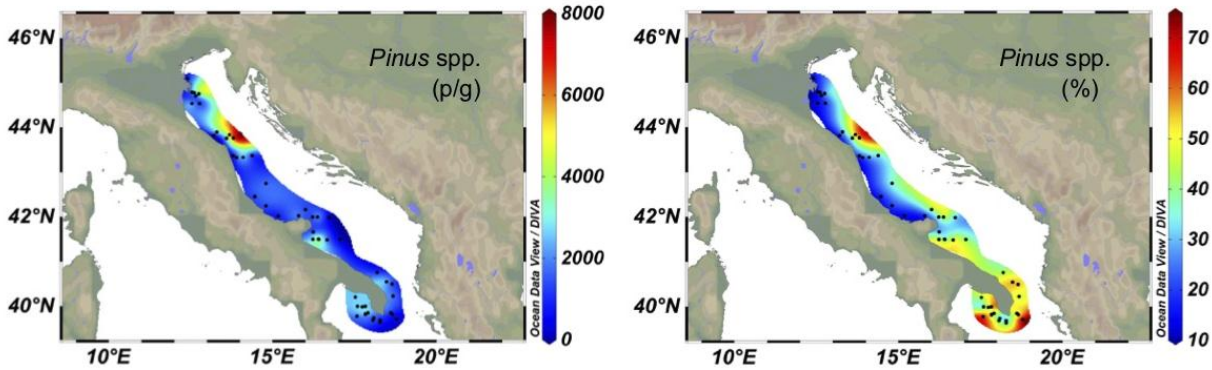

Figure 4-4. Distribution map depicting the distribution of Pinus a) left: Concentrations in pollen/gram. b) right: Relative abundance.

A clear onshore to offshore trend can be observed in the pollen and spore associations. Offshore samples that are recovered from regions where surface waters are formed by Ionian Surface waters (ISW) contain almost exclusively pollen of arboreal taxa notably of Gymnosperms (*Pinus* and to a lesser extend *Abies* and *Picea*) and *Quercus*. When *Pinus* is excluded from the pollen sum, Gymnosperms form about 5 % of the association whereas *Quercus* forms about 22 % (Figure 4-6). Onshore samples recovered from regions influenced by ASW waters contain a rich association of pollen from trees, shrubs and herbs (Figures 4-5, 4-6). This is also reflected in the diversity (Figure 4-5). In the northern part of our assemblage more morphotypes can be found, up to 38, and a higher taxa richness ~3 (Shannon and Wiener) than in the southern samples where the minimum is reached with 6 morphotypes and a value of 0.85 for the Shannon-Wiener diversity index.

A north-south trend can be observed both in the river and marine samples. The northern samples, notably Po River sediments and samples collected off the Po River mouth have high relative proportions of pollen belonging to monocot species (Poaceae including *Poa* and Cerealia, Cyperaceae, *Typha*) further referred to as "monocots". Furthermore, these samples are characterized by relatively high percentages of *Larix,* C*orylus*, *Picea* and Asteraceae (Figure 4-7). The relative abundances of all these species decrease towards the south. This trend of decreasing relative abundances towards the south is most prominent for *Larix* which is not recovered from samples south of Ancona and for Poaceae (*Poa*, Cerealia) that decrease from ~20 % in the north to ~5 % in the south.

In comparison, the southern samples contain high relative abundances of species characteristic for sclerophyll forests such as *Olea europaea*, other Oleaceae, *Pistacia and Quercus ilex* type (Figure 4-8). Maps of relative abundances for other species are presented in Appendix A-1.

The presence of *Alnus* is quite uniform along the assemblage, with a maximum proportion of 15 %.

We observe strong coherences in association composition between the river sediments and marine sediments near the river mouths of these rivers (Figures 4-5, 4-6). This is especially visible in the distribution patterns of major herbs and ferns that occur in higher relative abundances in sediments located near river mouths compared to the other marine samples.

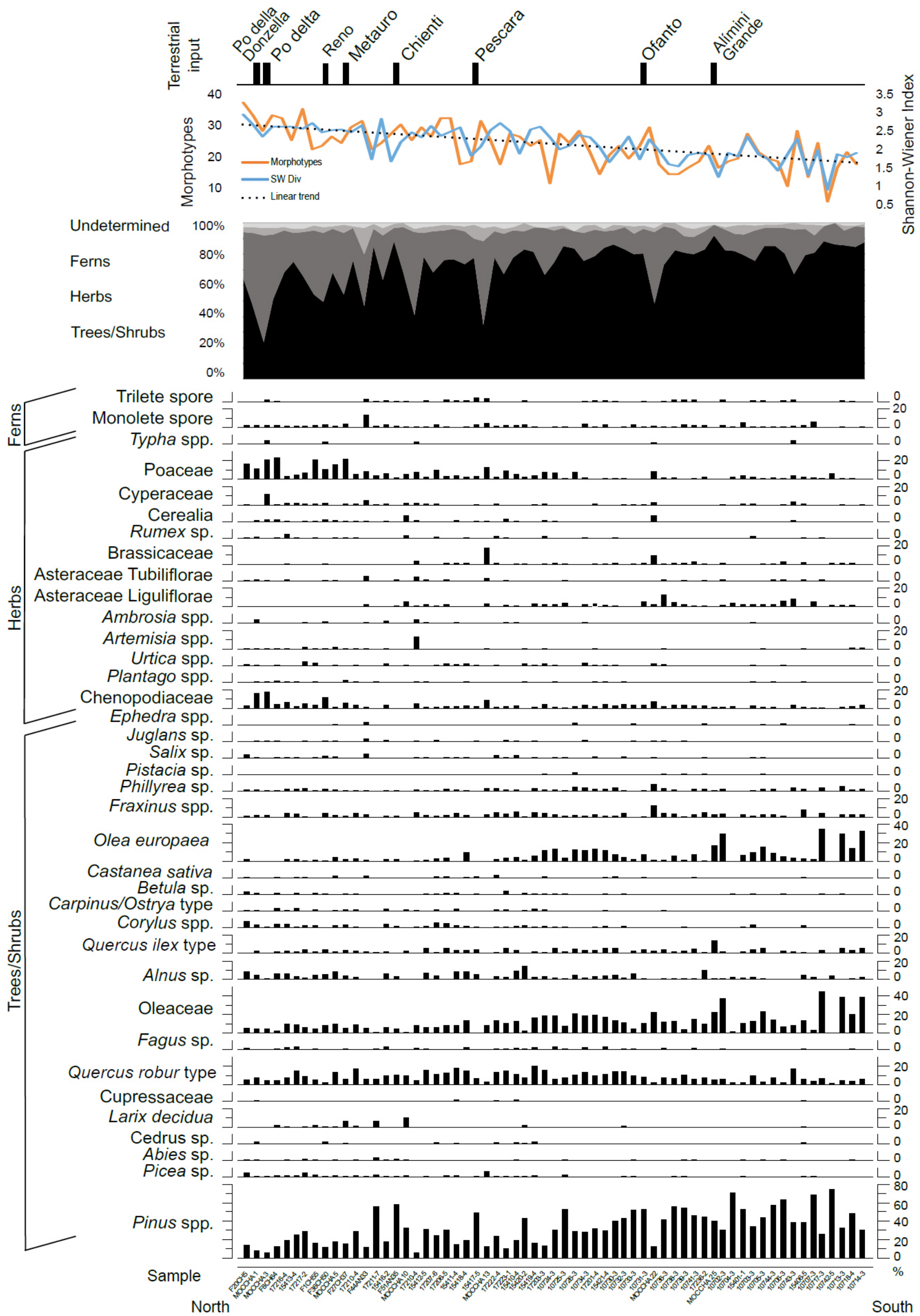

Figure 4-5. Top: Location of investigated terrestrial samples. Shannon-Wiener-Index and morphotypes trend across the studied region. Percentages of the different vegetation groups presented below. Bottom: Relative abundances of major palynomorph types and species diversity with Pinus included in the pollen sum.

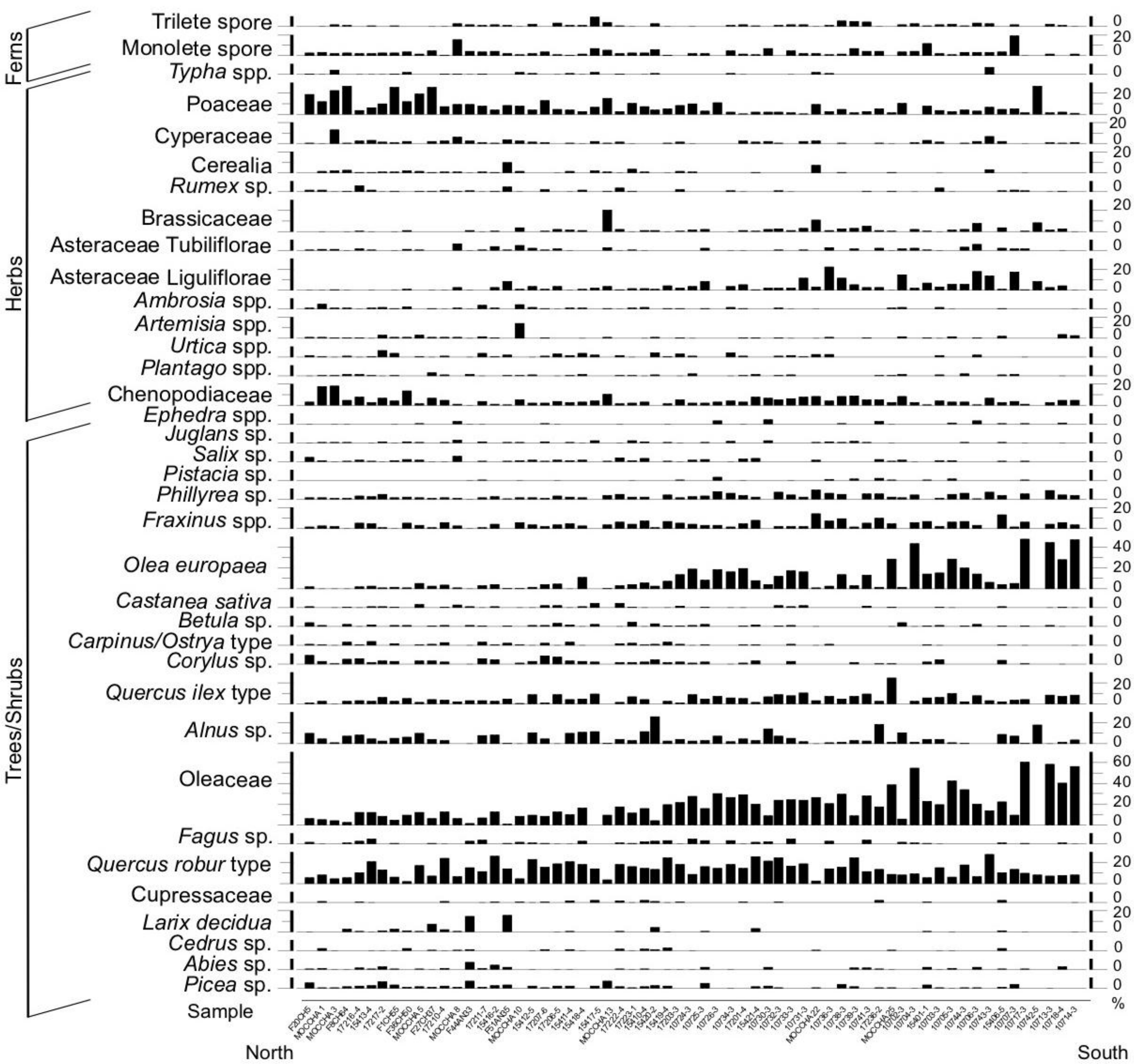

Figure 4-6. Relative abundances of major palynomorph types with Pinus excluded from the pollen sum.

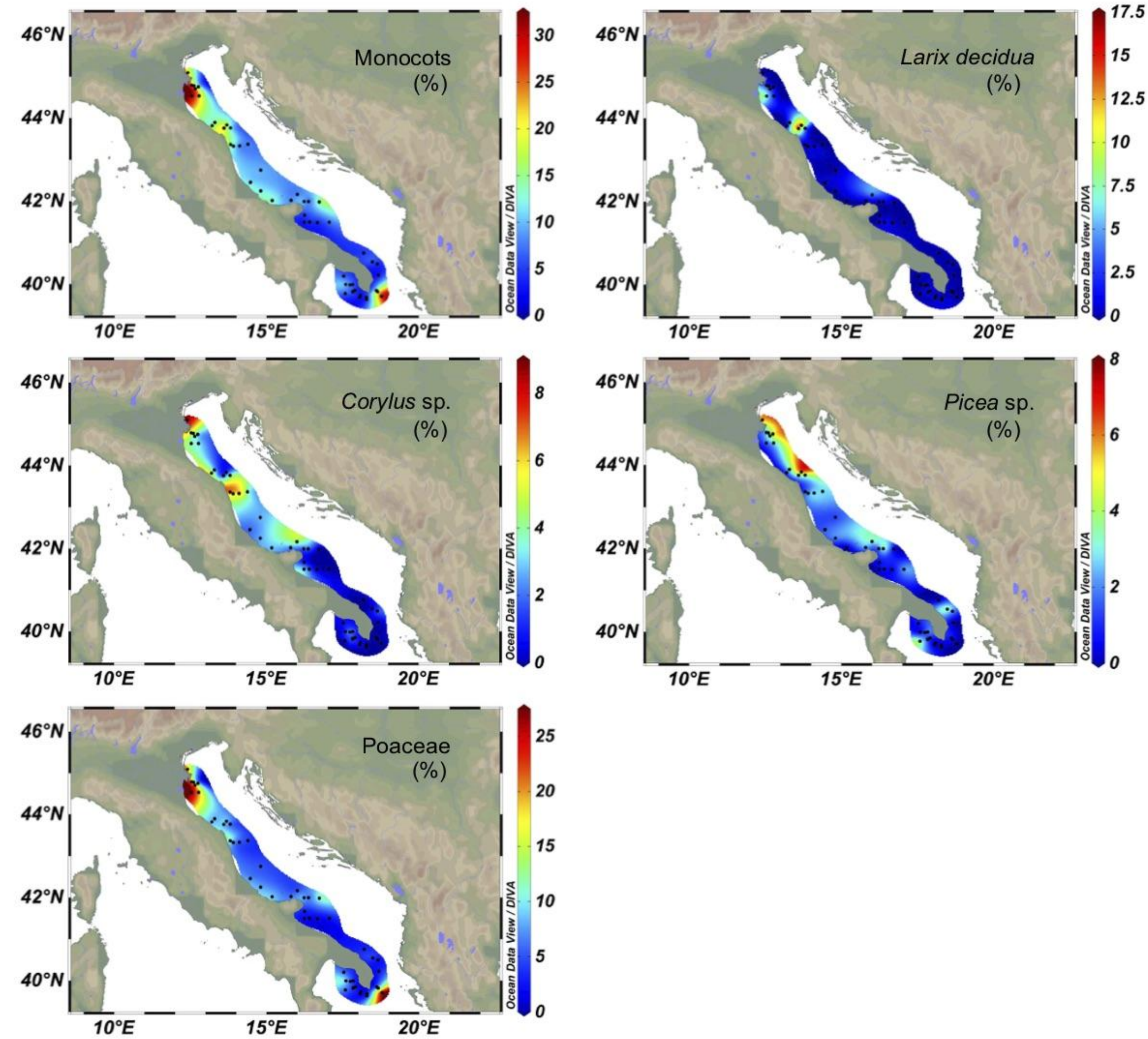

Figure 4-7. Distribution maps of key species characteristic for the "Po River association".

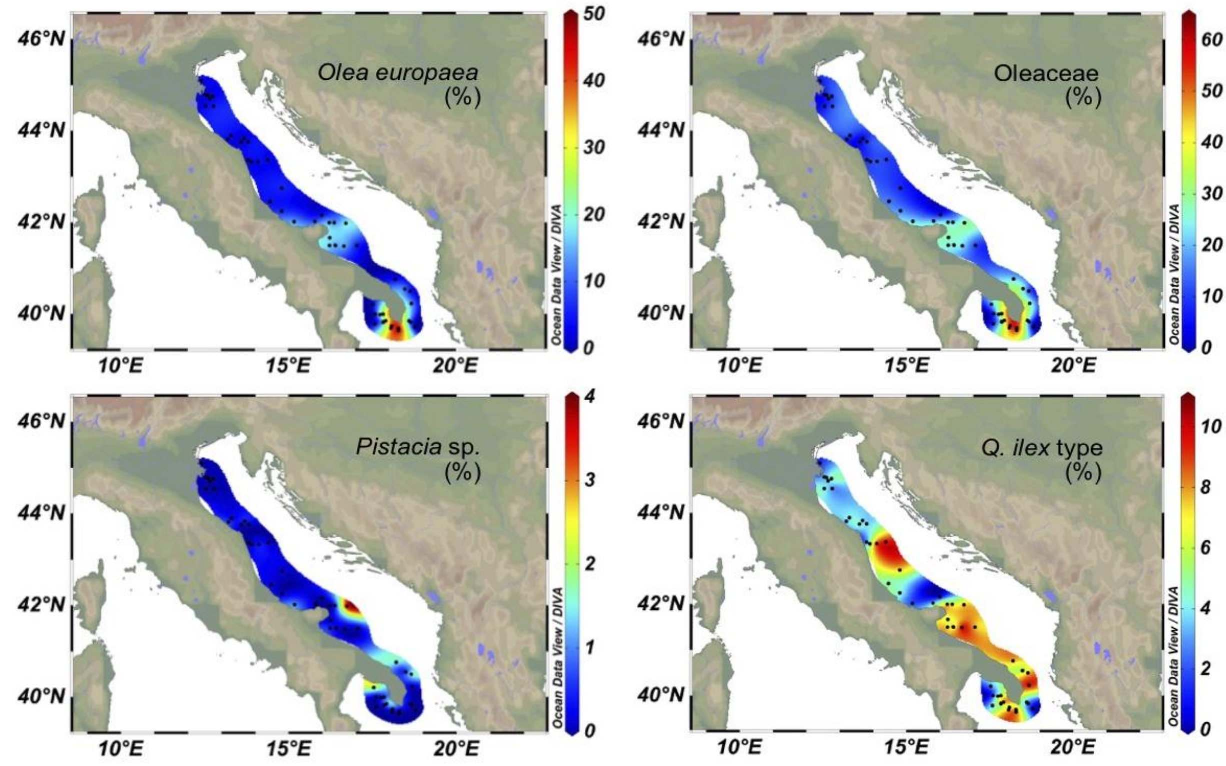

Figure 4-8. Distribution maps of key species characteristic for the "southern association".

3.3. Ordination results

The patterns observed in relative and absolute abundances of pollen/spore assemblages are reflected in the results of the Principal Component Analyses (PCA) as well (Figure 4-9, 4-10).

When *Pinus* is included in the pollen sum, the species and sample ordination is completely dominated by the distribution of this species that is ordinated at the positive side of the first axis (Figure 4-9). All other species are ordinated at the negative side of this axis or near the centre. The first axis covers 56 % of the variance within the dataset. The second axis accounts for 26 % of the variance (Table 4-1).

When *Pinus*, is excluded from the pollen sum the first and second axes cover 23 % and 11 % of the variance in the dataset respectively (Table 4-1, Figure 4-10). Samples ordinated along the first axis show a strong north-south gradient (Figure 4-10b).

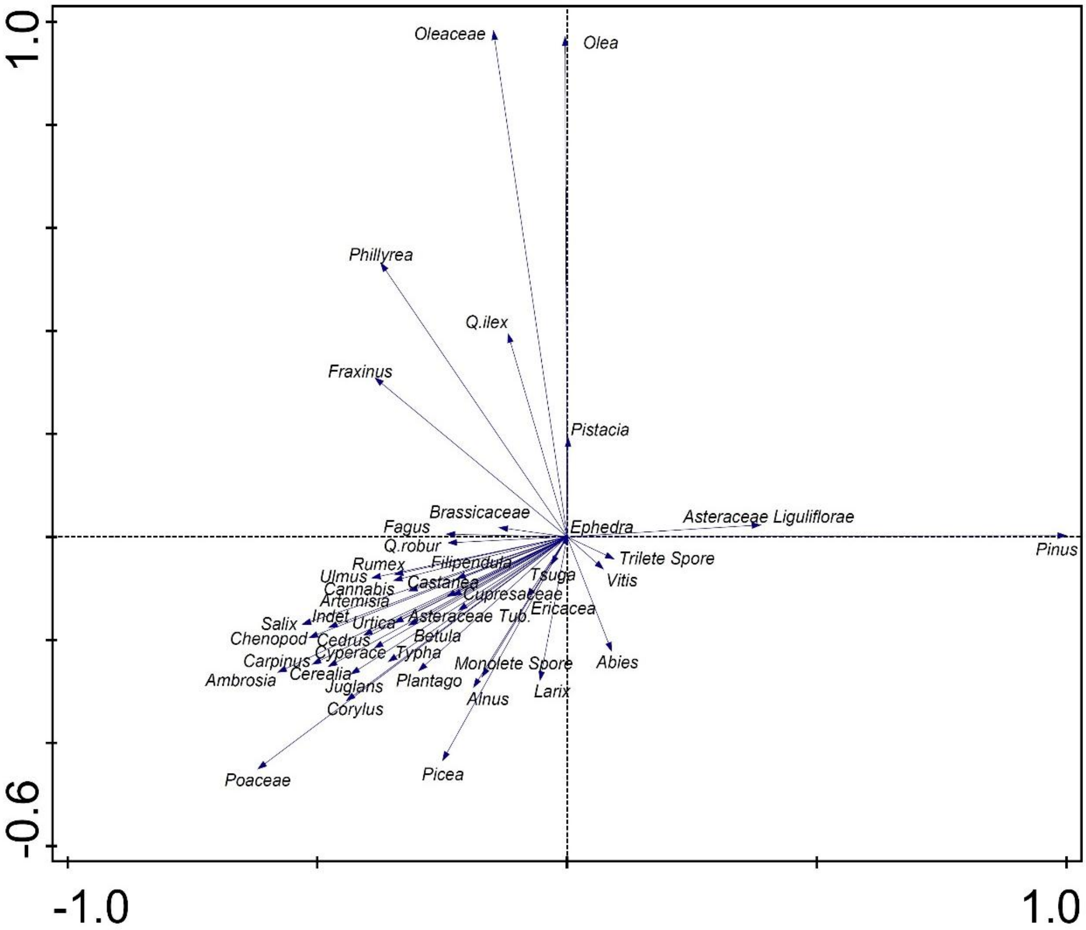

Figure 4.9. Species ordination diagram based on a PCA analysis with Pinus included into the pollen sum.

Table 4-1. Eigenvalues and length of gradient of the first two PCA axis of the two analyses.

Analysis with *Pinus* included, Total variation: 37281.979				
Statistic	**Axis 1**	**Axis 2**	**Axis 3**	**Axis 4**
Eigenvalue	0.5564	0.2626	0.0494	0.0281
Explained variation (cumulative)	55.64	81.9	86.84	89.65
Analysis without *Pinus*, Total variation: 1031.019				
Statistic	**Axis 1**	**Axis 2**	**Axis 3**	**Axis 4**
Eigenvalue	0.2285	0.1144	0.0829	0.068
Explained variation (cumulative)	22.85	34.29	42.58	49.38

Four groups of samples with specific species associations can be identified (see Appendix A-2); for spatial ordination of the groups see Figure 4-11:

A. Po River association. The first group is formed by samples ordinated at the most negative scores on the PCA 1. These samples were recovered from the Po della Donzella and Po River delta as well as in front of the Po River mouth, two samples off Ancona that underlie the most offshore part of the ASW influenced region and one from more central Adriatic Sea waters off Pescara (Figures 4-10, 4-11). Pollen associations in these samples are dominated by Monocots that belong to the Poaceae, Cerealia and *Typha*. Furthermore, samples of this group are characterized by, relative to other samples, high relative abundances of *Picea*, *Larix* and Chenopodiaceae compared to other samples.

B. Central Italian river association. A second group of samples is formed by samples collected from the central Italian rivers Reno, Foglia, Potenza and Vomano. Furthermore, samples collected from the coastal Adriatic Sea regions in front of these rivers characterize this group (Figures 4-10, 4-11). Samples of this group are ordinated at the negative central part of the first PCA axis. Pollen/spore associations of these samples are characterized by high proportion of pollen from trees that are specific for mesophilous forest including *Carpinu*s, *Corylus*, *Fagus* as well as herbs such as *Rumex*, *Plantago* and *Artemisia*.

C. Mixed association. A third group of samples is formed by samples that are characterized by a mixed pollen association that cannot be assigned to one of the other groups (Figures 4-10, 4-11). This group consist of a sample from the river mouth of the Ofanto River, samples collected from ASW influenced regions south of Ancona, onshore samples at the most distal end of the river plume influenced waters and a few more offshore Adriatic Sea and Gulf of Taranto samples. These samples are ordinated at the central part of the first PCA axis. Their pollen/spore association is characterized by relatively high relative abundances of cosmopolitan species, notably from herbs such as those from the Asteraceae, Chenopodiaceae and *Ephedra*.

D. Southern association. The last group is formed by samples that are located in the Alimini Grande and the coastal regions of the Adriatic Sea south of the Gargano Peninsula and in the Gulf of Taranto (Figures 4-10, 4-11). These samples are ordinated at the most positive side of the first PCA axis. These samples are characterized by a pollen/spore assemblage that is typical for Mediterranean sclerophyll forest with high relative abundances of *Olea*, other Oleaceae, *Pistacia*, *Phillyrea* and *Quercus ilex* type. The association of these samples is dominated by pollen belonging to the Oleaceae.

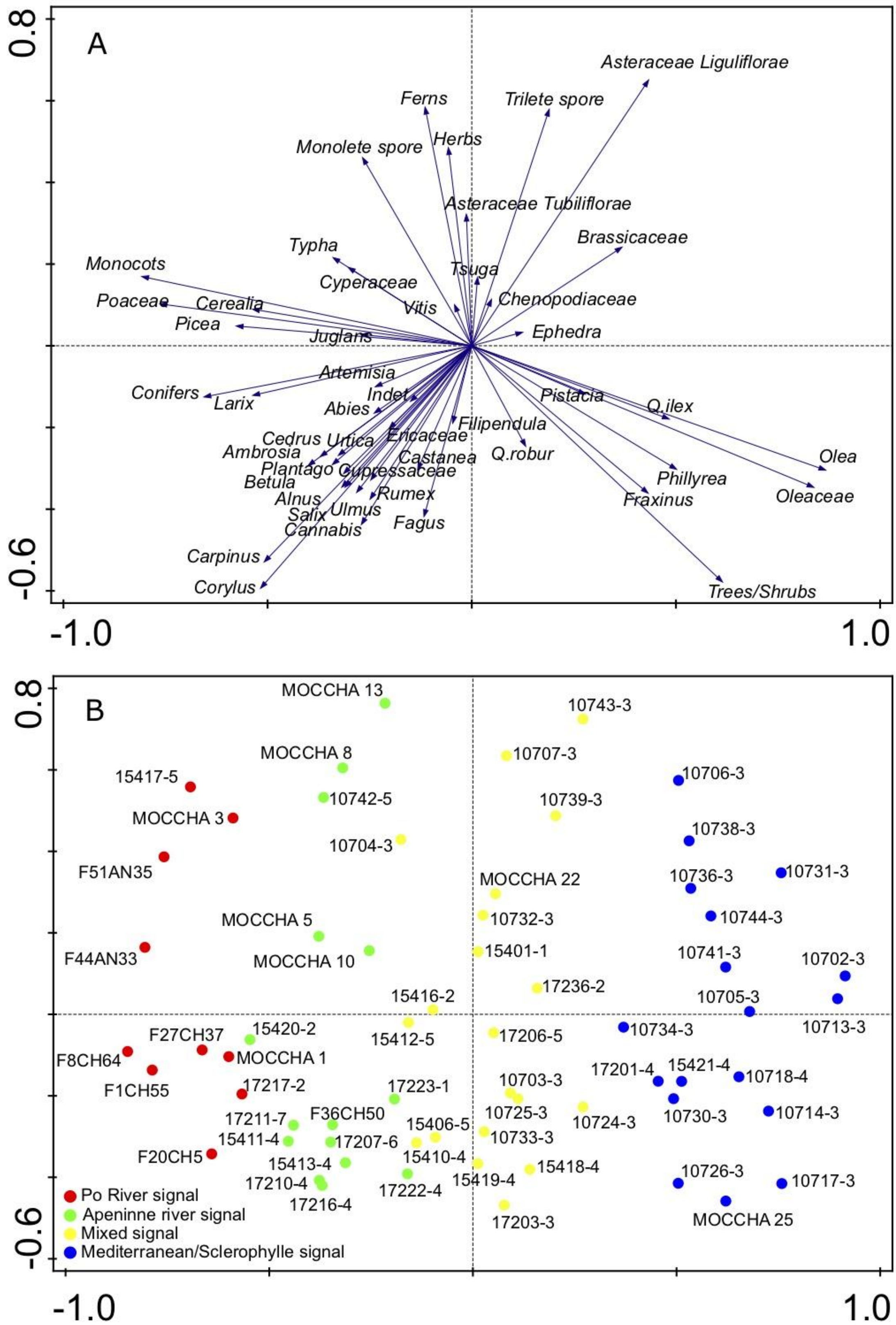

Figure 4-10. Results of the PCA analysis where Pinus being excluded from the pollen sum. a) top: Ordination diagram of species. b) bottom: Ordination diagram of samples. Colours represent different association groups. Red: Po River association, green: central Italian river association, yellow: mixed association, blue: southern association.

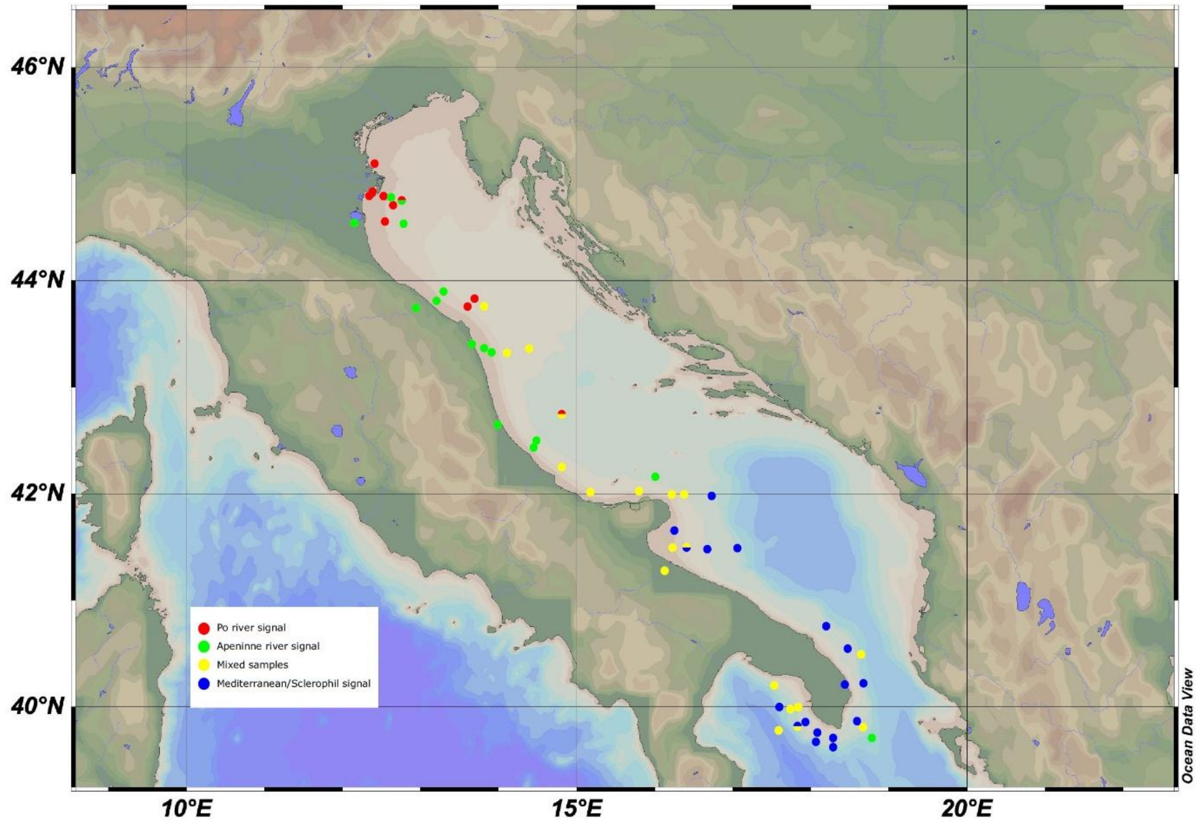

Figure 4-11. Map of the research area depicting sample positions of the four association groups. Red: Po River association, green: central Italian river association, yellow: mixed association, blue: southern association.

4. Discussion

4.1. Overall distribution of pollen/spores in the Adriatic Sea and Gulf of Taranto

Eastern Italian river sediments and marine sediment surface samples show that pollen and spore (p) associations in the Adriatic Sea and Gulf of Taranto region are dominated by arboreal pollen. This is a well-known distortion between pollen concentrations in surface sediment samples and vegetation cover in the source areas (e.g. Beauduoin et al., 2007; Ganne et al., 2016; Lambert et al., 2017, Lou et al., 2018a). This can be generally explained by a higher production of arboreal pollen compared to that of herbaceous plants (Broström et al., 2008; Mazier et al., 2015; Sjögren et al., 2015). Another factor that might have affected the sedimentary pollen spore association is species selective degradation (e.g. Keil & Hedges, 1998; Lebreton et al., 2010; Zhang et al., 2017). Recent studies show that when aerobic degradation affects the sedimentary pollen/spore association, several taxa such as *Artemisia*, Asteraceae, *Pinus*, Poaceae and Chenopodiaceae are less affected than other taxa (Zhang et al., 2017). This result in a biased higher relative abundance of these species when samples are affected by aerobic degradation compared to non-affected samples. Within the research area, bottom water oxygen concentrations co-vary with water depth. Bottom waters of the shallow coastal samples are embedded in ASW water are characterized by relatively low oxygen concentrations whereas the deeper samples are formed by oxygen rich Adriatic Deep Waters. Indeed, we observe in our dataset a clear onshore-offshore trend with the highest abundances of *Pinus* in the most offshore samples that are recovered from a greater water depth. However, if selective degradation would have been the cause of this increase in abundance, we could expect that other resistant taxa such as *Artemisia,* Asteraceae, Poaceae and Chenopodiaceae would have higher concentrations in these samples recovered from greater water depths as well. We however do not observe this in our dataset. Although we observe relatively high relative abundances of Asteraceae and Chenopodiaceae in the more offshore samples compared to the onshore samples, we observe an opposite trend for the Poaceae. Consequently, we assume that selective preservation did not influence the pollen pollen/spore association in the surface sediments of the river and marine surface sediment samples in the research area.

Within the research area, we observe highest concentrations and more different morphotypes of pollen/spores in samples that underlie ASW waters, which can be observed along the Italian coast throughout the Adriatic Sea and the coastal area around the southern tip of Italy. Lowest concentrations and lowest diversity are observed in samples that underlie ISW waters within the central Adriatic Sea as well as the central and northern part of the Gulf of Taranto (Figures 4-3, 4-5). ASW waters are formed by river discharge waters that contain high concentrations of suspended sediments. Since pollen/spores behave similarly to silt particles due to the similar particle size and density (Rossignol, 1961), it can be therefore expected that the majority of the pollen/spores that are observed in the coastal samples are brought into the system by the Italian river systems. This assumption is supported by the observation that there is a striking coherence between position of sites of maximal pollen/spore concentrations and sediment deposition in the region. Maximal concentrations of p/g are observed in sites that are known to be sediment depocenters where maximal sedimentation rates of fluvial sediments have been registered (e.g. Frignani et al., 2004, 2005 and references therein; Palinkas & Nittrouer, 2006, 2007; Petrinec et al., 2012). Furthermore, although the marine samples show a higher diversity, we observed a striking coherence between the pollen/spore association of the river mouth sediments and those of the marine samples in close vicinity.

The sediment samples collected outside the ASW influenced area contain almost exclusively Gymnosperms (notably *Pinus* and to a lesser extend *Abies* and *Picea*) and *Quercus* (Figures 4-4, 4-5). These regions are not influenced by fluvial sedimentary input and it can therefore be expected that pollen input is mainly caused by wind transport. Considering the wind system in the research area, the alluvial transport can occur by so called “local Italian winds” that blow in the region mainly from the west and the two principal wind systems, the Bora and the Sirocco, blowing from the northeast and the southeast, respectively (Artegiani et al., 1993; Orlić et al., 1994; Cavaleri et al.,1997; Kourafalou, 1999; Palinkas & Nittrouer., 2006; Pasaric et al., 2007). Bora winds are likely to bring pollen from the Balkan and the Dalmatian mountains where vegetation is largely dominated by *Pinus, Abies*, *Picea* and *Quercus* (Sostaric, 2005). Although a likely origin, the pollen taxa in the open Adriatic Sea are common on the

Italian side of the Adriatic as well. Furthermore, *Pinus* pollen is known for being transported largely and long distances (Whitehead, 1983*). Abies*, *Betula* and *Quercus* pollen can also be transported by long distances (Rousseau et al., 2008). However, *Picea* pollen are thought to be transported by wind only for short distances (Hicks, 2001). Therefore, it is difficult to assess the source of these pollen types and we cannot address the transport of this pollen either to the Bora or to "local Italian winds". The same holds for the Sirocco. Strong Sirocco winds can bring dust including pollen over the Mediterranean Sea all the way to the Northern border from the African continent under strong Sirocco events (Avila et al., 1997; Prezerakos et al., 2010; Chuvochina et al., 2011). Sahara vegetation is characterized by the presence of Asteraceae or Chenopodiaceae mainly, and the north African coastal vegetation has a typical Mediterranean component with *Olea, Pistacia, Quercus robur* type*, Quercus ilex* type, *Artemisia* or *Pinus halepensis* (You et al., 2016; Abu-Aziza et al., 2017). Although these species do not dominate the pollen association in the open Adriatic Sea samples and northern and central Gulf of Taranto, they are all found in these samples. However, these taxa also grow on the Italian Peninsula. As such, it is extremely difficult to determine what part of the pollen association observed in our samples has been transported by Sirocco winds. What seems to be clear enough is the strong effect that both predominating wind systems (Bora and Sirocco) have over the region, bringing pollen and spores even outside the confined waters of the river plume, which could only reach that far brought by the wind.

Moreover, *Pinus* represents a particular case due to its provenance. It is well known that *Pinus* pollen can be transported over extremely long distances thus no clear source region can be attributed (Whitehead, 1983).

4.2 Geographic distribution of specific pollen/spore association

Based on visual examination of the dataset and statistical analyses (after excluding the *Pinus* in the PCA analysis), four groups of samples with specific pollen/spore association have been identified.

The "Po River association" is observed in samples recovered from the northern part of the research area; sediments from the Po River delta and off the Po River mouth as well as two samples within the most offshore region of ASW influenced waters off Ancona and in the central Adriatic Sea off Pescara (Figures 4-5 to 4-7). These samples have high relative proportions of Pollen monocots (a.o. *Poa*, Cerealia and *Typha*) as well as trees and herbs such *Picea*, *Larix* and Chenopodiaceae. It can be expected that the large majority of the pollen/spores in these samples originate from the Po-catchment and are transported by this river in accordance to previous studies (e.g. Sangiorgi & Donders, 2004). We observe very high concentrations of Poaceae (Poaceae and Cerealia) in the north with a decreasing trend southward. This is also observed for the monocot group. These high relative abundances in these northern samples reflect the intensive cultivation of maize, corn, soybean, rice and other crops in the Po Valley (Giglio, 2006). Moreover, there is an intense modification of the land use in detriment to the natural vegetation with the extensive use of different woody species (Cencini, 1998), what contributes to the already established strong influence of *Pinus* in the assemblage. These samples, with a Po River signal, can be observed in samples that have been recovered from sites that underlie the offshore part of ASW waters off Ancona and in one central Adriatic Sea sample recovered from a more southern location. This suggests that Po-discharge waters are being transported southwards with the more offshore ASW waters

The "Central Italian river association" is observed in samples recovered from the central Italian rivers Reno, Foglia, Potenza and Vomano, as well as marine samples collected in front of the river mouths of these rivers. These samples are rich in pollen from trees and shrubs from mesophilous forest (*Corylus, Quercus robur* type*, Castanea sativa, Alnus, Fagus, Corylus* and *Carpinus)* providing a clear Apennine signal. The Apennines are covered by mesophilous forest (max. 1500 m a.s.l) and a thermophilous forest (max 900 m a.s.l) in the lower flanks, suggesting a dominance input of the central Italian rivers catchment areas in these samples.

The spatial distribution of groups one and two is consistent with earlier findings on the transport of suspended sediment material (Tomadin, 2000). By investigating the distribution and composition of muds in the Adriatic Sea, Tomadin (2000) could clearly distinguish two main fluxes towards the southeast; one so called "Apennine flux" close to the western coast and one parallel "Padane flux" in the more open Sea. Based on the distribution maps of illite and smectite, Tomadin (2000) concluded that the muds transported by the "Apennine flux" had an Apennine origin whereas the "Padane flux" contained muds that had brought into the Adriatic Sea by the Po River.

A third group of samples containing a "mixed association", contains a mixture of pollen and spore association associations of the Po River, central Italian river and Southern associations. Samples of this group are dispersed throughout the region influenced by ASW waters from Ancona to the distal end of the plume in the Gulf of Taranto. These samples have, in comparison to the rest of the assemblage, higher relative abundances of herbaceous pollen; Chenopodiaceae, Brassicaceae, Asteraceae and trilete spores. Within this group, a gradual change of species association can be observed. The northern samples roughly contain an association that shows similarity to the association of group 1; the Po River association whereas more southwards this signal becomes less prominent. Samples of this mixed signal group are mainly observed along the coast north of Gargano as well as the most distal part of the ASW influenced regions in the Gulf of Taranto. This suggests that pollen originating from the Po and Apennine rivers is transported south to the Gulf of Taranto where they still dominate the local associations. These samples contain a mixed signal of both river systems, supported by presence of pollen from plant species which geographic distribution is restricted to northern Italy, such as *Picea*. *Picea* nowadays only grow in the Alps, northern Apennines or in the Balkans. Its presence in higher relative abundances in samples collected along the so-called "Padane flux" influenced region and at the distal end of the ASW influenced region suggest a long way transport by ASW with Po River discharge water being traced all the way down to the Gulf of Taranto. Similar distribution patterns can be observed for pollen of *Betula*, *Ulmus*, *Cedrus*, *Cannabis*, *Filipendula*, and *Rumex*. Although these species not restrictively grow in northern Italy and the Apennines their geographic distribution in the marine sediments studied in this paper suggest that they primarily originate from the Po- and Apennine river catchment areas. These findings are in line with geochemical and sedimentological investigations of the sediments deposited in the Gulf of Taranto (Goudeau et al., 2013 and references therein). Goudeau et al. (2013) concluded based on the element ration's Ca/Ni and Zr/Cr as well as on the bulk organic matter carbon isotopes and the C/N ration, that the provenance for Gallipoli shelf sediments can be for ~80 % attributed to Po River/northern Apennines sources and for ~20 % to southern Italian sources. On the contrary, based on organic geochemical analyses Leider et al. (2013) concluded that plant-wax n-alkanes have a local origin at the land mass surrounding the Gulf of Taranto. However, our results clearly show that part of the pollen found in the sediments has a northern/central Italian source.

In the southern part of the studied region, "Southern association", samples share a general pollen pattern characterized by species that are typical for a Mediterranean sclerophyll forest, which reaches in this region its maximum abundance. In these samples, high percentage of *Olea*, *Quercus ilex* type, *Pistacia*, Oleaceae and *Phillyrea* are observed. In the southern part of Italy *Olea europaea* is widely cultivated for its culinary uses. We therefore suggest that the majority of the pollen/spores recovered from these samples will have a local origin.

4.3. Significance for palaeo-environmental reconstructions

Our observations that the pollen/spore associations that are deposited in offshore regions most probably have undergone Aeolian transport, whereas the associations of surface waters that underlie the river discharge plume show a strong coherence with the content of surface sediments of river mouth in close vicinity reflecting fluvial transport, is in line with palynological provenance studies in other marine environments throughout the world (e.g. Beaudouin et al., 2007; Montade et al., 2011; Luo et al., 2016a, b; Yang et al., 2016; Lambert et al., 2017; Luo et al., 2018a and references therein). All provenance studies show as well, that the palynomorph distribution in marine surface sediments and the rate in which pollen/spores are being transported by wind, river discharge or ocean currents is dependent on the local climate, atmospheric, oceanographic and geomorphologic conditions and current systems. The region dependent provenance of the palynomorph associations observed in this study underlines that it is essential to obtain detailed insight local pollen/spore distribution to be able to establish adequate palaeoenvironmental reconstructions using marine sedimentary archives (e.g. Montade et al., 2019 and references therein). The results of our study also show that it can be expected that different environmental signals can be reconstructed depending on the position of the sediment archives. For instance, when the archive is located in the northern Adriatic Sea of below the more offshore part of the ASW waters (below the so called "Padane flux") it can be expected that the down core association mainly reflect vegetation, climate and environmental changes from the drainage area of the Po River. In case the archive is located in more coastal ASW-influenced regions north of the Gargano Peninsula (below the so called "Apennine flux") it can be expected that the down core association most likely reflects vegetation, environmental and climate change in the drainage areas of the eastern Italian rivers draining the north and eastern Apennines. South of the Gargano Peninsula and in the Gulf of Taranto, the mixture of Po River association and central Italian river associations as well as the southern associations might allow the establishment of detailed reconstructions using end-member modelling (e.g. McGregor et al., 2009; Vriend et al., 2014) or other quantitative methods such as Weighted Averaging Least Squares or Modern Analogue Techniques (e.g. Lopez-Merino et al., 2017; Montade et al., 2019). However, future studies are required that compare reconstructions established with these techniques with historical documented vegetation and climate change in the research area to obtain insight into which of these techniques is most suitable to be used in this research area.

5. Conclusions

The palynomorph content of surface sediments from 7 eastern Italian river mouths and 55 marine sites are dominated by arboreal pollen. In the palynomorph associations, an onshore-offshore and north-south gradient is observed.

Pollen/spore associations that are deposited in offshore regions Palynomorphs that are not influenced by Adriatic Surface Waters have a low species diversity containing almost exclusively Gymnosperms (notably *Pinus* and to a lesser extend *Abies* and *Picea*) and *Quercus*. These regions are not influenced by fluvial sedimentary input and pollen input is expected to have an Aeolian origin. Our data provide no clear evidence which of the predominating wind systems; the Bora, Sirocco or local winds, have transported these palynomorphs into this region.

More coastal sites where surface water masses consist of river discharge waters, the so-called Adriatic Surface Waters (ASW) are characterized by a high diversity. In these samples, a striking coherence can be observed between the pollen/spore association of the river mouth sediments and those of the marine samples in close vicinity, indicating that fluvial transport is the major transport way of the ASW-sample palynomorphs. Based on visual and statistical examination 4 major associations can be characterized in these samples reflecting a north-south gradient:

1.) "Po River association" dominated by Monocot pollen and containing high relative abundances of *Picea*, *Larix* and Chenopodiaceae.

2.) "Central Italian river association" characterized by high proportion of pollen from trees that are specific for mesophilous forest including *Carpinu*s, *Corylus*, *Fagus* as well as herbs such as *Rumex*, *Plantago* and *Artemisia*.

3.) "Mixed association" characterized by relatively high relative abundances of cosmopolitan species such as those from the Asteraceae, Chenopodiaceae and *Ephedra*.

4.) "Southern association" characterized by a Mediterranean sclerophyll forest pollen/spore assemblage with high relative abundances of *Olea*, other Oleaceae, *Pistacia*, *Phillyrea*, and *Quercus ilex* type.

Between the region off Ancona and the Gargano Peninsula, the "Po River association" signal is observed in samples recovered from sites that underlie the most offshore part of ASW waters whereas more coastal ASW samples contain a "central river association". South of the Gargano Peninsula and in the Gulf of Taranto the signals are mixed or formed by a "southern association".

We also show for the first time a long way transport of pollen by ASW with Po River discharge water being traceable all the way down to the Gulf of Taranto. This is concluded based on the presence of *Picea,* nowadays only growing in the Alps. This species is observed in samples collected along the so-called "Padane flux" influenced region and at the distal end of the ASW in the Gulf of Taranto.

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Chapter 5 - Manuscript 2

“Reconstructing Italian vegetation development across the Roman Climate Optimum (50 BC – 186 AD) using pollen, spores and dinoflagellates found in marine sediments from the Gulf of Taranto”

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Abstract

Vegetation, human impact and climatic changes during the Roman Climate Optimum have been reconstructed by using pollen, spores and dinoflagellate cysts (dinocyst) from a well-dated marine sediment core (~3.5 years temporal resolution) retrieved from the Gulf of Taranto (SW Italy) covering the period 50 BC - 186 AD.

During this time period, general vegetation patterns remain rather constant, as does the climate. A major disturbance is seen around the year 79 AD related to the most famous Vesuvius eruption. After this eruption, arboreal pollen decrease notably, particularly *Quercus robur* type, with herbaceous pollen increasing in relative abundance. Remarkable is the appearance of *Pistacia* at the upper end of the core section compared to scattered appearances before 767.5 mm. *Olea europaea* or *Castanea sativa* were not extensively used and cultivated in the region, which is reflected in low abundances in general, if present. The presence of *Picea,* a typically alpine pollen type, supports the hypothesis that pollen found in the more southern sediments are transported suspended in marine waters tracing back to the Po River. The combined analysis between pollen and dinocysts shows a striking correlation for both trends along the studied section.

1. Introduction

Within the scope of current climate change, there is a debate about to what extent this change is influenced by natural steering factors and/ or by anthropogenic activities. The Quaternary, and more specifically the Holocene, is characterized by constant climatic changes in short periods of time, with no fixed patterns and duration but usually on a millennial-scale as many different proxies show (Guiot, 1987; Adams et al., 1999; Bradley, 1999, Ljungqvist et al., 2019). However, besides anthropogenic activities influencing the climate, natural processes play an important part in the global climate as well (Stott et al., 2000; Carslaw et al., 2002). Society at large, as well as the scientific community, argue about the degree of human-induced influence on present climate change compared to the ones occurred in history. A proven method to investigate this is to study environmental changes of pre-industrial times that are preserved in sedimentary archives and compare these to changes of industrial times. Changes reflected in such pre-industrial archives provide hereby information about how the environment varies as a response to natural forcing. The topic of interest presently is inferring the trend in climate development, and the media can help to spread the interest and knowledge about it (Eurobarometer, 2011; Anderson, 2017). These interests and concerns established within the population are responses to episodes with extreme weather phenomena and the constant rise of global temperatures and what future scenarios humanity could have to face (IPCC, 2013; Collins et al., 2013). The fact that the net contribution to climate change caused by natural forcing is not known yet in combination with the uncertainty about the reason of the current temperature rise sets both groups apart. The human impact in this playset is of major importance to evaluate future scenarios, bearing in mind that many of these changes occurred on a very short time scale, when compared to a human lifespan (Adams et al., 1999). What remains unclear is to what extent climate changes recorded in pre-industrial archives reflect less disturbed conditions or are influenced by pre-industrial anthropogenic activities. Of special interest are times in which humans were expanding their influence on the natural world accompanied by a rise in the population and a change of the landscape like seen in the Roman Empire epoch.

In the history of the Earth, many periods are described in which temperature was higher than at present. Nevertheless, to date, the debate centres in knowing if there were climatic perturbations in the late Holocene that can be compared to the temperature perturbation experienced after the Industrial Revolution and what extent the causes and the effects had. Furthermore, there is an inconsistency in global temperatures over the preindustrial Common Era (Neukom et al., 2019), which makes it even more important to know about the specific conditions of the studied region.

According to historical records, current climate conditions could have been similar during the so called "Roman Warm Period" (200 yr. BC – 400 yr. AD) (Lamb, 1977). During this period, also named as the "Roman Classical Period" due to the expansion of the Roman Empire, Roman culture was widely spread. Unfortunately, the exact climate conditions are not established as well as if the "Roman Warm Period" was similar, warmer or cooler than today cannot be precisely defined (Bianchi & McCave, 1999; deMenocal et al., 2000; Frisia et al., 2005; Giraudi, 2009; Taricco et al., 2009). Moreover, the forcing mechanisms suggested to steer the short-term climate fluctuations for this time interval do not provide a concrete answer. Oceanic and atmospheric circulations in the Gulf of Taranto and the Adriatic Sea are well described, and riverine input into the basin by the Po River has been extensively studied. The particular feature of this study area lays in the excellent historical record available since the Roman times, facilitating to a significant extent the use and calibration of different proxy-based methods to discern between natural and human-induced climate change (e.g. Milligan & Cattaneo, 2007; Zanchettin et al., 2008; Camuffo et al., 2012; 2013; 2014; Harper, 2017).

To obtain this information, natural archives that contain high temporal resolution records of both climatic and anthropogenic changes are needed. Such archives are worldwide extremely rare (Dupont, 1999). One of these archives is located in the Gulf of Taranto (SE Italy), at the distal end of the Po River discharge plume (e.g. Zonneveld et al., 2009, 2012, 2016; Chen et al., 2011, 2013; Goudeau et al., 2014). These sediments, which are characterized by high sedimentation rates, low bioturbation, absence of hiatus and the presence of ash layers related to Vesuvius eruptions. provide approximately 3.26 year resolution and so enable the detection of paleoenvironmental changes according to linear interpolation of the radiocarbon dates (See Castagnoli et al., 2002; Goudeau et al., 2014; Zonneveld et al., 2016).

For the Gulf of Taranto, high temporal resolution climate records are available covering the transition from the warm climatic conditions in Italy during the Roman Classical Period towards a cooling that started around 125 AD (Chen et al., 2011; Goudeau et al., 2014)

Here we aim to contribute to the discussion to what extent the pre-industrial human activity influenced the environment leading to a change in the vegetation by investigating the effect on the vegetation caused by the Romans previous and during the cooling phase. Hereby, we hypothesize that if Roman activity had influenced and changed the vegetation, this forced distortion must have been recorded in the sediments. The hypothesis is tested by studying pollen and spores development across the Roman Climate Optimum. To avoid dating inconsistencies, we use the same sample set as studied by Chen et al. (2011), which allows us to obtain a direct land-sea correlation. By combining this information and the findings of Ruiz Soto et al. (to be resubmitted; See Chapter 4.) about present palynomorph distribution in the Adriatic Sea and the Gulf of Taranto we will provide the terrestrial input for this time period, leading to the finding of a marine archive showing mainly less disturbed conditions during this time period.

2. Research area

2.1. Ocean circulation

Water mass characteristics from the Gulf of Taranto are influenced by the ocean circulation in the Adriatic Sea (Figure 5-1). Surface circulation in the Adriatic Sea is dominated mainly by two water masses, the Adriatic Surface Waters (ASW) and the Ionian Sea Surface Waters (ISW). The ASW have their origin in the river discharge from the Po River and other Italian rivers which drain the Apennines, supplying cool waters, that are low in salinity and high in suspended matter content into the system (Degobbis et al., 1986; Boldring et al., 2005). Although many rivers drain water from the Apennines into the Adriatic Sea, the Po River contributes the most to the ASW, whose discharge water imprint can still be detected in the most distal part of the discharge plume in the Gulf of Taranto. Moreover, the amount of nutrients and suspended matter supplied by the Po River is also higher in comparison to the rest of the rivers draining the Italian Peninsula to the east (Degobbis et al., 1986; Milligan & Cattaneo, 2007). These discharged waters flow southward and are pressed against the western margin of the basin due to the cyclonic surface currents induced by the Coriolis force (Artegiani et al., 1997b; Lee et al., 2007; Colombaroli et al., 2009). ISW shows high temperature and salinity but a relatively low concentration of nutrients and suspended matter (Socal et al., 1999; Boldrin et al., 2005; Caroppo et al., 2006). Besides ASW and ISW, it is possible to find another distinctive water mass, the Adriatic Deep Water (ADW) with cool and dense water in depths greater than 600 m (Sellschopp & Alvarez, 2003; Hainbucher et al., 2006; Rubino & Hainbucher, 2007). In the Gulf of Taranto, Levantine Intermediate Water (LIW) characterized by high salinity, temperature and relatively high nutrient concentration is located above the ADW, between 150 - 440 m water depths (Nittis & Lascataros, 1999; Sellschopp & Alvarez, 2003). Between LIW and ADW the transition zone called Modified LIW occurs, which has characteristics from both water masses (Sellschopp & Alvarez, 2003). Like in the Adriatic Sea, ocean circulation in the Gulf of Taranto follows a cyclonic pattern. ASW mixes gradually with Ionian Sea Waters (ISW) in the Adriatic Sea, but the total mixing happens within the Gulf of Taranto (Artegiani et al., 1997a,b; Socal et al., 1999; Boldrin et al., 2005; Caroppo et al., 2006).

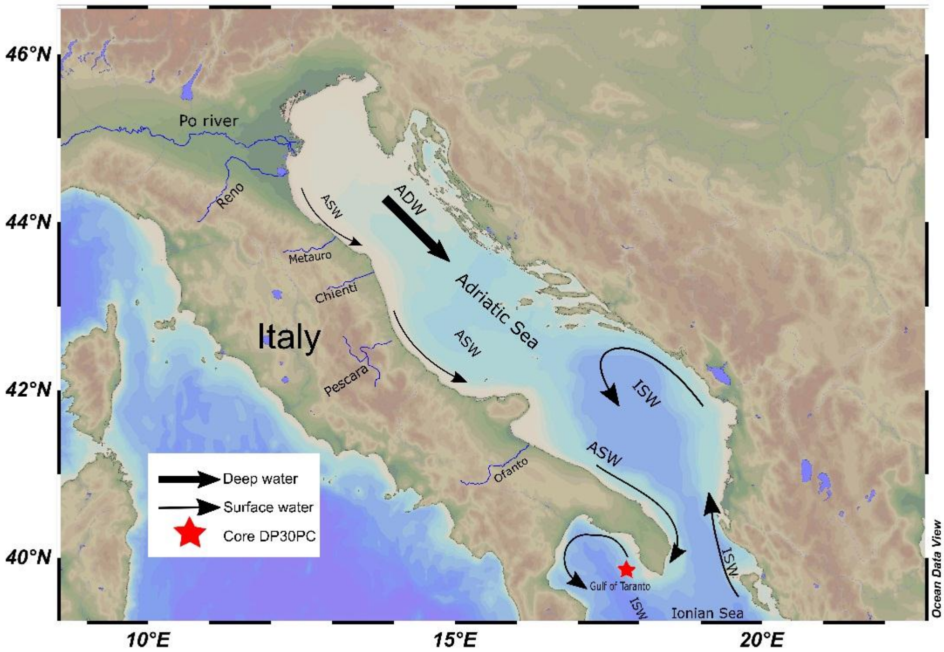

Figure 5-1: Map of the research area depicting core location and main upper ocean surface and deep water current systems.

2.2. Climate of the region

The climate in the Mediterranean region is mainly controlled by a dipole between hot and dry summers but cool and wet winters. This fine adjustment is regulated by the combination of three main air circulation systems, the Scandinavian pattern, the East Atlantic (EA) pattern and the North Atlantic Oscillation (NAO) (Hurrell & van Loon, 1997; Cassou et al., 2004; Fil & Dubus, 2005). The relationship among these atmospheric systems establishes clear characteristics and differences between winter and summer. The NAO is the air system with the greater influence in winter, exhibiting the difference between the high-pressure zone in the Azores and the low-pressure zone in Iceland (Hurrell, 1995). Rainfall and temperatures all across Europe and the Mediterranean Sea as well as in the Adriatic Sea are under strict control of the NAO. When the difference between the high and low pressure zones is strong, the region is under a positive NAO and the northern regions of Europe are affected by excess of rainfall in winter. When the difference between the two pressure systems is weaker, a negative NAO index dominates the region resulting in an increase in rainfall and temperatures in the southern regions of Europe in winter. This affects both the Mediterranean Sea and the Adriatic Sea (Lionello & Sanna, 2005). As a result of changes in the NAO index, the precipitation can be focused either in the north or in the south of Europe. When rainfall is concentrated in the southern regions due to a negative NAO Index, rivers including the Po River and the rest of the Italian rivers discharge more water in the Adriatic Sea. This phenomenon increases the amount of ASW, forcing the plume southwards down to the Gulf of Taranto (Chen et al., 2011).

Regionally, the atmospheric circulation is additionally controlled by two main wind systems, the Bora and the Sirocco (Kourafalou, 1999; Artegiani et al., 1993). The Bora, blowing from the northeast of Europe, is characterized by a dry, cold and continental wind (Artegiani et al., 1993; Orlić et al., 1994; Palinkas & Nittrouer, 2006). Its importance is due to the capacity to induce evaporation and heat loss of the water mass in the Adriatic Sea (Artegiani et al., 1993). In combination with the Coriolis force (Lee et al., 2007), the formation of deep water in the northern part of the basin is enhanced (Hendershott & Rizzoli, 1976). The Sirocco, blowing from the southeast, acts as a good carrier of humid and warm air from the Mediterranean Sea and the northeastern territory from Africa into the Adriatic region (Artegiani et al., 1993, Cavaleri et al., 1997; Pasaric et al., 2007). Both wind systems can take place at anytime without a seasonal preference with the exception that strong Sirocco winds are more frequent in spring (Sivall, 1957). Although they do not have an extensive duration, Bora and Sirocco winds play an important role on the Adriatic Sea level and water masses circulation but with different results (Orlić et al., 1994).

The effects of Bora winds are in general that Po River discharge waters are pressed against the coast. It also contributes, in the northern Adriatic Sea, to the formation of dense and deep water (Hendershott and Rizzoli, 1976; Kourafalou, 1999). Sirocco winds tend to spread the discharge waters eastwards across the northern part of the basin (Orlić et al., 1994; Kourafalou, 1999; Palinkas & Nittrouer, 2006). Moreover, flooding events can occasionally take place in the northern part of the basin due to the accumulation caused by Sirocco winds (Orlić et al., 1994; Pirazzoli & Tomasin, 2002; Ferrarese et al., 2008; Jeromel et al., 2009). The formation and changes of water masses, with the alteration of the characteristics of the water masses like density and temperature, control the amount of dissolved oxygen (DO), which can potentially alter the preservation of pollen and spores (Versteegh & Zonneveld, 2002).

3. Materials and methods

Section 8 (725.0-910.0 mm) of the piston core DP30PC retrieved during the RV Pelagia DOPPIO cruise in 2008 (39°50'07"N, 17°48'05" E, water depth 270 m, 23 km offshore (Figure 5-1)) has been studied (de Lange, 2009).

Samples were prepared for palynological analysis based on standard palynological preparation procedures according to the aliquot method as described by Zonneveld et al. (2009) and subsampled at every 2.5 mm (~3,5 years temporal resolution) (more core details in Chen et al., 2011). The identification of the different pollen types was conducted following Moore et al. (1991), Trigo et al. (2008), Beug (2015) in combination with the reference collection of the Department of Palynology and Paleoecology at the University of Utrecht (The Netherlands).

Palynomorphs percentages were plotted using C2 Version 1.5 (Juggins, 2007). Relative abundances have been calculated by dividing the number of palynomorphs of a particular species by the total sum of observed palynomorphs (including pollen and spores). In the plotted diagram, only the pollen taxa with climatic or anthropological importance are presented. The shown data, in Figure 5-2, represents the relative abundances of pollen and spores. The analysis was performed under two different conditions, with and without including *Pinus* pollen in the pollen sum due to its consistent high proportion across almost the entire assemblage. In Figures 5-3 and 5-4, relative abundances, with and without including *Pinus*, are displayed. Rarefaction analysis was performed with Past3 (Hammer et al., 2001).

Dinocyst data used to spot river run-off episodes and the general trend in freshwater input is used from the published dataset by Chen et al. (2011).

3.1 Age model

The studied core interval of piston core DP30PC is dated by three ^{14}C-datings calibrated as described in detail in Goudeau et al. (2014) (Table 5-1).

Table 5-1. Age model of core DP30PC as calibrated by ^{14}C-dating (Goudeau et al., 2014).

Core depth (mm)	**Radiocarbon age (^{14}C age BP)**	**Calibrated age (cal. yr. BP)**	**Calibrated age (cal. yr.)**	**Error (95 %; ± yr.)**
789	1290 ± 25	755	1195	135
1792	2535 ± 30	2140	-190	120
2794	3830 ± 40	3635	-1685	195

The age model was complemented by tephra analysis. The percentage of glass shards content in non-calcareous mineral particles of the fraction between 100 – 20 µm was analysed in each sample. Beforehand, the samples were treated with 10 % HCl to remove carbonates. In order to remove particles bigger than 100 µm, the material was sieved over a high precision metal sieve. Successively, material was sieved using a 20 µm high precision sieve. The residual material was placed on a slide, embedded in glycerine gelatine and sealed with paraffin wax. The relative amount of volcanic glass particles relative to other particles was determined by polarized light microscopy.

Several volcanic eruptions of Vesuvius and Monte Pilato are known to provide an imprint of volcanic ash deposits in marine sediments of the southern Tyrrhenian Sea and central Mediterranean Sea (Figure 5-4; Table 5-2. e.g. Rolandi et al., 1998; Castagnoli et al., 2002; Taricco et al., 2008; Wulf et al., 2008; Albert et al., 2012) but also in higher ^{14}C content in tree rings in the Alps (Scafetta, 2013; Sigl et al., 2015).

Based on a linear regression between the core depth of samples with enhanced volcanic glass particles and the age of known volcanic eruptions, a sedimentation rate of 0.7637 mm/yr. was calculated ($R^2 = 0.9999$). Due to the high similarity (± 0.75 years difference with a correlation coefficient 1 and *p*-value for T-test of 0.96 x 10-10) between this age model and the one proposed by Chen et al. (2011) combined with the linear compound of the age model, further extrapolation between depths 840.0 mm and 910.0 mm was calculated.

Table 5-2. Peaks in pyroxenes concentrations and the associated volcano eruption.

Enhanced pyroxene concentrations (core depth (mm))	Eruptive event (yr. AD)	Volcano	Reference
1064	774-776	Monte Pilato	Albert et al., 2012; Sigl et al., 2015
1134	685	Vesuvius	Rolandi et al., 1998
1244	540	unknown tropical eruption reflected in Arctic and Antarctic ice cores	Sigl et al., 2015
1251	536	unknown tropical eruption	Sigl et al., 2015
1299	472	Vesuvius (Pollena)	Wulf et al., 2008
1599	79	Vesuvius (Pompeii)	Wulf et al., 2008

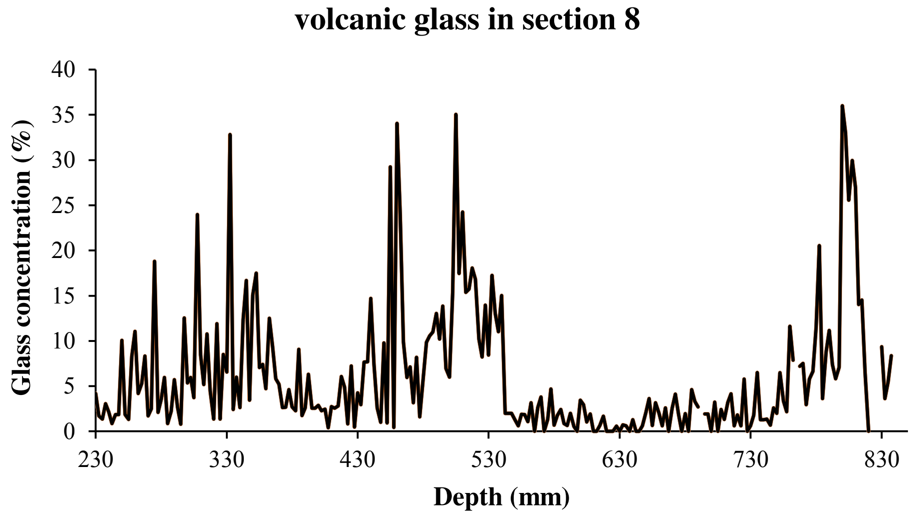

Figure 5-2: Concentration of volcanic glasses (%) in section 8 of piston core DP30PC.

4. Results

A total of 49 morphotypes were identified in the samples (Appendix A-3). The number of palynomorph per sample found range between 100 at 815.0 mm depth and 379 at 820.0 mm depth. *Pinus* shows, in general, a high proportion all across the entire core. Pollen analysis, with *Pinus* excluded from the pollen sum, reveals a vegetation pattern in which the different vegetation zones can be identified (Figure 5-4). For palynomorph flux of the studied section see Appendix A-4. The number of morphotypes ranges between 13 at 802.5 mm depth (~89 yr. AD) and 29 at 760.0 mm (~144 yr. AD). Rarefaction analysis shows values between 11 at 777.5 mm depth (~121 yr. AD) and 21 at 762.5 mm depth (~141 yr. AD). Data will be stored on www.pangaea.de.

4.1. Absolute abundance data

Total pollen (including *Pinus*) and spores concentration as well as accumulation rate values vary greatly along the studied section, ranging between 3078.1 and 297.9 palynomorphs/ gram (p/g) with an average of 1029.2 p/g. Between the depths 835.5 and 815.0 mm (46.53 and 78.58 yr. AD respectively) pollen concentrations are higher compared to the periods previous and after this time interval.

4.2. Relative abundance data

Pinus is the most common species across the entire section and is present in all the analysed samples. It reaches its highest proportion of 69 % at 775.5 mm (121.4 yr. AD) and a minimum of 38 %, with an average proportion of 53 %. The second most common species is *Quercus robur* type. When *Pinus* is excluded from the pollen sum, *Quercus robur* type represents on average 29 % of the assemblage with a maximum percentage of 51 % at 812.5 mm (75 yr. AD). *Quercus ilex* type shows an average abundance of 11 % with a maximum of 20 % at 895.0 mm (31.6 year BC). It is the last species from the group of trees with an average percentage greater than 5 % (Figure 5-3). Trees and shrubs species range between 72 % and 26 %.

Herbaceous species represent on average 36 % of the assemblage (without *Pinus*). However, at certain depths individual herbaceous species can make up to 12 % of the assemblage, like Asteraceae (liguliflorae + tubiliflorae) at 790.0 mm (105.15 yr. AD). Common grass pollen, Poaceae, has a low relative proportion of 3 %. Ferns are the less represented group with an average of 15 %, a maximum of 26 % (~87 yr. AD) and a minimum of 3 % (~27 yr. AD).

Throughout the studied section, the pollen and spore association remains relatively constant with the exception of the time interval between 810.0 mm - 790.0 mm (~79 yr. AD - 105 yr. AD).

According to this, the studied core section can be divided into the following three zones:

- Zone 1: 910.0 mm - 810.0 mm, ~50 yr. BC - 79 yr. AD
- Zone 2: 810.0 mm - 790.0 mm, ~79 yr. AD - 105 yr. AD
- Zone 3: 790.0 mm - 725.0 mm, ~105 yr. AD - 190 yr. AD.

Zone 1 and Zone 3 are characterized by a dominance of trees/ shrubs (average of 50 %) that are mainly formed by the combination of *Quercus robur* type and *Quercus ilex* type, representing about 40 % of the association. Apart from the *Quercus* pollen type, *Picea*, *Abies*, and *Fagus sylvatica* constitute a considerable part of the association. Pollen from *Castanea* and *Vitis* is found sporadically, with 2 (in Zone 1) and 1 (in Zone 3) occurrences respectively. *Olea* has a more stable presence in the assemblage in the Zone 1. Cerealia is present sporadically in Zone 3 with a percentage smaller than 1 % but it is not present in Zone 1.

Zone 2 is characterized by an abrupt decrease in pollen from trees/ shrubs as a result of a decrease in pollen from *Quercus robur* type. In this zone, pollen from herbs dominate, notably due to an increase in the relative abundance of Asteraceae and Brassicaceae. At the end of this zone, after about 30 years, the pollen association gradually returns to the situation before this disturbance.

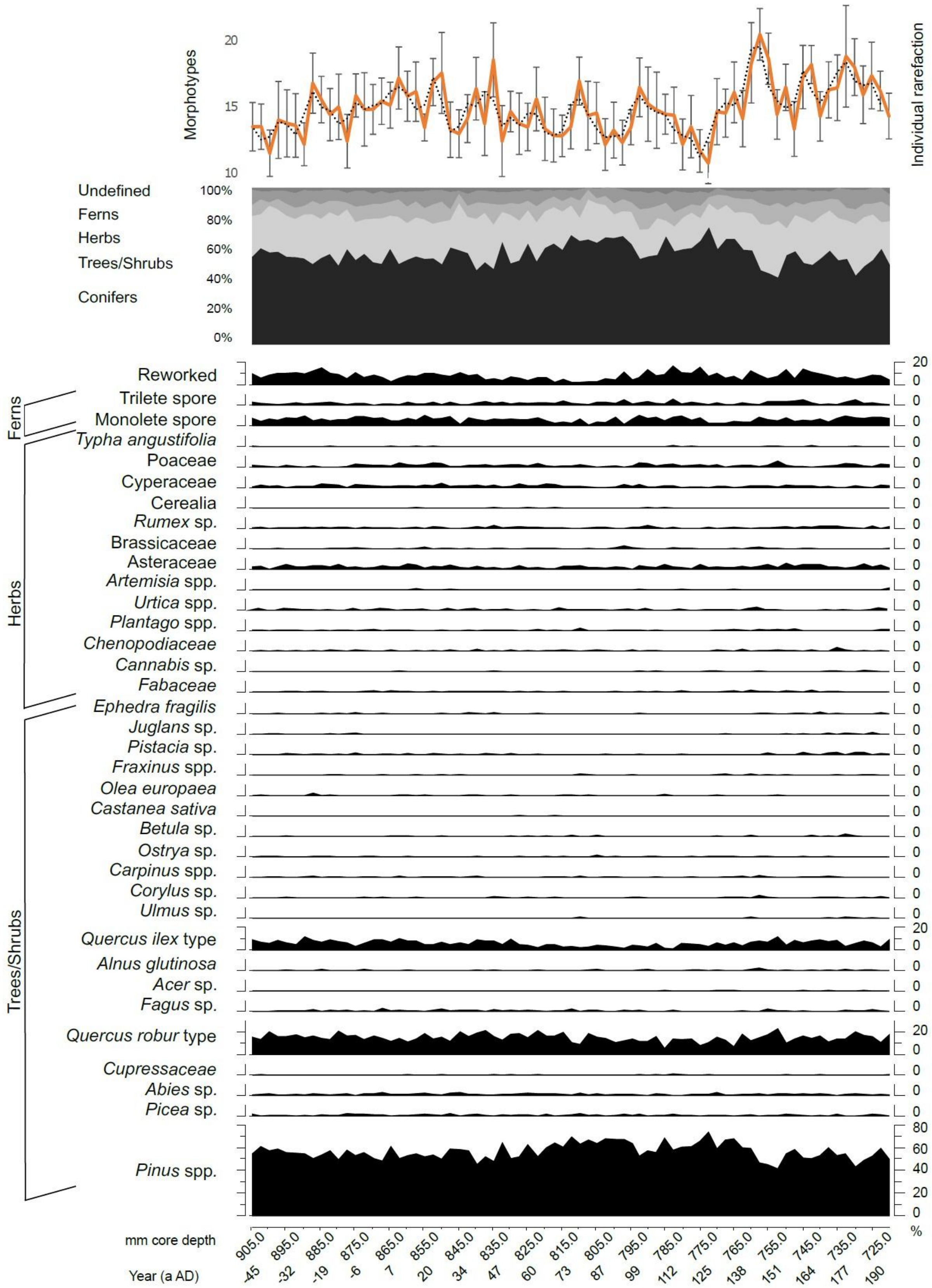

Figure 5-3. Top: Morphotypes and individual rarefaction per sample. Middle: Percentages of the different vegetation groups presented below. Bottom: Relative abundances of pollen and spore species found in core section 8 (725.0-910.0 mm) of the piston core DP30PC.

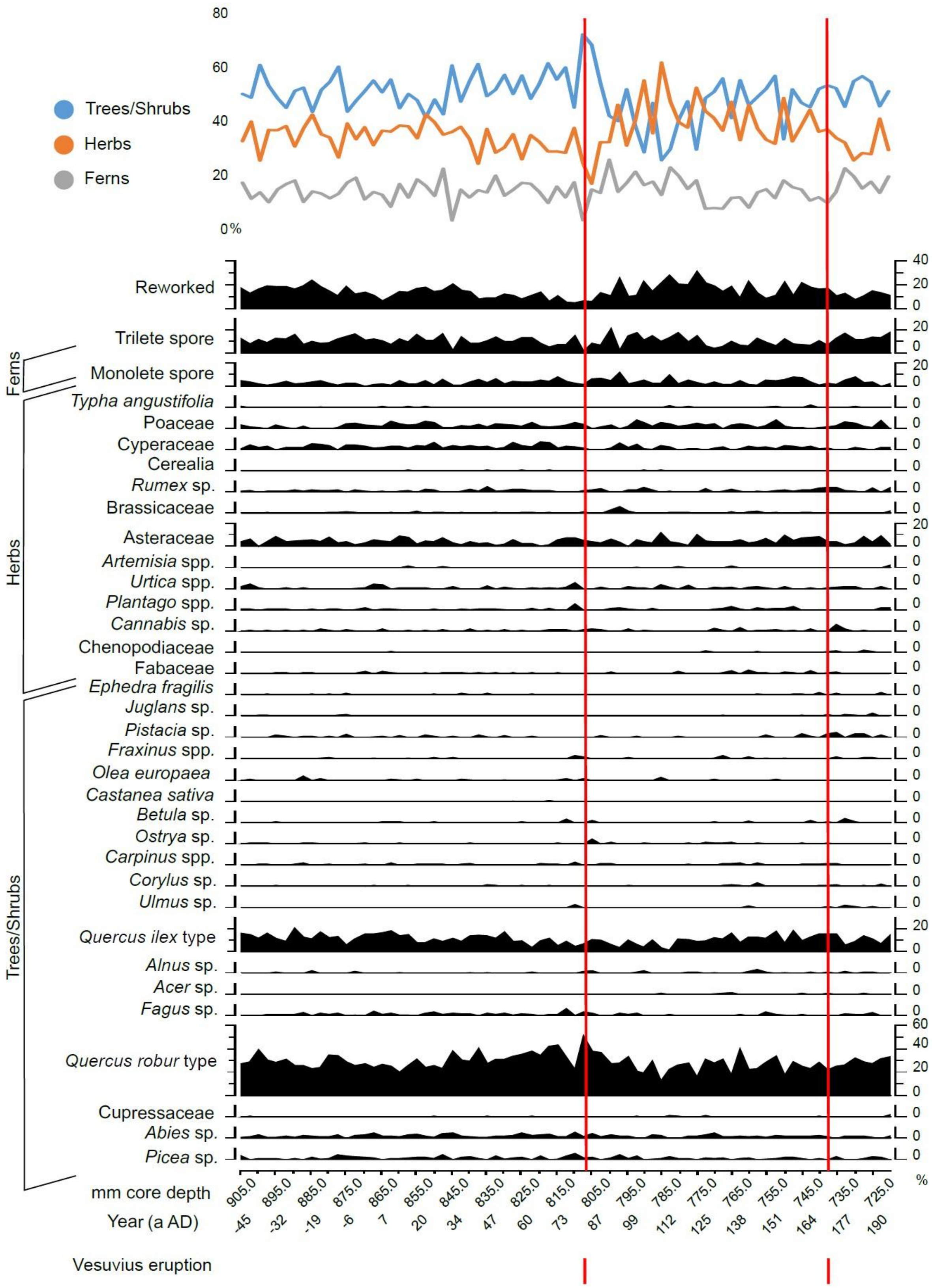

Figure 5-4. Top: Percentages of the vegetation groups. Bottom: Relative abundances without considering Pinus. The red lines mark two known eruptions of the Vesuvius.

4.3. Pollen, spores and dinocysts, a land-sea comparison

A comparative study between dinocyst/g and pollen/g is done using total palynomorphs counts. In addition, further calculation of accumulation rates for both types of palynomorphs is performed.

Concentration values vary between 297 and 3078 pollen/g, with an average of 1029 pollen/g. For the same section, dinocyst concentration varies between 178 and 2003 dinocyst/g, with an average of 679 dinocyst/g. Throughout the studied section, only at five different depths, values of dinocysts are greater than the ones of pollen, concretely at depths 717.5 mm, 720.0 mm, 722.5 mm, 875.0 mm and 910.0 mm (Figure 5-5).

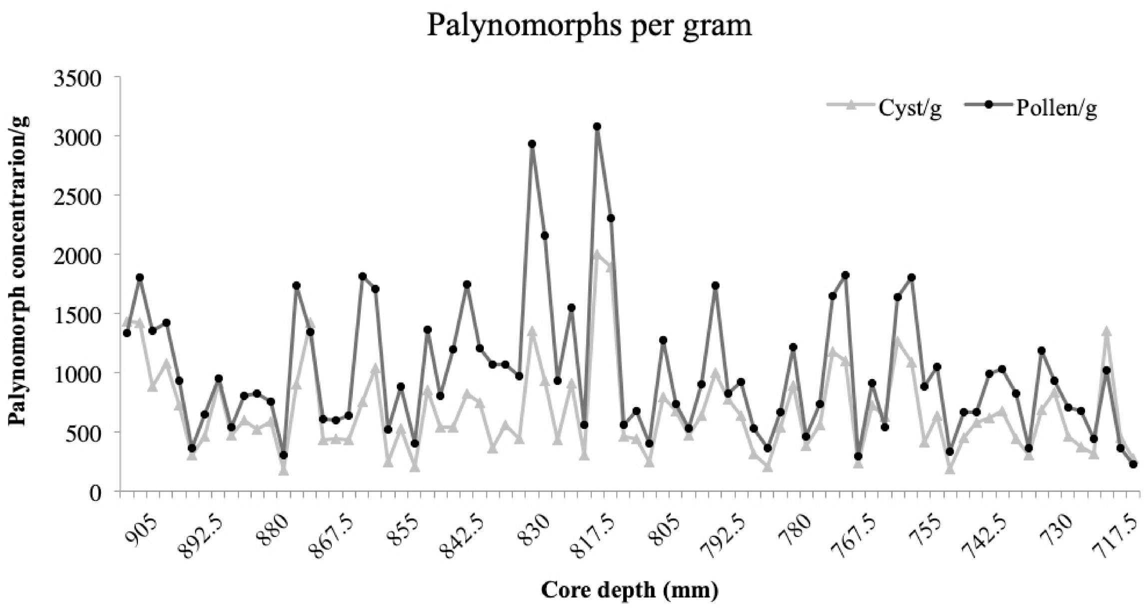

Figure 5-5. Concentrations of palynomorphs (dinocysts and pollen in palynomorph/g) in core section 8.

Pollen and dinocysts show a similar trend for pollen and dinocyst during this period with a positive correlation of 0.8596 (Pearson's Correlation R with a confidence interval 95 %). Pollen-spores/ dinoflagellates proportion varies between 75 % and 46 % with an average terrestrial palynomorph composition for the assemblage of 60 %. No degradation rate is considered for the analysis. No lag phase between the different fluctuations for both groups can be seen. Within the fluctuations, when both groups show an increase in their values, terrestrial palynomorphs express a greater increase than marine palynomorphs. Plotted data (palynomorphs/g) of pollen-spores against dinocyst show a positive and linear correlation too (Figure 5-6).

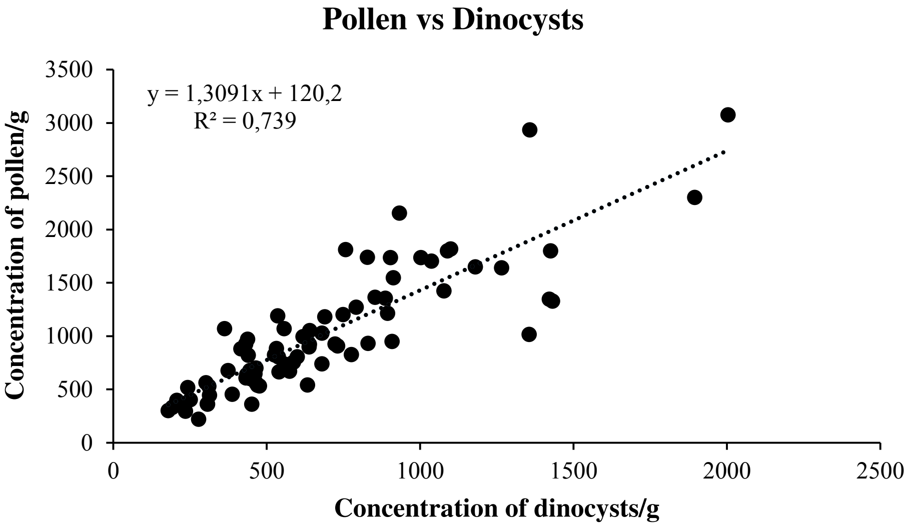

Figure 5-6. Concentrations of pollen/g vs. dinocysts/g in core section 8.

5. Discussion

Throughout the core samples we observe that the pollen/ spore associations are dominated by arboreal pollen, mainly by *Pinus*, which has the capacity to be transported long distances (Whitehead, 1983). The arboreal dominance is suggested to be a typical characteristic of modern marine sediments (e.g. Ganne et al., 2016; Lambert et al., 2017) and it is also seen when atmospheric studies about pollen and spores are performed (e.g. Luo et al., 2018 and references therein). Our data seems to be in line with the results for the Italian vegetation published by Magri et al. (2015). In general, it is broadly accepted that this dominance is caused by higher pollen production through arboreal species compared to the ones of herbaceous plants (Broström et al., 2008; Mazier et al., 2015; Sjögren et al., 2015). Species-specific degradation may have altered the palynomorph association (pollen and spores) found in the sediments (e.g. Keil, 1994; Lebreton et al., 2010; Zhang et al., 2017). Recent studies shed some light about species-specific aerobic pollen degradation that can have a negative effect on some taxa. Other such as *Artemisia*, Asteraceae, *Pinus*, *Taraxacum*, Poaceae, and Chenopodiaceae appear to be more resistant against aerobic degradation (Zhang et al., 2017). This has an effect in the pollen/spore association in which some taxa will be overrepresented and others underrepresented not only because of a difference in net production but also due to selective aerobic degradation. However, the core location is characterized by very high sedimentation rate and therefore, it is expected that pollen and spores have been embedded rapidly in the sediments with minimal exposure to aerobic conditions. This is why we assume that post-depositional selective degradation did not overprint our signal.

The homogeneous distribution of Mediterranean species across the basin do not allow to determine a concrete source of the pollen found in the sediments. Species like *Olea europaea*, *Pistacia*, *Quercus robur* type, *Quercus ilex* type, *Artemisia*, Chenopodiaceae or *Pinus halepensis* can be found not only in the Italian Peninsula (Pignatti, 1982) but also in the north of Africa (You et al., 2016; Abu-Aziza et al., 2017). It means, that under a strong Sirocco pollen from North Africa and even from the Saharan regions, could reach our studied area (Avila et al., 1997; Prezerakos et al., 2010; Chuvochina et al., 2011).

However pollen can also potentially be brought into the system from the Balkan and the Dalmatian mountains, providing in this case pollen of *Pinus*, *Abies*, *Picea* and *Quercus* (Sostaric, 2005). These pollen types not exclusively occur in these regions but can also be found in the Italian Peninsula (Pignatti, 1982). In addition, these species can be present in areas where human activity modifies the land, increasing the proportion of herbs (*Artemisia*, *Plantago*, *Rumex*) and cultivated trees (*Olea europaea*, *Tilia*, *Ulmus*, *Corylus avellana*) (Sadori et al., 2010a,b).

A comparison between terrestrial and marine palynomorphs provides an excellent tool to spot river run-off episodes and the general trend of freshwater input (Versteegh, 1994). We observe that pollen concentrations, and so accumulation rates, vary synchronously to the dinoflagellate cyst concentrations (Figure 5-5). This synchronous behaviour is also found by Beaudouin et al. (2007) in a study in the south of France. Based on the dinoflagellate cyst association, Zonneveld et al. (2009) and Chen et al. (2011) concluded that enhanced cyst accumulation rates were induced by enhanced nutrient availability in surface waters related to high river discharge periods of the Po River and the eastern Italian rivers draining the Apennines. Eker-Develi et al. (2006) attributed this increase in phytoplankton to Sahara-Sahel mineral input. According to Box et al. (2008), this flux is described to be higher in the Roman Classical Period than the average flux for the Holocene, however the study conducted by Chen et al. (2011) rejects the Sahara-Sahel dust flux as the trigger for the dinocyst production in favour of the supply of nutrients by the Po River discharge. The exact relationship between the paired data of the concentration between terrestrial/ marine palynomorphs cannot be determined and the question why they are present in the same order of magnitude remains unknown. Moreover, pollen concentration in marine sediments do not only rely on river discharge input but also on the pollen transported by wind which reach the ocean surface and sink.

The observed synchronous change in the concentration of marine and terrestrial palynomorphs suggests that pollen and spores are transported by rivers. This is seen from the presence of pollen types in the samples from species whose distribution is expected to be located much further northwards, like in the Alps or the northern part of the Apennines. This supports the hypothesis that Po River plume in conjunction with the freshwater inputs from smaller rivers can be traced all the way down to the Gulf of Taranto like suggested by Zonneveld (2009). In Italy, *Picea* is confined to the Alps (Magri et al., 2015) and northern Apennines (Pignatti, 1982) but in this time period of the Roman Climate Optimum it can also be found in the Lake Ohrid in the Balkans (Sadori et al., 2016). So there is strong evidence that its presence in the Gulf of Taranto can be explained by the input from the Po River or secondarily brought with Bora winds from the Balkans (Combourieu-Nebout et al., 2013). However, according to Hicks

(2001), the consideration that *Picea* pollen is brought by wind would only apply to short distances. This long transport wouldn´t be applicable to the distance between the alpine region and the Gulf of Taranto. If temperature in the studied time period was warmer than today, *Picea* could have not thrived in the southern part of the peninsula and this confinement in the north part would have been ever more restrictive. In chapter 4, a study about pollen provenance in the Adriatic Sea also underlines the river input of pollen and spores in this region and the reflection of the vegetation of the drainage area for the rivers discharging their waters on the east side of the Italian Peninsula.

The presented results show no major change in the vegetation of the drainage area of the Po River and the Apennine rivers distributed along the east coast of the Italian Peninsula throughout the studied time interval with the exception of zone 2. Our observation of high relative abundances of trees/ shrubs suggests that these regions were probably mainly covered with forests. The low abundances shown by Cerealia, *Olea*, and *Vitis* suggest as well that Roman activity did not severely alter the regional vegetation. Agriculture was not intensively widespread in southern Italy and trees exploited for culinary usage still represented a minority in the pollen percentage and were more restricted to local plantation (e.g. Sadori et al., 2010a,b; di Rita & Magri, 2009; di Rita et al., 2010).

The Italian population at this time was very low compared to the present-day population. According to Lo Cascio & Malanima (2005 and references therein), population between 200 BC and 200 AD experienced a decrease during the first half of the period reaching 15-16 Mio. inhabitants between years 1 and 100 AD. By 200 AD the population had shrunk to 12 Mio. inhabitants (See Table 5-3). Comparing this data and the associated population density with the current situation it is logical to think that anthropogenic activities couldn't lead to massive land distortions. Currently, the Po River valley is populated by 16 Mio. inhabitants (Po River Basin Authority; In Italian Autorità di bacino distrettuale del fiume Po), which means that during the Roman Climate Optimum, the population for the whole country was as big as the population today accounting only for the Po Valley. So, no vegetation change can be attributed to the population size as zones 1 and 3 do not reflect Roman activities and influence on the vegetation.

We also observe no major change in the association related to the cooling described at about 120 - 150 yr. AD based on dinoflagellate cysts association, dendrochronology and speleothems (Lauritzen, S.-E., & Lundberg, J., 1999; Chen et al., 2011; Lutherbacher et al., 2012, 2016). The average temperature for this period is described as non-stable and warmer than the one today. This is reflected in the analysed pollen assemblage with very marginal fluctuations amongst the identified species. However, the accumulation rate shows large shifts, which could be related to differences in water discharge from the Italian rivers but mainly from the Po River as shown for dinocyst in Chen et al. (2011). The small temperature variation shown by Schönwiese (1995), Lauritzen & Lundberg (1999) or Scafetta (2016), in which a progressive warming to the year 1 AD is followed by a cooling of less than 2 °C, seems to be not large enough to cause a change in the vegetation of the Italian Peninsula. Like other authors have suggested, there is no clear data and evidence to support that this period was warmer than today (Bianchi & McCave, 1999; deMenocal et al., 2000; Frisia et al., 2005; Giraudi, 2009; Taricco et al., 2009). Moreover, the anthropogenic activities may have suppressed the natural vegetation trend caused by the small climatic variation that occurred during this period. There is no clear species preference, wood clearance or indications of extensive agricultural activities. Even though it is known that the Romans modified the land and conducted intensive agricultural activities, the most prominent episodes of deforestation in the Po Valley and the eastern and northern flanks of the central and northern Apennines didn't occur during the Roman times but around 3600 BP and again around 700 BP as sedimentation rates in sediments from the Adriatic Sea show (Oldfield et al., 2003). According to the study by Oldfield et al. (2003), it is also not clear to what extent climate change caused a cultural change or amplified the effects caused in the vegetation by human activities.

Table 5-3. Population estimation, density and % in comparison to present-day population between the years 200 BC and 2006 (Lo Cascio and Malanima (2005) and references therein).

Year	**Estimated population (Mio. Inhabitants)**	**Population density (inhabitants/km^2)**	**% In comparison with today**
-200	6-8	23.2	11.53
1	10	33.2	16.49
100	15-16	49.8	24.7
200	12	39.8	19.8
2016	60	201.3	

A major short-term association change between 79 - 105 yr. AD is observed. During this time, a decrease in *Pinus* pollen occurs. The beginning of this period coincides with the year of the most known Vesuvius outbreak. Within the following 30 years after the volcanic eruption, also *Quercus robur* type pollen concentration declined. It is quite unlikely that this eruption affected the Po River region or the rivers from the Apennines. The exact reasons for this decrease in *Quercus robur* type remain unknown. The relationship between the Vesuvius eruption (79 AD) and its effects at larger scales is not clear. The major destructive effects happened in the surrounding cities of Pompeii, Herculaneum, Oplonti and Stabiae (Giacomelli et al., 2003). The eruption took place at the same time as the most devastating plague to date, which was causing around 10000 casualties per day in Rome but no direct relationship between the two events are known (Harper, 2017 and references therein). It seems to be obvious that no natural force directly caused this decrease in arboreal pollen and especially in *Quercus* *robur* type. The high number of destroyed properties like houses or boats led to an increased need for wood and so intensified the logging rate in the region. The rebuilding was probably mainly done with Oak, which caused this abrupt decrease. After this forest clearance, more rapid growing, yearly and opportunistic species, like herbs, covered the surrounding land as the data shows. It took several years for the forest to recover its previous state, which is reflected by the gradual decrease of non-arboreal species in favour of trees and shrubs.

6. Conclusions

In order to obtain insights into the changes in vegetation and climate, and to what extent the Romans wielded influence on these parameters, we studied the pollen and spore content from a well-dated marine sediment core from the Gulf of Taranto (SW Italy) covering a time period within the Roman Climate Optimum. The investigated sediments show a pollen spectrum including species that are confined to the northern part of the Italian Peninsula, like *Picea*. The fact, that these northern species are still present in the southern Gulf of Taranto leads to the conclusion that Po River waters, in combination with other eastern Italian rivers, reach far southward. This is in line with many other studies in this region.

Overall, vegetation did not change dramatically during this time period and human influence on the regional vegetation was low. We observe that agriculture was more locally restricted and not intensively spread probably due to population size. Trees and herbs with culinary uses were still not spread enough to constitute a big part of the association. An exception is the short vegetation change with *Quercus robur* type decreasing in the pollen sum following the Vesuvius eruption around 79 AD. We suggest that the Romans selectively cleared oak trees from the forest probably related to rebuilding and reparation works. For the first time, such short-term vegetation disturbance by Romans after the Vesuvius eruption is recorded.

Past high river discharge episodes of the Po River in this period might be reflected by the observed similarity in the dynamics of the pollen/ spores and dinoflagellates concentrations. A fixed pollen/spore-dinoflagellates ratio cannot be derived from our data but further studies in this region could reveal, due the singular characteristics, if such index could be possible to achieve. The specific contribution of wind and rivers to the palynomorphs supply into the system can't be distinguished. Therefore, a more detailed study is required, combining core analyses from adjacent regions and from the Po River mouth in order to elucidate the net income that both riverine and wind have in our studied area have.

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Chapter 6 - Manuscript 3

“Vegetation trend reconstruction during the Post-Industrial Revolution based on pollen and spores using a well-dated marine sediment record from the Gulf of Taranto (South Italy)”

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Abstract

In this study, the pollen and spores trends after the Industrial Revolution (in Italy) are reconstructed by analysing a well-dated sediment core (~3.5 years temporal resolution) retrieved from the Gulf of Taranto covering the period 1838 - 2006 AD. The proportions of arboreal and non-arboreal pollen remain quite constant over this time interval, with a dominance of arboreal pollen. Pollen types related to the north of the Italian Peninsula are found in the sediments, as they are transported in suspension southwards to the Gulf of Taranto in plume waters discharged by the Po River. No evidence of a significant human intervention is found in the pollen assemblage, due to low percentages for pollen species related to human activity. Pollen and dinocyst concentrations show a positive linear correlation. Years, in which high Po River discharge or floods are described, are also reflected as peaks in the palynomorph concentration.

The results demonstrate the tight relationship between Po River discharge and palynomorphs found in the marine sediments from the Gulf of Taranto. However, it’s not possible to obtain a specific vegetation reconstruction for the Po Valley region from this dataset due to a progressive dilution of the Po River signal through mixing with other discharge waters of small rivers also draining into the Adriatic Sea.

1. Introduction

The Industrial Revolution is known as the period in recent human history with the most drastic and rapid changes. It led to an exponential growth of the industrial activities accompanied by a population growth never seen before. Also the expansion of the cities, the improvement of the communication amongst territories to distribute goods as well as the use of natural resources (Peterson, 2008; Buchanan, 2018) increased. The development of new machines and processes resulted in an improvement in productivity and the increase in the population (Buchanan, 2018). For these reasons, more land needed to be claimed for new settlement and traffic areas in detriment of the local wild and less disturbed conditions.

Of special interest is the Italian Peninsula and especially the Po River valley, where a great number of inhabitants are concentrated, and where agricultural and livestock activities are the key economic activity. This activity has modulated and changed the region dramatically, from the number of inhabitants and vegetation type to the Po River discharge volume and chemical content of the waters (e.g. Sangiorgi & Donders, 2004; Sangiorgi et al., 2005; Zanchettin et al., 2008; Zonneveld et al., 2009, 2012).

Another marked environmental consequence is that forest mass has decreased since the beginning of the Industrial Revolution. Forests provide wood to industry and species with higher economic turnover are preferred, leading to a change in the natural vegetation balance. This can be seen in the pollen spectrum in the regions analysed, usually with a shift to species related to human activities.

Pollen studies have demonstrated that there is a great correlation between the relative abundances of pollen from the major taxa found in an assemblage and the relative abundance of terrestrial vegetation. Therefore it is postulated that pollen/ spores diagrams are an excellent tool to study vegetation development patterns because they reflect the change in the vegetation over time (Prentice, 1988), driven notably by changes in temperature and precipitation. Pollen and spores preserved in marine archives are therefore a valuable tool to reconstruct these parameters in the past.

Since the beginning of the Industrial Revolution, neither policies looking after the climate or a strong land regulation existed until the foundation of the Intergovernmental Panel on Climate Change (IPCC) in 1988. Additionally the effects caused by these activities were only partially known. Nowadays, it is established that burning of fossil fuels has led to an increase in CO_2 concentration in the atmosphere, and that this trend has been increasing constantly since the beginning of the Industrial Revolution. It was found that these observations do not have a natural but rather anthropological origin (Neftel et al., 1994; Keeling & Whorf, 2005), denoted by an abrupt increase for all greenhouse gases accelerating the greenhouse effect (IPCC, 2013). Besides the human-induced climate change, natural processes also play an important role in steering the climate (Stott et al., 2000; Carslaw et al., 2002).

This study combines, for the first time, marine and terrestrial proxies to reconstruct the vegetation spectrum change that occurred in the Italian Peninsula and affected the Adriatic Sea during since the 1840s. It will attempt to discern between the pre-industrial state of the oceanic basin and the consequences of anthropogenic activities and their impact on the vegetation. The marine and terrestrial signals are derived from palynological (pollen and spores assemblages and organic-walled dinoflagellate cysts, hereafter dinocysts) analyses of a well-dated sediment core from the Gulf of Taranto. According to Heusser & Morley (1985), Hooghiemstra et al. (2006) or Combourieu-Nebout et al. (2013), marine sediments and hence, its interpretation reflect the regional vegetation. The sediments from the Gulf of Taranto, especially in Gallipoli shelf, are attributed to Po River and the northern Apennines but also to southern Italian rivers (80 % and 20 % respectively) following the results from Goudeau et al. (2014). This study also represents a completion of the work by Zonneveld et al. (2012), in which a deep study and analysis of dinocysts and the relationship between natural and human impacts on the trophic state of the region was conducted.

2. Research area

2.1. Ocean circulation

The ocean circulation of the Adriatic Sea (Figure 6-1) with the dominating water masses Adriatic Surface Waters (ASW) and Ionian Sea Surface Waters (ISW) influences the water masses of the Gulf of Taranto. The cool, low in salinity and high-suspended matter content waters of the ASW have their origin in the river discharge from the Po River and other Italian rivers which drain the Apennines (Degobbis et al., 1986; Boldrin et al., 2005). The highest contribution to the ASW can be assigned to the Po River, even if many rivers drain water from the Apennines into the Adriatic Sea. Its discharge water imprint can still be traced in the most distant part of the discharge plume in the Gulf of Taranto. Additionally, in comparison to the rest of the rivers, the amount of nutrients and suspended matter supplied by the Po River is also higher (Degobbis et al., 1986; Milligan & Cattaneo, 2007). The southward flowing Po River discharge waters are pressed against the western margin due to the Coriolis force inducing cyclonic surface currents (Artegiani et al., 1997b; Lee et al., 2007; Colombaroli et al., 2009). In contrast to the ASW, the ISW is high in temperature and salinity but relatively low in concentration of nutrient and suspended matter (Socal et al., 1999; Boldrin et al., 2005; Caroppo et al., 2006). Another characteristic water mass of the Adriatic Sea is the Adriatic Deep Water (ADW) with cool and dense water in water depths greater than 600 m (Sellschopp & Alvarez, 2003; Hainbucher et al., 2006; Rubino & Hainbucher, 2007). Above the ADW, the so-called Modified Levantine Intermediate Water (LIW) is situated (Sellschopp & Alvarez, 2003). This water mass is followed by the Levantine Intermediate Water (LIW) located between 150 – 440 m water depths. The Modified LIW represents a transition zone between the ASW and the LIW, showing a mixture of the properties of both water masses (Sellschopp & Alvarez, 2003). In the Gulf of Taranto, the LIW is characterized by high salinity, temperature and relatively high nutrient concentration (Nittis & Lascataros, 1999; Sellschopp & Alvarez, 2003). Comparable to the Adriatic Sea, the ocean circulation in the Gulf of Taranto follows a cyclonic pattern and the ASW is mixed completely with the ISW in the Gulf of Taranto after such mixing started to take place already in the Adriatic Sea (Artegiani et al., 1997a,b; Socal et al., 1999; Boldrin et al., 2005; Caroppo et al., 2006).

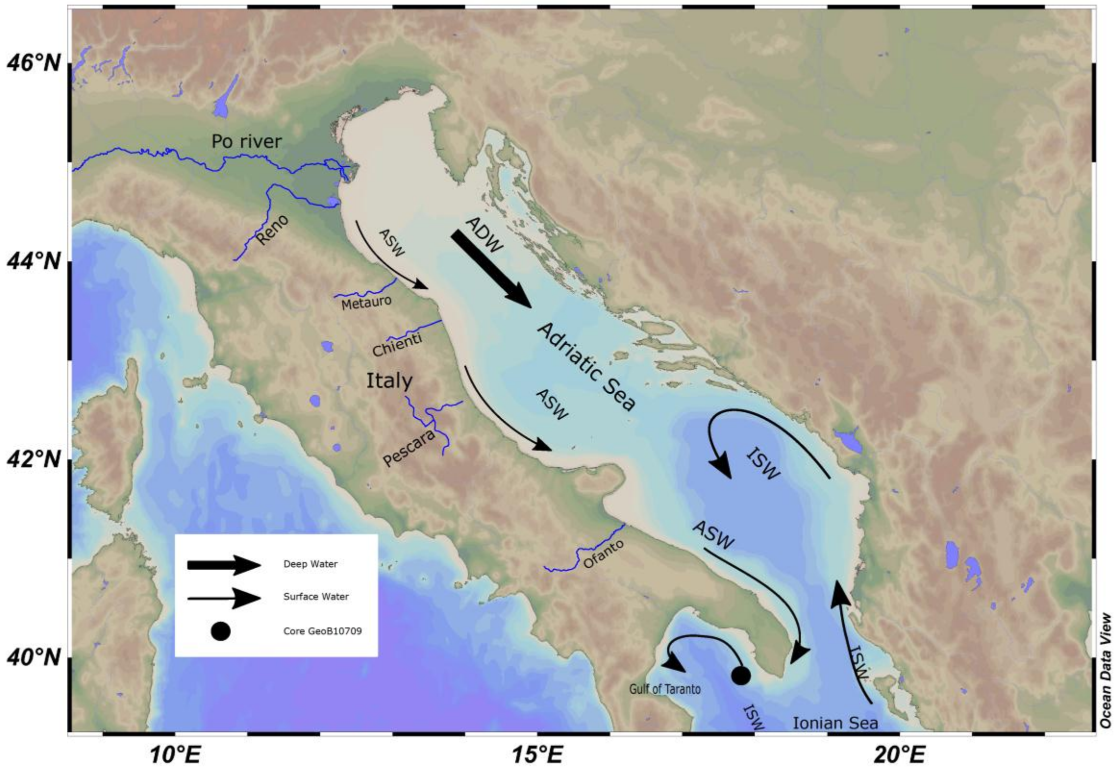

Figure 6-1. Map of the research area depicting core location and main upper ocean surface and deep-water current systems.

2.2. Climate

The climate in the Mediterranean region is characterized by hot, dry summers and cool, wet winters. This dipole is regulated by the combination of three main air circulation systems, the Scandinavian pattern, the East Atlantic (EA) pattern and the North Atlantic Oscillation (NAO) (Hurrell & Van Loon, 1997; Cassou et al., 2004; Fil & Dubus, 2005). Their relationship establishes the clear characteristics and differences between winter and summer. Caused by the difference between high-pressure zone in the Azores and the low-pressure zone in Iceland, the NAO is the air system with the greater influence in winter (Hurrell, 1995).

Also, the rainfall and temperatures all across Europe, the Mediterranean Sea as well as in the Adriatic Sea are influenced by the NAO. A high difference between the high and low-pressure zones results in a positive NAO in this region, whereby the northern regions of Europe are affected with an excess of rainfall in winter. In contrast, a weaker difference between these two pressure systems lead to a negative NAO index in the region. The results are an increase in rainfall and temperatures in the southern regions of Europe in winter. This affects the Mediterranean Sea and the Adriatic Sea as well (e.g. Lionello & Sanna, 2005). As a consequence, the precipitation can be focalized either in the north or in the south of Europe due to changes in the NAO index. Excess rainfall in the southern regions provoke an increase in river discharge by i.a. the Po River and the rest of the Italian rivers into the

Adriatic Sea. For this reason, the amount of ASW increases, forcing the plume southwards to the Gulf of Taranto (Chen et al., 2011).

On a regional scale, the atmospheric circulation is also controlled by the main wind systems Bora and Sirocco (Artegiani et al., 1993; Kourafalou, 1999). The dry and cold continental winds of the Bora blow from the northeast of Europe (Artegiani et al., 1993; Orlić et al., 1994; Palinkas & Nittrouer, 2006). It has the capacity to induce evaporation and heat loss of the water mass in the Adriatic Sea (Artegiani et al., 1993), resulting in an activation of the deep water in the northern part of the basin (Hendershott & Rizzoli, 1976) as well as of the Coriolis forcing (Lee et al., 2007). Blowing from the southeast, the Sirocco acts as a good carrier of humid and warm air from the Mediterranean Sea and the northeastern territory from Africa into the Adriatic region (Artegiani et al., 1993, Cavaleri et al., 1997; Pasaric et al., 2007). Both winds do not show a seasonal preference and can happen at any moment with the exception that strong Sirocco winds are more frequent in spring (Sivall, 1957). Although with different results, Bora and Sirocco winds play an important role on the Adriatic Sea level and water masses circulation, even if they do not have an extensive duration (Orlić et al., 1994).

In general, the Bora winds lead to a confinement of the Po River discharge waters against the coast. In the northern Adriatic, it also contributes to the formation of dense and deep water (Hendershott & Rizzoli, 1976; Kourafalou, 1999). Sirocco winds result to a spreading of the discharge waters eastwards across the northern part of the basin (Orlić et al., 1994; Kourafalou, 1999; Palinkas & Nittrouer, 2006). Furthermore, the accumulation caused by Sirocco winds can cause occasional floods episodes in the northern part of the basin (Orlić et al., 1994; Pirazzoli & Tomasin, 2002; Ferrarese et al., 2008; Jeromel et al., 2009). The formation and changes of water masses, including the alteration of the characteristics like density and temperature, control the amount of dissolved oxygen (DO), which can potentially alter the preservation of pollen and spores (Versteegh & Zonneveld, 2002).

2.3. Po River

The Po River drains the southern part of the Alps and the northern part of Italy forming an extended drainage basin, the Po Valley. Both Alpine and Apennine mountain ranges have approximately an equal contribution to the sediment load of the Po River (Nelson, 1970). For this freshwater input from the rivers, the Po River itself is responsible for around one-third of the riverine input (Kourafalou, 1999). There are other rivers providing sediments to the system but the amount is significantly low compared to the Po River discharge (Penna et al., 2004; Milligan & Cattaneo, 2007). In particular, a mix of sediments from the Po River and Apennine rivers characterizes the sediments from the northern part of the Adriatic Sea (Cattaneo et al., 2003). Following Wang & Pinardi (2002), a bifurcation of the sediments supply occurs at the Po Delta, and some sediment are being transported to the northeast by the northern Adriatic gyre. The fine-grained sediments are as well transported southwards. In addition, resuspension of the material also occurs in the marine realm (Wang & Pinardi, 2002). According to

Boldrin et al. (2005), the amount of freshwater input from the Po River is on a long-term average of 1511 m^3/s.

The hydrology of the Po River consists of two seasonal increases in the discharge of around 2000 m^3/s (in spring and autumn) and some high-discharge events (Kourafalou, 1999; Boldrin et al., 2005). Po River discharge influences the temperature, salinity, sedimentation and nutrient availability but also the water circulation in the Adriatic Sea (Giordani et al., 1992; Tankéré et al., 2000). Po plume water is loaded with sediments, nutrients and freshwater elements from local eastern Italian rivers (Penna et al., 2004; Milligan & Cattaneo, 2007). Water from the Eastern Mediterranean enters the basin through the Strait of Otranto. This water mass, known as Ionian Surface Water (ISW), has a high temperature, high salinity but low suspended matter and nutrient concentration balancing the effects of the freshwater input (Artegiani et al., 1997a,b). It spreads up along the eastern side of the south Adriatic Sea. The completion of the mixing of ASW and ISW occurs outside the basin, within the Gulf of Taranto (Socal et al., 1999; Boldrin et al., 2005; Caroppo et al., 2006; Lee et al., 2007). The amount of Dissolved Oxygen (DO) varies across the Adriatic Sea, both with the latitude but also with depth. It is also possible to find zones with different hypoxia. There are zones eutrophic and hypoxic with an episodic, seasonal or unknown character mostly concentrated in the northern part (European Environment Agency, 2016). This hypoxia in the north has been intensified recently due to eutrophication (Justič, 1991). The three sub-basins in which the Adriatic Sea can be divided (northern, central, and southern) differ in the concentration in DO. Below 100 m, the central and the southern Adriatic show a clear distinction in DO suggesting a weak exchange connection between the two water masses (Lipizer et al., 2014).

The discharge of the Po and Apennine rivers is strongly influenced by the atmospheric circulation. The atmospheric circulation is characterized by two principal wind systems, the Bora and the Sirocco (Kourafalou, 1999; Artegiani et al., 1993). The Bora is a dry, cold and continental wind blowing from the northeast (Artegiani et al., 1993; Orlić et al., 1994; Palinkas & Nittrouer, 2006). It also plays a key role on the evaporation and heat loss (Artegiani et al., 1993), triggering the formation of deep water in the north (Hendershott & Rizzoli, 1976) and Coriolis forcing (Lee et al., 2007). The Sirocco is a moist wind blowing from the southeast transporting humid and warm air into the Adriatic region (Artegiani et al., 1993, Cavaleri et al., 1997; Pasaric et al., 2007). Although it cannot be attributed to a season, it is more common to have strong Sirocco winds in spring (Sivall, 1957). Both wind systems have a different effect on the Po plume as well as an important influence on Adriatic Sea level and circulation, even though they do not have an extensive duration (Orlić et al., 1994).

Bora winds tend to confine and press the Po plume against the coast, meanwhile, Sirocco winds spread the plume eastwards across the northern part of the region (Orlić et al., 1994; Kourafalou, 1999; Palinkas & Nittrouer, 2007). Sirocco winds can occasionally cause flooding events due to the accumulation of water in the shallow North Adriatic (Orlić et al., 1994; Pirazzoli & Tomasin, 2002, Ferrarese et al., 2008; Jeromel et al., 2009). Bora winds participate in the formation of dense and deep water in the northern Adriatic (Hendershott & Rizzoli, 1976, Kourafalou, 1999).

3. Materials and methods

Multicore GeoB 10709-5 was collected with the R.V. POSEIDON during the cruise P339 "CAPPUCCINO" in June 2006 at 39°45.39'N and 17°53.57'E and a water depth of 172.3 m (Figure 6-1) (Zonneveld et al., 2008). After retrieval, the complete multicore was immediately frozen and stored at -20 °C. In order to sample the core in high-precision slices of 2.5 mm, the temperature was raised to -4 °C for 24 h before cutting, which took place at 4 ºC. From each slice, 1 ml was weight and dried overnight at 60 ºC. After the drying time, the remaining pellet was reweighted to calculate the dry bulk density. The material was treated and decanted in subsequent steps with HCl 10 % and HF 40 % according to standard palynological techniques described by Zonneveld et al. (2009) for the aliquot method. Oxidative agents or heavy liquid separations were avoided to preserve selective cyst degradation during sample preparation. For the complete study on organic-walled dinoflagellate cysts (dinocysts) see Zonneveld et al. (2012).

Pollen grains were identified following Moore et al. (1991), Trigo et al. (2008), Beug (2015) and the reference collection of the Department of Palynology and Paleoecology at the University of Utrecht (The Netherlands). Relative abundances of pollen and spores are expressed as percentages of total pollen including herbs, shrubs, and trees throughout the whole study, for the calculation in which *Pinus* is excluded a clarification is made. Relative abundances have been calculated by dividing the number of palynomorphs of a particular species by the total sum of observed palynomorphs (pollen and spores). Dinocyst data to assess the influence of river run-off episodes is part of the paper published by Zonneveld et al. (2012).

Graphs showing palynomorphs percentages were plotted with the software C2 Version 1.5 (Juggins, 2007). On the plotted diagram, only the pollen taxa with climatic or anthropological importance are presented. Data shown represents the relative abundances of pollen and spores of the most abundant taxa (Figure 6-2). Data is analysed with and without *Pinus* pollen to avoid underrepresentation of other pollen types with lower proportion. Each case, with and without considering *Pinus* for the calculation, is mentioned in the text.

3.1 Age Model

As described in Zonneverld et al. (2012), the age model of core GeoB 10709-5 is based on $^{210}Pb/^{137}Cs$ dating with gamma spectroscopy that was provided by the Institute of Environmental Physics, University of Bremen. The wet samples were put into round plastic Petri dishes with a diameter of 7 cm and sealed with Rn-tight foil. In order to ensure a radioactive equilibrium between ^{226}Ra and ^{222}Rn, the sealed samples were stored for 3 weeks prior to the measurements. For the low level, low-background gamma spectroscopy, a coaxial HPGe detector (Canberra Industries) with 50 % relative efficiency housed in a 10 cm Pb shielding with Cu and plastic lining was operated under Genie 2000 software. The applied efficiencies were calculated using the LabSOCS™ (Laboratory Sourceless Calibration System), Genie 2000 software calibration tool in order to deal with self-attenuation (Pittauerová et al., 2009).

The excess-^{210}Pb activity ($^{210}Pb_{exc.}$) was calculated by subtracting supported-^{210}Pb ($^{210}Pb_{sup.}$) from the $^{210}Pb_{total}$ signal that was measured by using the 46.5 keV line. After the establishment of Rn progeny equilibration, the 351.9 keV line of ^{214}Pb was used to determine $^{210}Pb_{sup.}$. Zonneveld et al. (2012) calculated a sedimentation rate of 1.17 ± 0.266 mm/yr under the CIC model (Appleby & Oldfield, 1978).

4. Results

A total of 55 plant taxa were identified in the samples of core GeoB 10709-5 (Appendix A-3). Pollen and spores counted vary between 24 and 254 grains, at 300.0 mm and 67.5 mm depth respectively. The results are shown as pollen and spores percentages across the studied section (Figure 6-2). *Pinus* has, in general, a great proportion in the entire set of samples, suppressing the proportion of the rest of the species in the analysis. Pollen analysis is therefore performed without considering *Pinus* (Figure 6-3). The number of different morphotypes range between minimum 7 at 300.0 mm depth (year ~1853) and maximum 33 at 67.5 mm (year ~1972). Shannon-Wiener index values range between 1.04 and 2.43 at 7.5 mm (year ~2006) and 135.0 mm depth (year ~1937), respectively. Data will be stored on www.pangaea.de.

4.1. Absolute abundance

Total pollen (including *Pinus*) and spores concentration as well as accumulation rate values vary greatly along the studied section, ranging between 1620 and 106 palynomorphs/gram (p/g) at 30.0 mm (year 1990) and 300.0 mm (year 1852), respectively, with an average of 592 p/g.

4.2. Relative abundance data

The pollen spectrum shows different trends and variations during the studied period for the species shown in Figure 6-2. Only 6 out of 55 species appear regularly in more than 75 % of the samples across the section. Most of the species show an intermittent frequency, some of them a constrained distribution. Ferns are present across the entire section, with trilete spores being more abundant than monolete spores.

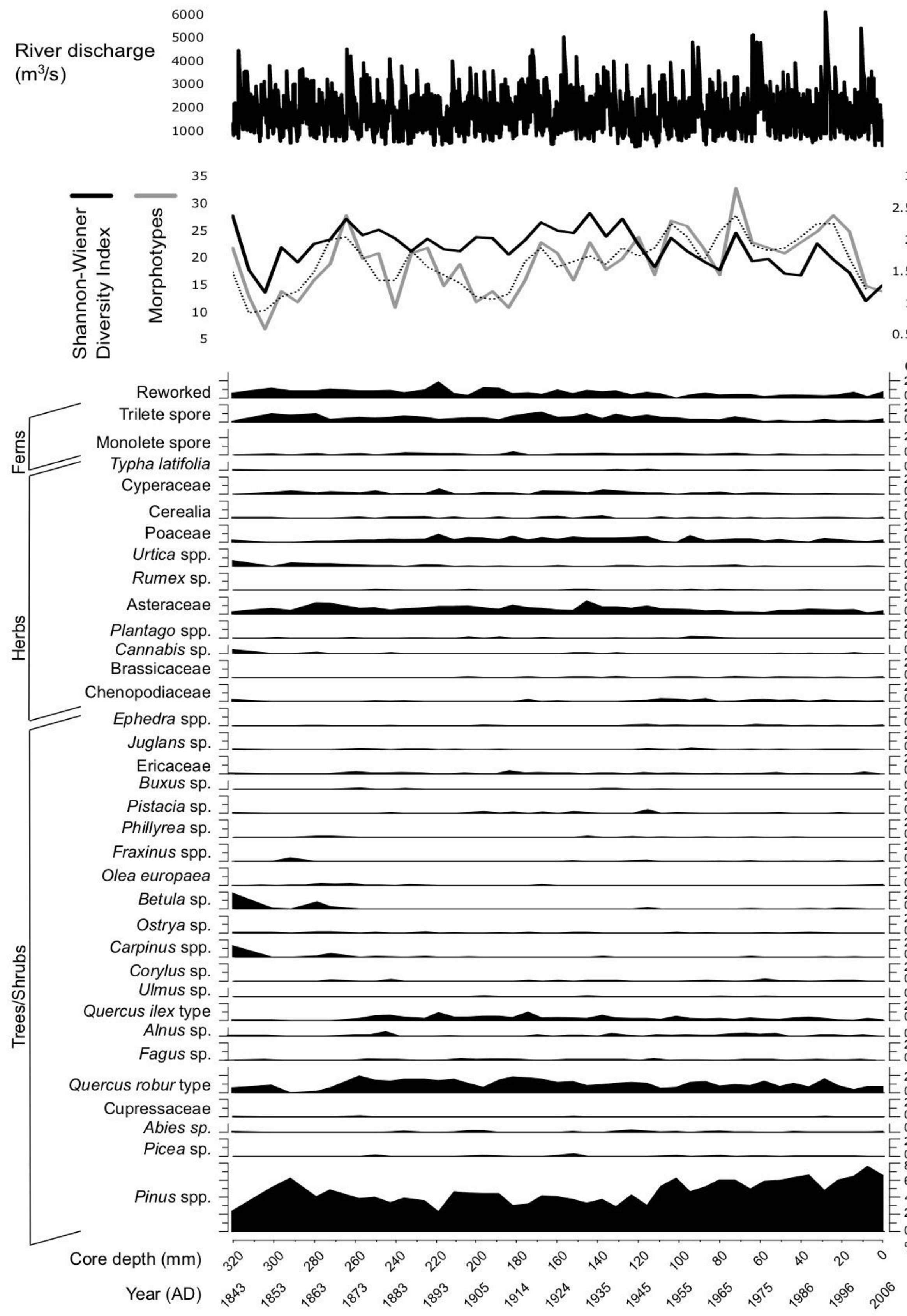

Figure 6-2. Top: discharge of the Po River in m³/s (after Zanchettin et al., 2008). Middle: Shannon-Wiener Index and morphotypes.Botton: Relative abundances of pollen and spore species found in core GeoB 10709-5.

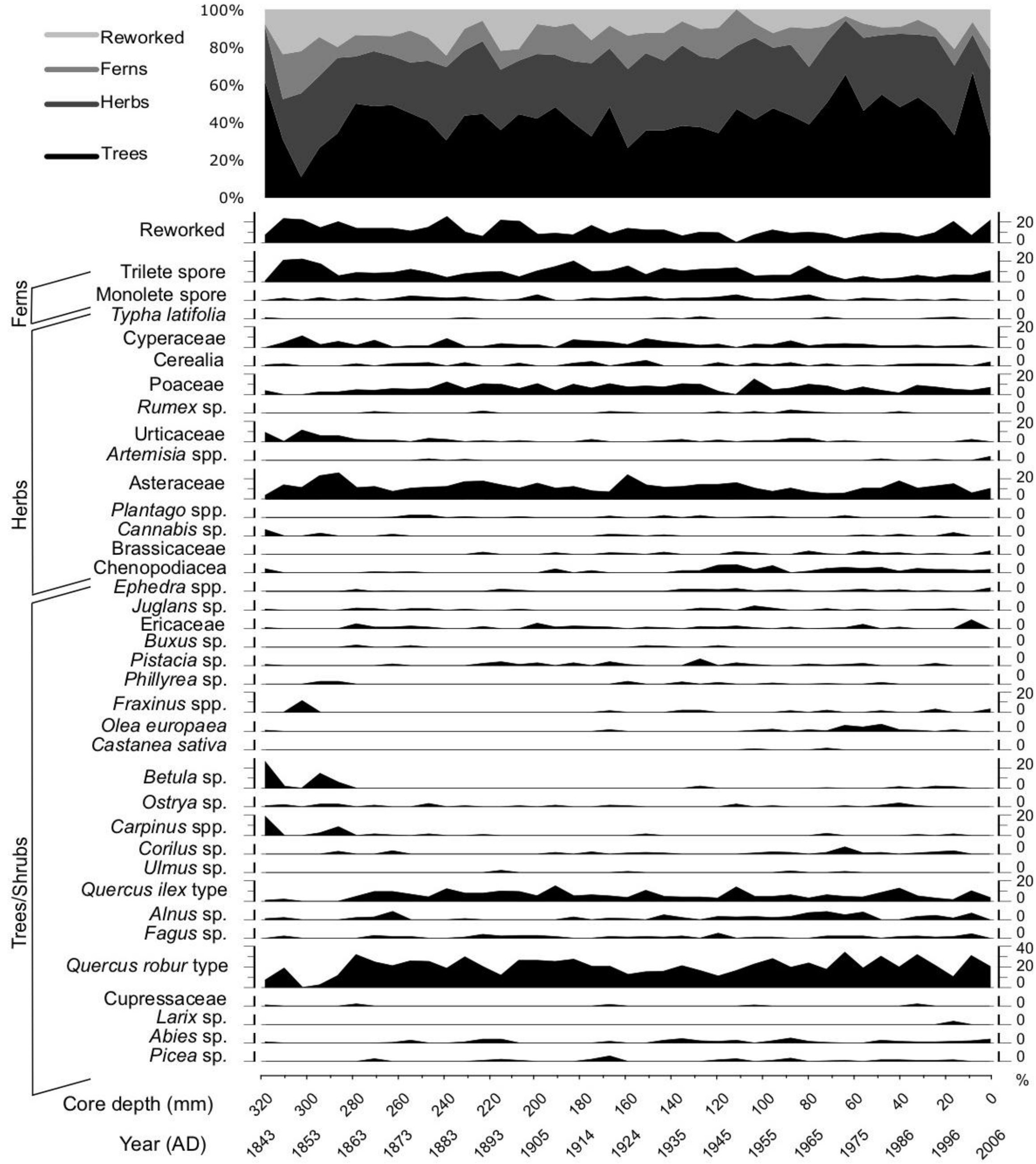

Figure 6-3. Top: Proportions of major groups and reworked pollen. Bottom: Relative abundances without considering Pinus for the calculations.

Pinus is found in all the samples and with the highest proportion of 76 % at (7.5 mm, (year 2002) and the lowest of 23 % at (225.0 mm, (year 1891). The average proportion is 47 %. Without considering *Pinus* for the calculations, trees and shrubs species represent between 64 % and 11 % of the assemblage at 60.0 mm (year 1975) and 300.0 mm (year 1853) with an average of 40 %. After *Pinus*, the next most abundant species in the assemblage is *Quercus robur* type*,* which is only missing at 300.0 mm (year 1852). It represents on average 11 % of the assemblage and has a maximum percentage of 19 % at 265.0 mm (year 1870). *Quercus ilex* type has an average abundance of 3 %.

Alnus, Fagus, Betula and *Abies* are, after *Quercus* types (*robur* and *ilex*), the species with a greater proportion among the other arboreal species. However, not all the species appear constantly in the pollen spectrum or their distribution is confined to specific years. *Betula* and *Carpinus* present a similar distribution and proportion across the studied section, with two peaks between years 1838 and 1863 (330.0 and 280.0 mm, respectively). After the second peak, the proportion for both species decreases and remains low. The decrease in the proportion of these two species is coincident with the increase of *Q. robur type* and *Q. ilex type* in the assemblage. *Fraxinus* has only a single but prominent peak around year 1853 (300.0 mm). From year 1853 (300.0 mm) to 1926 (157.0 mm) it is absent within the sediments but then appears again in the pollen spectrum in a rather constant proportion. Relative abundances of the remaining species of trees are lower than 2 % (Figure 6-2).

Without considering *Pinus*, herbaceous species represent on average 33 % of the assemblage. Certain herbaceous species, like Asteraceae liguliflorae type, can reach proportions of 25 % at 280.0 mm (year 1863). Poaceae group has an average relative proportion of 6 %, with the highest percentage of 15 % at 97.5 mm (year 1956). Poaceae shows an initial increase, which is followed by a long stable period between year 1859 and 1907. After that, intervals with distinct alternating increases and decreases occur. In contrast, the abundance of Cerealia is low but rather stable during the entire time period. Cyperaceae is represented with an average percentage of 3 % and a maximum value of 11 % at 300.0 mm (year 1853). Chenopodiaceae, Brassicaceae and *Ephedra* gain importance within the pollen spectrum to the middle-upper part of the core (Fig. 6-2). Ferns show an average proportion of 10 %.

4.3. Terrestrial vs. marine palynomorphs

According to Versteegh (1994), the comparison of terrestrial and marine palynomorphs provides valuable information about high river run-off events and the general freshwater input pattern. Therefore, the total palynomorphs counts data is used to calculate the concentration for both marine (dinocyst/g) and terrestrial (pollen/g) palynomorphs as well as the ratio between them.

Regarding the concentrations, the pollen/g value averages 573 pollen/g and varies between 272 and 1620 pollen/g. At the same time, dinocyst concentration varies between 219 and 849 dinocyst/g, with an average of 413 dinocyst/g. Throughout the studied section, only at six different depths values of dinocysts/g exceed the ones of pollen/g, namely at 0 mm, 187.5 mm, 202.5 mm, 280.0 mm, 287.7 mm and 300.0 mm depths. Pollen and dinocyst trends described for this period shows a positive correlation of 0.7655 (Pearson´s correlation r with confidence interval 95 %). Pollen-spores versus dinoflagellates ratio varies between 76 % and 30 % with an average terrestrial palynomorph composition for the assemblage of 57 %. Degradation rate was not taken into account for the analysis. Within the fluctuations, when both groups show an increase in their values, terrestrial palynomorphs express a greater increase than marine palynomorphs. Plotted data (palynomorphs/g) of pollen-spores against dinocyst show a positive and linear correlation too. (Figure 6-4).

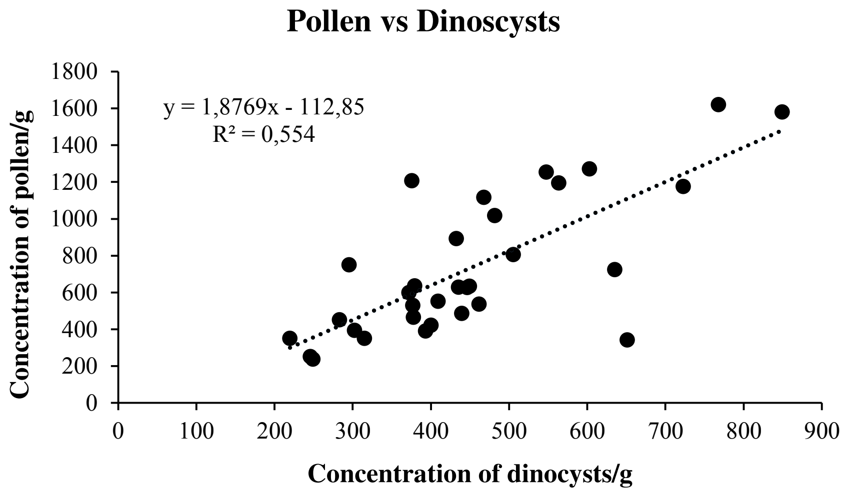

Figure 6-4. Concentrations of pollen/g vs. dinocysts/g in core GeoB 10709-5.

5. Discussion

Vegetation reconstruction, especially the detection of human impact is always a difficult task due to the number of different aspects that affect pollen dispersion and sedimentation. For adequate reconstruction, it is essential to have precise information about how much input is caused by wind and how much by riverine systems. Once in the marine realm, pollen are expected to behave similarly to silt particles due to the comparable particle size and density (Rossignol, 1961). In our case, the difficulty lay on one hand in assessing the relative influence on the transport of pollen as elements forming the dipole wind-water. On the other hand, it is unfeasible to have a completed continuous record covering the whole Adriatic Sea and sediments from all the rivers of the Italian coast. Moreover, to conduct reconstruction analysis it is important to know the possible relationship between the numerical values and the vegetation, in terms of pollen producers. Otherwise, the risk of false analysis due to over- or underrepresentation of different species is very high. Furthermore, there is a difference in pollen dispersion for each species. Herbaceous pollen is diffused a few meters only, meanwhile pollen from trees may travel several hundred meters by air (Erdtman, 1969). This is the case for *Pinus*, whose pollen is distributed largely and transported over long distances (Whitehead, 1983). Another crucial aspect is the (annual) variation of pollen production, which plays a key role for the abundance in the sediments. Also critical is the pollen preservation that can challenge the identification, with corrosion being the most important effect altering the shape and the characteristics (Andersen, 1970). Filtering processes happening in the forest or physical structures, like human constructions highly reduce the pollen load far from the source (Whitehead, 1983).

Po River water influences and changes physical and chemical parameters of the Adriatic Sea water due to the input of nutrients and suspended matter (Degobbis et al., 1986; Kourafalou, 1999;

Boldrin et al., 2005). The discharged waters, as described by Sangiorgi & Donders (2004), are rich in pollen and spores content but also promote the increase of the dinoflagellate population in the marine realm, not only where the waters are discharged but also along the plume formed by these discharge waters (Zonneveld et al., 2009, 2012; Chen et al., 2011, 2013). Its high content in suspended organic matter facilitates the growth of dinoflagellates, appearing in a higher concentration in the plume waters on their way south (Zonneveld et al., 2009; Chen et al., 2011) until they reach the distant Gulf of Taranto transporting pollen, spores and dinocyst (Zonneveld et al., 2009, 2012; Grauel et al., 2013; Goudeau et al., 2014). For this time period, Sangiorgi & Donders (2004) described a vegetation scenario in which the Po River constitutes a great vector for pollen transport into the Adriatic Sea. This is in line with our correlation between pollen-spores and dinoflagellates trends is, where they experience similar increases and decreases over time.

In general, concentrations of terrestrial (pollen and spores) and marine palynomorphs (dinocysts) have a linear relationship, as shown in Beaudouin et al. (2007). Of special interest is the connection between high pollen, spores and dinocyst concentration and years for which high river discharge or floods are described. In Marchi et al. (1995), a list of flood periods is presented. The flood peaks in the years 1971 and 1986 are concordant to the peaks of palynomorph concentrations of our results. We also find high palynomorphs concentrations for the years 1878, 1906, 1929, 1948 and the period comprising 1956-1960. For these years, flood events are also described by Marchi et al. (1995). In contrast, another big flood event, taking place in 2004, is not visible in our results. For this time period, pollen, spores and dinocyst concentration is lower than the concentration found in another core (PC30DP, see chapter 5) but in the same location during the Roman Climate Optimum. The continuous man-made modifications and alterations in the Po River network since the 16th century, with the most recent work on the levee system completed during the 1960s to mitigate destructive floods nearby cities and also to sustent the agricultural activities (Marchi et al., 1995; Zanchettin et al., 2008), may have influenced the discharge of palynomorphs by the Po River . This strong intervention could have even lowered the overall pollen concentration in the southern Adriatic Sea and the Gulf of Taranto.

Overall, Sangiorgi & Donders (2004) detected a shift in the vegetation pattern from arboreal pollen to non-arboreal pollen in favour to Poaceae pollen. Similar is the result of Caroli & Caldara (2007) with a greater proportion of non-arboreal pollen than arboreal pollen in Lago Battaglia in this time period. Furthermore, Di Rita et al. (2018) found that within the last 180 years the pollen record in Lago Patria (SW Italy) is characterized by cultivations and pasturelands, with almost no trees in the spectrum. This shift is not reflected in our study as the plume waters reaching the Gulf of Taranto are also mixed with discharge waters of other rivers along the Italian coast southwards, buffering the local changes occurring in the Po Valley and providing a more integrated regional signal. This buffer effect is also mentioned in the work from Caroli & Caldra (2007) as they only could trace more regional changes concerning the Po River catchment area but no changes at local scale.

The fact that *Larix decidua* is found within our analysed core section even though it is a species described for the north of Italy, supports the hypothesis of a long marine pollen transport due to the Po River plume, and the freshwater contribution from smaller rivers along the Italian coast (Zonneveld et al., 2009). Nevertheless, a straight correlation between the results obtained and the exact numerical quantities for each taxon cannot be done because of the lack of information about pollen production and dispersion for each taxon (Behre, 1981).

The percentages found for *Olea europaea, Fraxinus* and *Phillyrea* differ from the results shown by Magri et al. (2015) and the current vegetation distribution (Blasi, 2005, 2010). Magri et al. (2015) describe a higher abundance of Oleaceae pollen types in the south of Italy compared to our results within the studied time period. For *Olea europaea*, low values are also found by Caroli & Caldara (2007). *Olea europaea* is widely and intensively cultivated due to its culinary uses, being described as a widely spread taxa for at least 3300 yr BP (Schneider, 1985; Magri & Follieri, 2000; Sadori & Narcisi, 2001; Russo Ermolli & di Pasquale, 2002). In our study, the low values recorded for these pollen types can be explained either by pollen loss during the treatment of the sediment or by the generally low richness in the used core. This is supported by the modern pollen surface distribution (see Chapter 4) with high concentrations of *Olea* pollen types in the southern Adriatic Sea and within the Gulf of Taranto. Besides *Olea* pollen abundances, the very low frequency of *Castanea sativa* in our dataset is also in agreement with Caroli & Caldara (2007), who found only one grain of Castanea in Lago Battaglia around 4400 yr BP.

In general, our data shows a more stable vegetation pattern without major changes in the species abundances within the investigated time interval. The described variations in the pollen spectrum over time are for most of the species not sensitive enough to see a clear trend or shift. An exemption for the quite stable proportions seen in the arboreal pollen is the change in the trend and the proportions observed for *Betula pendula* and *Carpinus* that more or less disappear after year 1863. As opposed to this, abundances of *Q. robur* type and *Q. ilex* type pollen rise significantly at the same time. This shift could be caused by land alteration due to anthropogenic influence.

Chenopodiaceae, *Plantago*, *Rumex* and *Urtica* are usually described as good indicators of anthropogenic activities (Behre, 1981). In our data, only Chenopodiaceae reaches significant values after 1941 (127.5 mm) to be considered as an indicator of human activities and land disturbances. The remaining anthropogenic indicators either appear in very low proportions or show frequencies too low to be used as indicators of land distortion. Sangiorgi & Donders (2004) found that these taxa gained importance within their analysed pollen spectrum after 1910. They conclude land alteration but also a higher distribution and dispersion of dry land taxa. Therefore, an increase in human activity after around 1940 is conceivable also in the south of Italy, as indicated by Chenopodiaceae.

Fern spores are brought into the system by rivers indicating areas of swampland. These so called Lidos or Lagoons are common in the region. Like Cerealia and Poaceae, fern spores also are found in a high proportion in the Po River mouth as shown by Ruiz Soto et al. (To be resubmitted, Chapter 4). Recent trends show that ferns tend to decrease in the Po River valley region due to the loss of swampland (Sangiorgi & Donders, 2004). In our dataset this decrease is less acute but it is also remarkable and in line with Caroli & Caldara (2007).

6. Conclusions

For the detection of vegetation changes during a strongly human influenced time period, a well-dated sediment core from the Gulf of Taranto, covering the Post-Industrial Revolution time period of 1838 - 2006 AD was analysed. The study reveals that a diluted signal from the Po River reaches the Gulf of Taranto. Small rivers discharging along the Italian east coast must buffer the detected Po River signal.

The presence of the northern/ alpine species *Larix decidua* in one of the sample indicates long marine pollen transport from the north of Italy to the southern Gulf of Taranto.

During the past last centuries, human intervention in Po River network has altered the general water discharge volume. Our data shows low concentrations in general and not clear trends for the majority of the species. Compared to the Roman Climate Optimum, a substantial decrease in total pollen concentration is found during this time period. Concentration of pollen, spores and dinoflagellates tends to be higher for years of which floods or high Po River discharge events are described.

The studied sediments do not provide a record strongly affected by humans and land alteration. Species indicating anthropogenic activities like *Olea europaea*, *Plantago*, *Rumex* and *Urtica* are underrepresented, making it difficult to assess the anthropological effect. Pollen types found in our core represent a more natural state of the vegetation of the Italian Peninsula with a dominance of arboreal species compared to studies from the north where more species related to human activities are described.

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Chapter 7

Summary, conclusions and future perspectives

The aim of this thesis is to shed some light on the development of the vegetation patterns, as well as the possible influence and alterations caused by humans on the vegetation of the Italian Peninsula using pollen and spores found in marine and riverine sediments as a proxy. The analyses were performed with marine sediments from the Adriatic Sea, Ionian Sea, the Gulf of Taranto and selected rivers. Besides the results of pollen and spores, dinoflagellates cysts data was also taken into consideration to gain insight into the freshwater discharge. This discharge plays an important role for the palynomorph association found in marine sediments. Riverine systems act as an aggregator of pollen and spores on their way to the discharge point in the marine realm providing regional information about processes happening inland (Muller, 1959; Dupont, 1999; Sangiorgi & Donders, 2004; Zonneveld et al., 2009, 2012).

The sea surface sediment study adds relevant and valuable information about the current state of the art in the region in terms of pollen and spores associations. It allows to understand the respective importance of each studied river according to its input, the importance of aerial transport and the transport of the palynomorphs once suspended in the marine waters. Comparing the data with the current vegetation described for the peninsula leads to a better approach in terms of the source suggested for the different pollen types. It also allows setting the basic understanding of the dynamic of the region in order to conduct vegetation analysis and reconstruction of past periods.

To investigate vegetation patterns in the past, sediment cores DP30PC and GeoB 10709-5, both retrieved in the Gulf of Taranto, were analysed for the second (Chapter 5) and the third project (Chapter 6) respectively. The aim was, in both cases, to reconstruct the development of the vegetation, considering the Roman Climate Optimum (~50 BC and 200 AD) as an example for less disturbed conditions in combination with the beginning of land alteration and degradation assigned to the expansion of the Roman Empire. Moreover, the current development after the significant changes caused by the Industrial Revolution was investigated.

The outcome from the sea surface sediment study is a regional, well-spread and high-value surface sediment dataset covering the area from the Po River mouth to the Gulf of Taranto with several rivers along the east coast. Results of the modern pollen and spores distribution and associations in marine, as well as in river sediments, enable the establishment of provenance at a regional scale for the palynomorphs found on each site. For individual cases, a more detailed study regarding specific wind regimes and marine currents over the studied time period would be needed to achieve a better approach and understanding. In addition, we show that mixing happens in the marine realm as the palynomorphs are transported, and potentially within in the sediments due to resuspension.

In general, the pollen distribution shows a north-south trend across the studied region. Even though the majority of the species are not confined to one specific locality, some of them show a clear preferential distribution. Of special interest and importance, due to their relationship with human activity, are the trends from Poaceae, Cerealia, and Oleaceae pollen types. Poaceae and Cerealia type pollen reach their maximum percentage in the vicinity of the Po River and stay relatively high in comparison to other pollen types in sediments north of Gargano Peninsula. This is highly related to the intense agricultural sector established in the Po Valley, supporting the hypothesis that many of the pollen found in the sediments have an alluvial origin, more concretely a Po River origin. Traces of Poaceae pollen are still found in the south and the Gulf of Taranto due to the long-range effect that Po River plume has on pollen and spores transport as shown in many studies (e.g. Chen et al., 2011, 2013; Goudeau et al., 2013; Zonneveld et al., 2009, 2012). Abundance of *Olea* pollen type increases in the pollen spectrum steadily from north to south, as so its cultivation, reaching the maximum percentage around Apulia.

Even though individual contribution of the studied rivers cannot be addressed, the pollen association found along the analysed cores still shows some river provenance. However, the dominance of the Po River signal in marine sediments buffers the input from other rivers distributed along the Adriatic coast. Rivers and marine sediments northern from Po River mouth were not investigated, though their contribution to the total pool is thought to be minimal.

A study covering the years between ~50 BC and 200 AD, a period also known as Roman Climate Optimum, was performed. This period is estimated to be warmer than today according to many studies, but there is no clear agreement as some other authors report cooler conditions or no differences from the ones we have today. The results show that the vegetation is in general dominated by arboreal pollen with no major human influence during this time period across the studied core section 8 (725.0 - 910.0 mm). It might be related to the population size calculated for that time. During the Roman Climate Optimum, the population of Italy is described to be very small compared to the current population size, hence land distortions and changes may have had rather local than regional scale effects. This study shows an integrated regional signal instead of local changes taking place on the Adriatic side of the Italian Peninsula. It is known that Romans cultivated *Olea europaea*, *Juglans* and *Castanea sativa* across the peninsula as the results from Mercuri et al. (2013) show. It can also be derived that agriculture was more local and not intensively spread due to population size. Such change in the pollen spectrum is more likely to be biased in sediments due to the proximity to villages or human settlements. However, this signal related to human activity is buffered at a larger scale making it difficult to assess the real distortion caused by humans.

The collected pollen data support other authors' findings (Zonneveld et al., 2009, 2011, 2012; Chen et al., 2011, 2013; Goudeau et al., 2014), that Po River waters and sediments reach the Gulf of Taranto. Species from the northern part of the peninsula, like *Picea*, or that are highly related to the Po

River catchment area (e.g. Poaceae and Cerealia) are still present in the southern Gulf of Taranto sediments or around Gargano Peninsula, like in case of *Larix decidua*.

Trees and herbs with culinary uses are still not spread enough to represent a big part of the association. A more detailed study needs to be done investigating sediment cores from adjacent regions that cover the same time period and from the Po River mouth in order to elucidate the net income that both riverine and wind systems have in the studied area. A pollen/spore-dinoflagellates index cannot be derived from our data but further studies could reveal, due to the singular characteristics, if such index could be possible to achieve. A remarkable effect on the vegetation can is seen after the Vesuvius eruption around 79 AD resulting in a decrease of arboreal pollen, especially of *Quercus robur* type, followed by a notable increase in herbs.

The last study, here presented, covers the years between ~1837 and 2006 AD. It shows a pollen spectrum with a dominance of arboreal over non-arboreal species. In general, pollen proportions are very stable during this time period with no major alterations. However, two opposite trends are distinguishable within the assemblage. *Q. robur type* and *Q. ilex type* are present in low proportions during the first years of the time period but then increase in abundance to become the second and the third most frequent species, respectively. *Betula pendula* and *Carpinus* are present in considerable proportions within the oldest samples, but decrease after year 1863 and appear only rarely and/or in very low proportions in the rest of the samples. Oleaceae pollen types (*Olea europaea*, *Fraxinus,* and *Phillyrea*) are present in lower proportions than in other studies considering the same time period but other Italian locations (e.g. Sangiorgi & Donders, 2004; Magri et al., 2015). A modification in the vegetation cannot be derived from our study, even though we still detect a diluted Po River signal in the Gulf of Taranto by the presence of pollen described to grow preferably in the north.

Palynomorphs concentrations reflect human intervention in the Po River network and high discharge events recorded for the Po River. The concentrations of pollen, spores and dinocysts are in general lower than the ones registered for the Roman Climate Optimum, which can be related to human activities. However, the pollen spectrum does not show strong human influence on the vegetation as described by Sangiorgi & Donders (2004). The long distance between the Po River mouth and the core location, and the discharge water of other small rivers along the Italian coast buffer the original Po River signal.

The results of the sea surface sediment analysis and the two down-core studies here presented add valuable information to the knowledge about vegetation development, potential human land change and the transport of palynomorphs in the Adriatic Sea and the Gulf of Taranto. However, to be able to distinguish between the changes in the vegetation caused by natural climate change or by human influence, a better coverage and dataset would be needed. The same idea applies to the understanding of pollen transport and deposition once in the marine realm. Due to the dynamics of the water mass, extremely different values of sedimentation rates for some locations are described which makes it difficult to interpret the results when the sedimentation rate is not known (Nelson, 1970; Boldrin et al., 2005; Palinkas & Nittrouer, 2007). Therefore, information for each core studied is needed in order to make a better use of the results obtained as interpolation does not provide reliable information in such cases.

The relationship between terrestrial and marine palynomorphs can be used as indicator of river run-off events (Versteegh, 1994). As presented in the two down-core studies, there is a direct relationship between the fluctuations in concentration of dinoflagellates and pollen and spores. This reinforces the conclusions of previous studies where the importance of riverine input for pollen found in sediments close to the coast or to river discharge locations is stated. A linear correlation between the concentrations of pollen-spores and dinocysts is obtained for the Roman Climate Optimum and the Post-Industrial Revolution time periods. These results are also supported by the study of Beaudouin et al. (2007) conducted in France, in which a linear relationship between marine and terrestrial palynomorphs is described.

A study with marine cores covering the gap between the Po River mouth and the Gulf of Taranto for the same studied period could reveal preferential deposition sites of the terrestrial palynomorphs, the implication of the river discharge and the remaining palynomorph load reaching the Gulf of Taranto. The period covering since 1838 to 2006 AD presents lower palynomorph concentrations compared to the findings during the Roman Climate Optimum. Human’s works and alterations in the Po River network since the 16th century to date in order to mitigate floods and to support the high agricultural activity of the region may explain the observed decrease in the concentration (Marchi et al.,1995; Zanchettin et al.,2008).

Even though it was not possible to differentiate between natural and human-induced climate change, this study expands the knowledge about palynomorphs distribution and provenance in the Adriatic Sea and the Gulf of Taranto. It also provides insights into the pollen trends for two past periods. This is not an easy task, as the study from Oldfield et al. (2003) on the late Holocene in the same region concludes. In his study, a multi-proxy approach combining seismic stratigraphy, pollen analysis, carbon, and nitrogen analysis, stable isotopes, magnetic measurements, transmission electron microscopy, tephra analysis, foraminiferal analysis, and alkenones, is used to tackle the same question about the degree of natural or human-induced contribution to climate change. Even by using many different analysis, they couldn't find a clear answer to what extent humans changed the climate or if the deviation in the cultural activities is a result of natural climate change.

In relation to the hypothesis and conclusions previously presented, several questions and interesting further investigations arise. For example, the aerial and riverine net contribution to the pollen found in the sediments investigated. To understand and quantify the contribution to the pollen pool by each wind system, it would be helpful to conduct research combining aerobiological and sedimentological analysis along the Italian and Balkan coast, as well as from North Africa. That would provide a pollen signal independent from the riverine input. Aerobiological data from the different locations could help to understand to what extend pollen transported by wind from each potential source region reach the marine realm, making the interpretation of the pollen source in the sediments easier and more accurate. With that, we would be a step closer to understand the preferential transport mechanism of the different pollen types either by the local rivers or by the winds, obtaining a clear separation for both vectors.

Some degree of uncertainty could be related to the sample preparation and the fact that for projects 2 and 3 the material was sieved through 20 µm sieve instead of 10 µm. Sediments from the core DP30PC, across the section 460.0-502.5 µm, were sieved using 20 µm and 10 µm high precision sieves showing no differences in the pollen content in terms of pollen types. No species were exclusively found in samples within the 10-20 µm range and in every case samples that were sieved through 10 µm contained no more than 5 % of the amount of pollen found for the same section using 20 µm sieve. In addition, the down core sections do not provide very rich samples since they present low palynomorphs concentration. In some cases, up to 40 % of the resulting fraction (aliquot method by Zonneveld et al., 2009) was analysed under the microscope with total results no greater than 200 pollen grains. In general, no more than 2 g of wet sediments were processed due to the low remaining material. That could have been the reason for such low palynomorphs values. Incrementing the grams processed would bring higher values of palynomorphs, even for the same number of slides analysed, making the whole study and interpretation more robust.

Chapter 8

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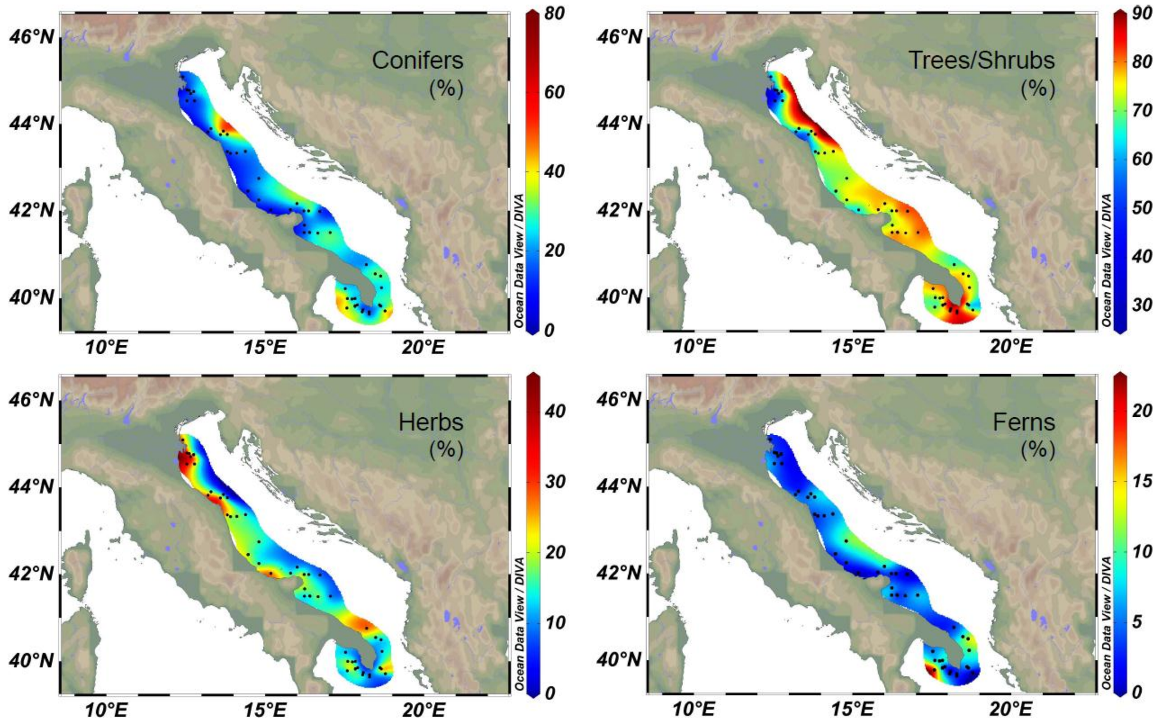
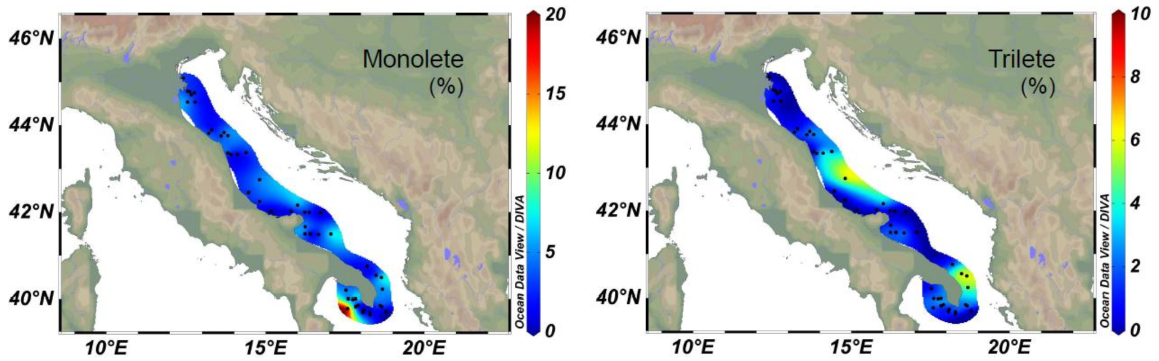
Appendix

A-1 Distribution maps based on relative abundance - Manuscript 1
(Main Groups & Spores with *Pinus*, Trees/ Shrubs & Herbs without *Pinus*)

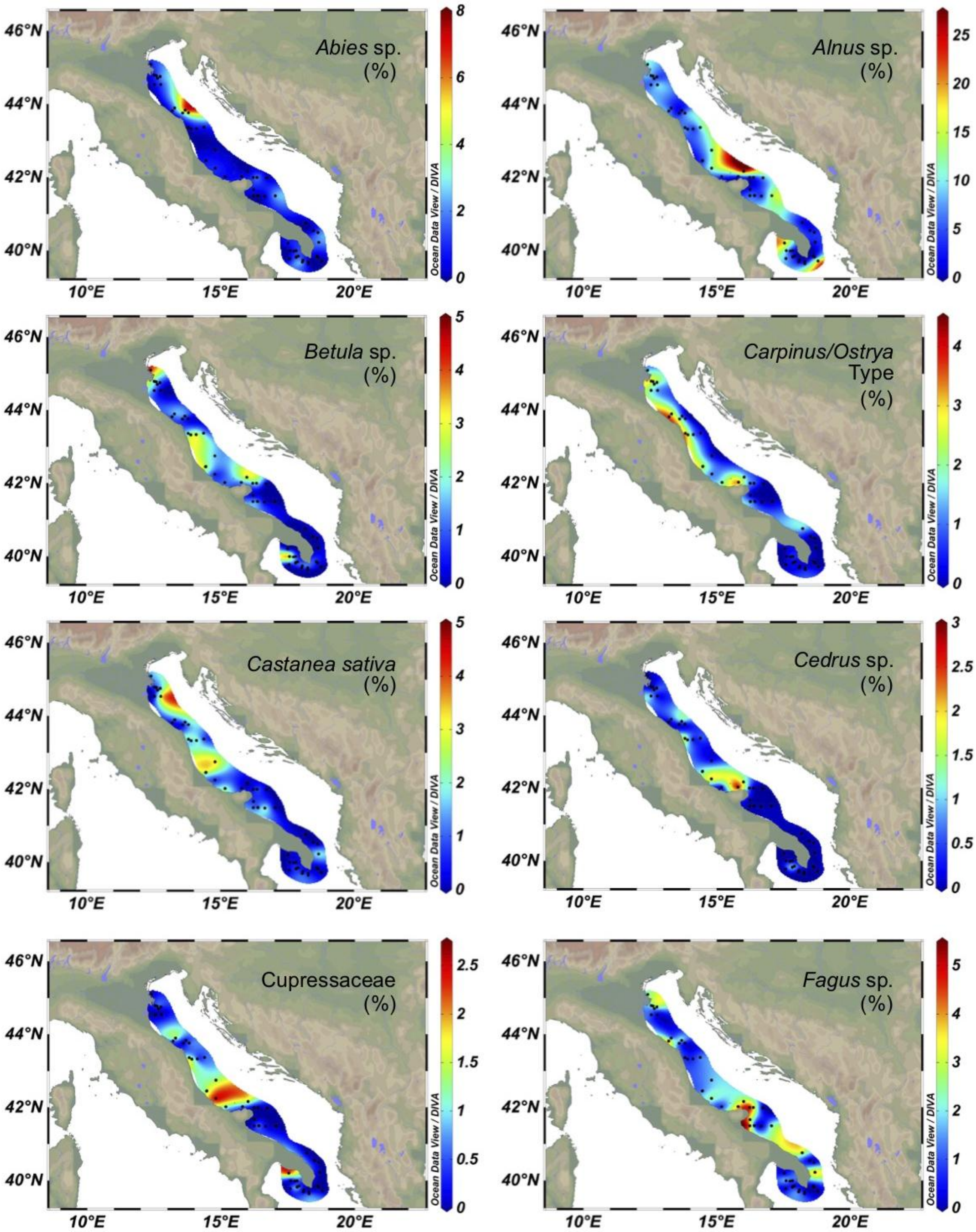
Main Groups



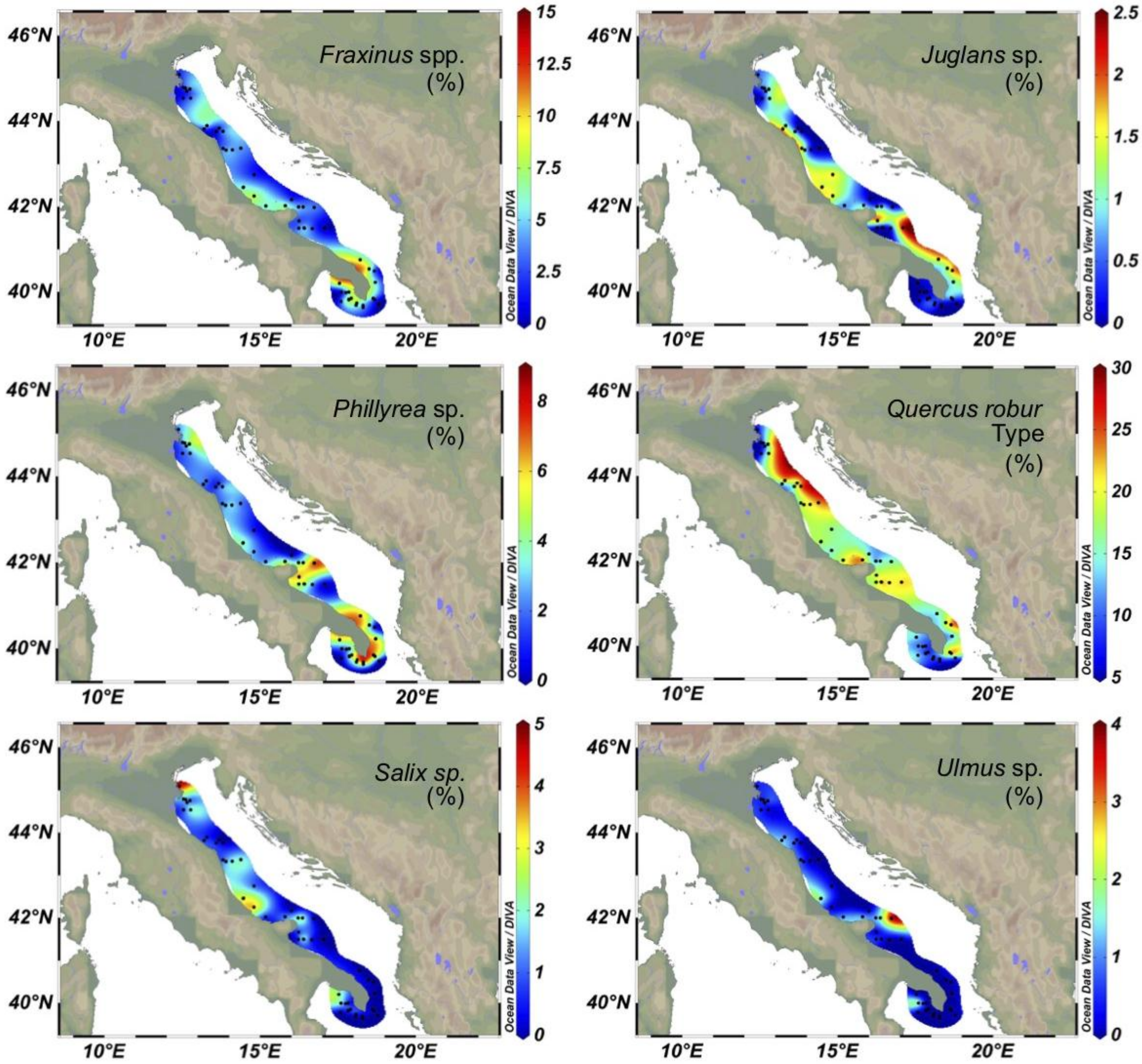
Spores



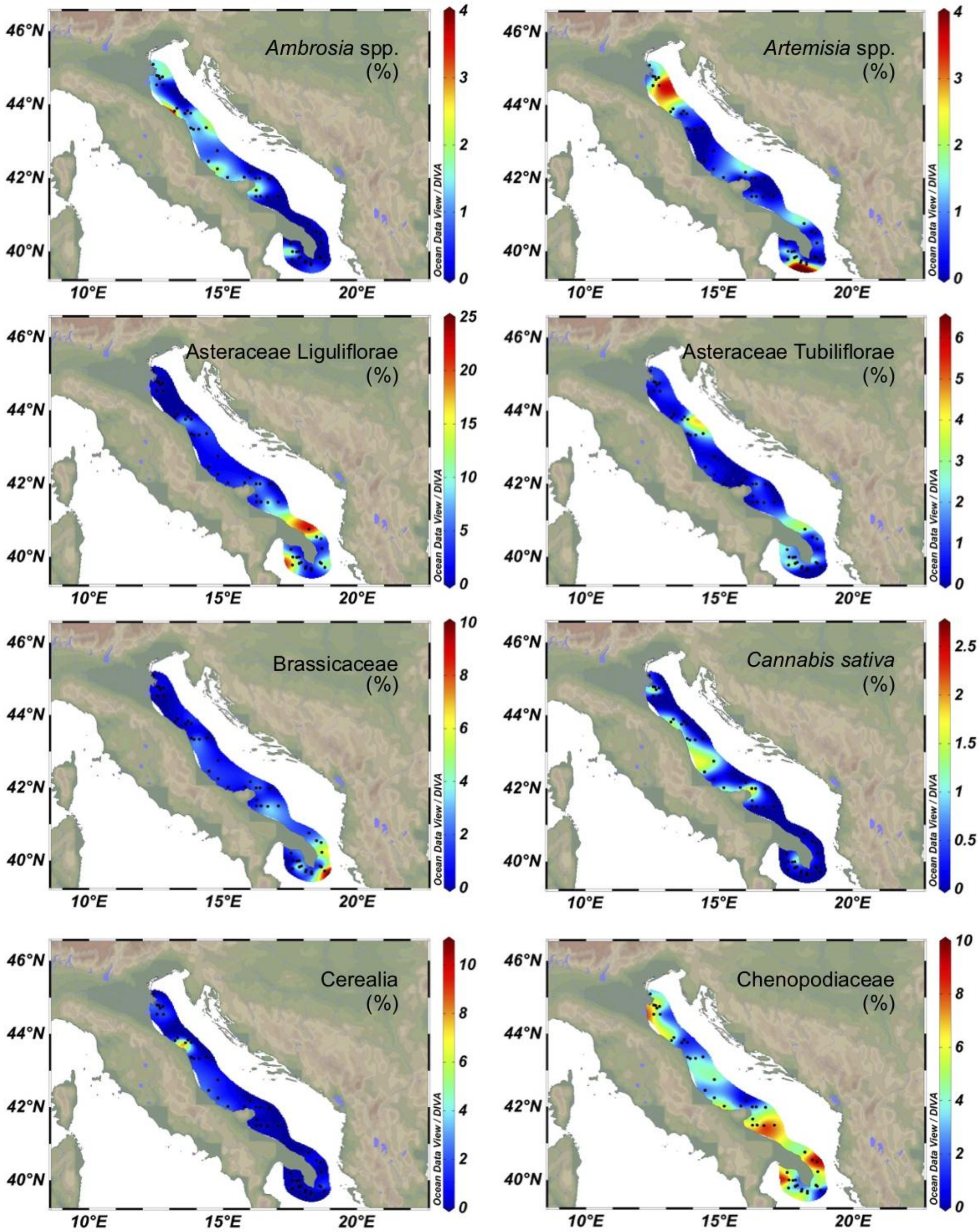
Trees/Shrubs



Trees/Shrubs

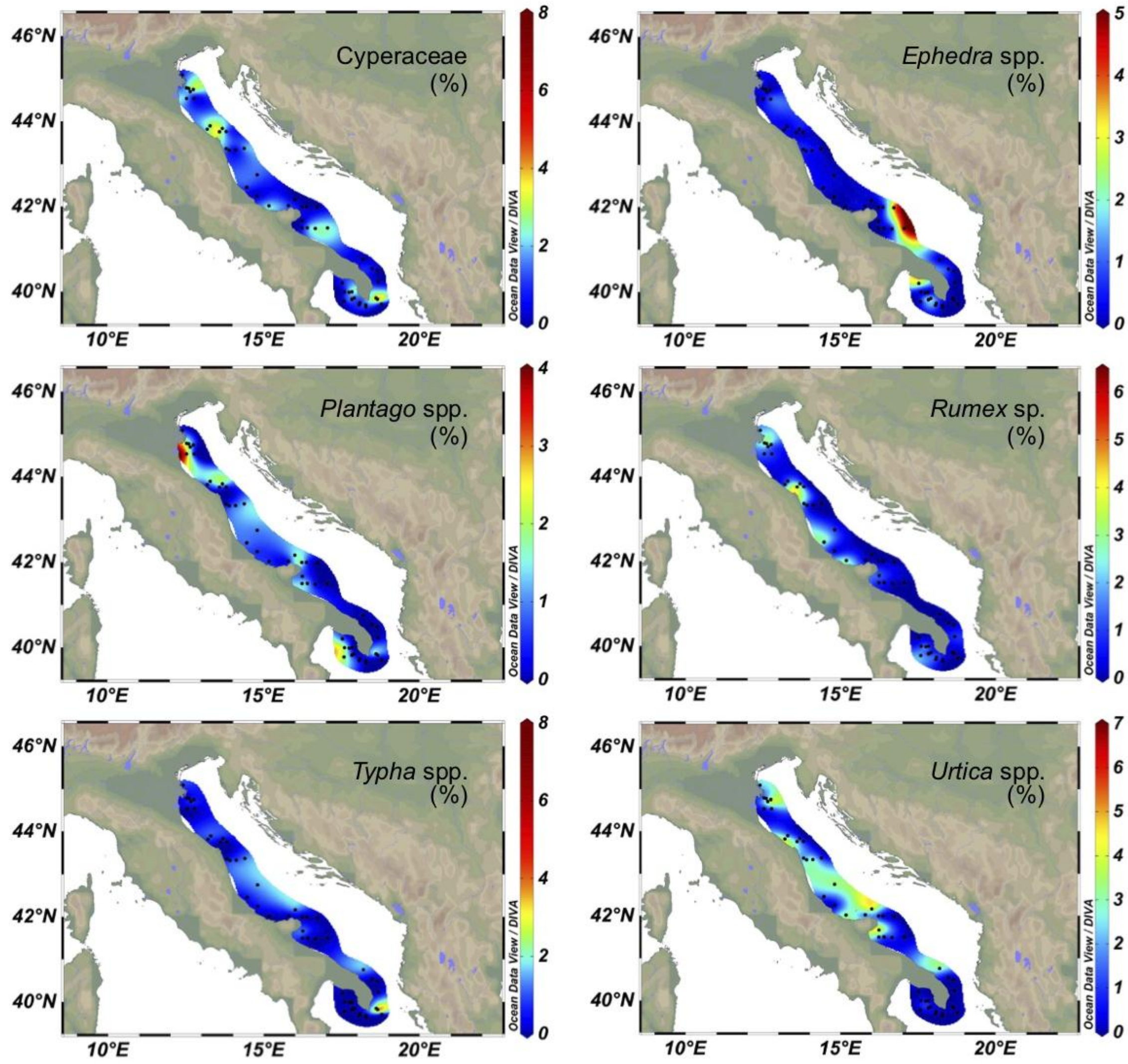


Herbs



Ambrosia spp.
(%)
Artemisia spp.
(%)
Asteraceae Liguliflorae
(%)
Asteraceae Tubiliflorae
(%)
Brassicaceae
(%)
Cannabis sativa
(%)
Cerealia
(%)
Chenopodiaceae
(%)
46°N
44°N
42°N
40°N
10°E
15°E
20°E
Ocean Data View / DIVA

Herbs

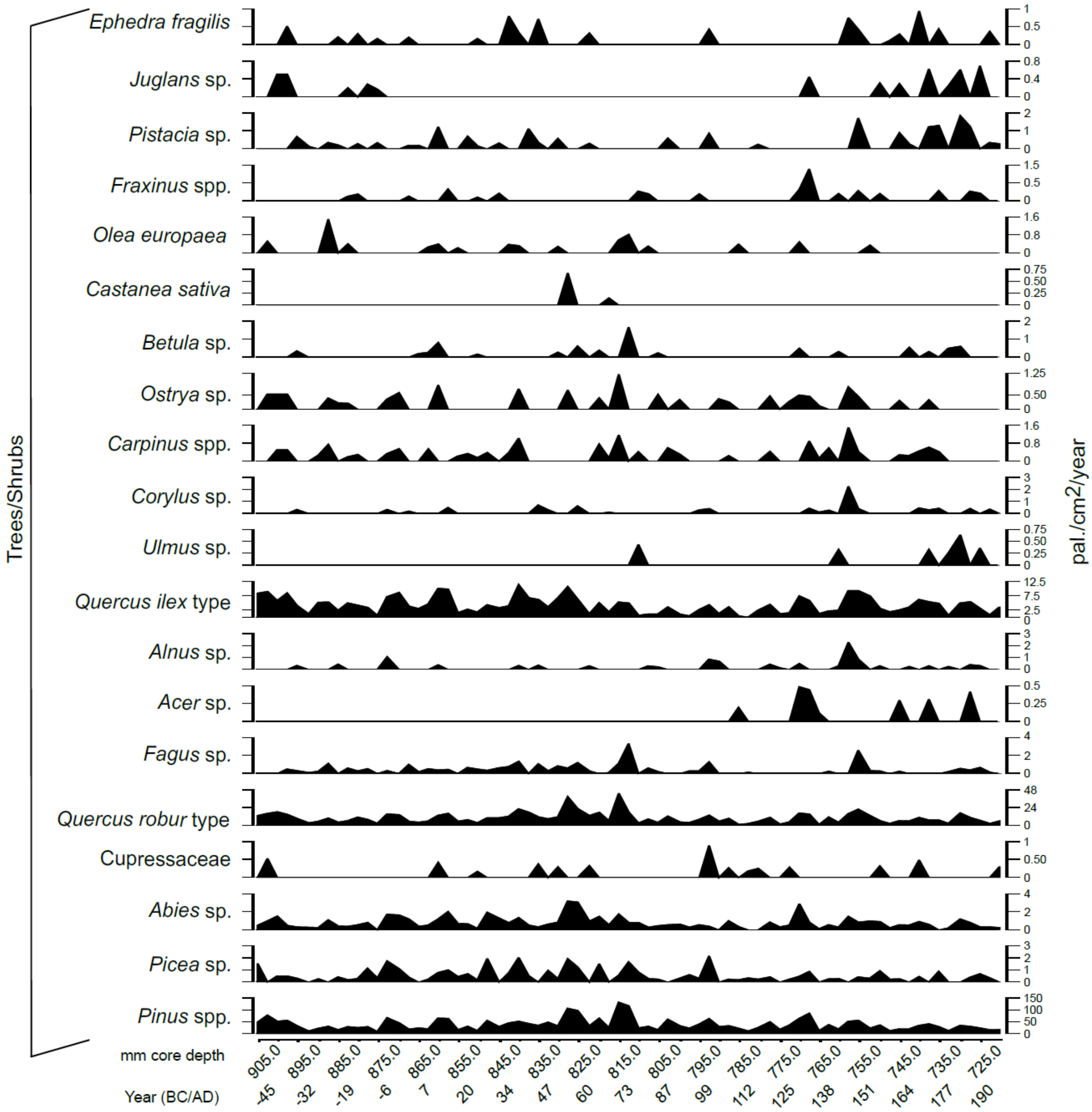


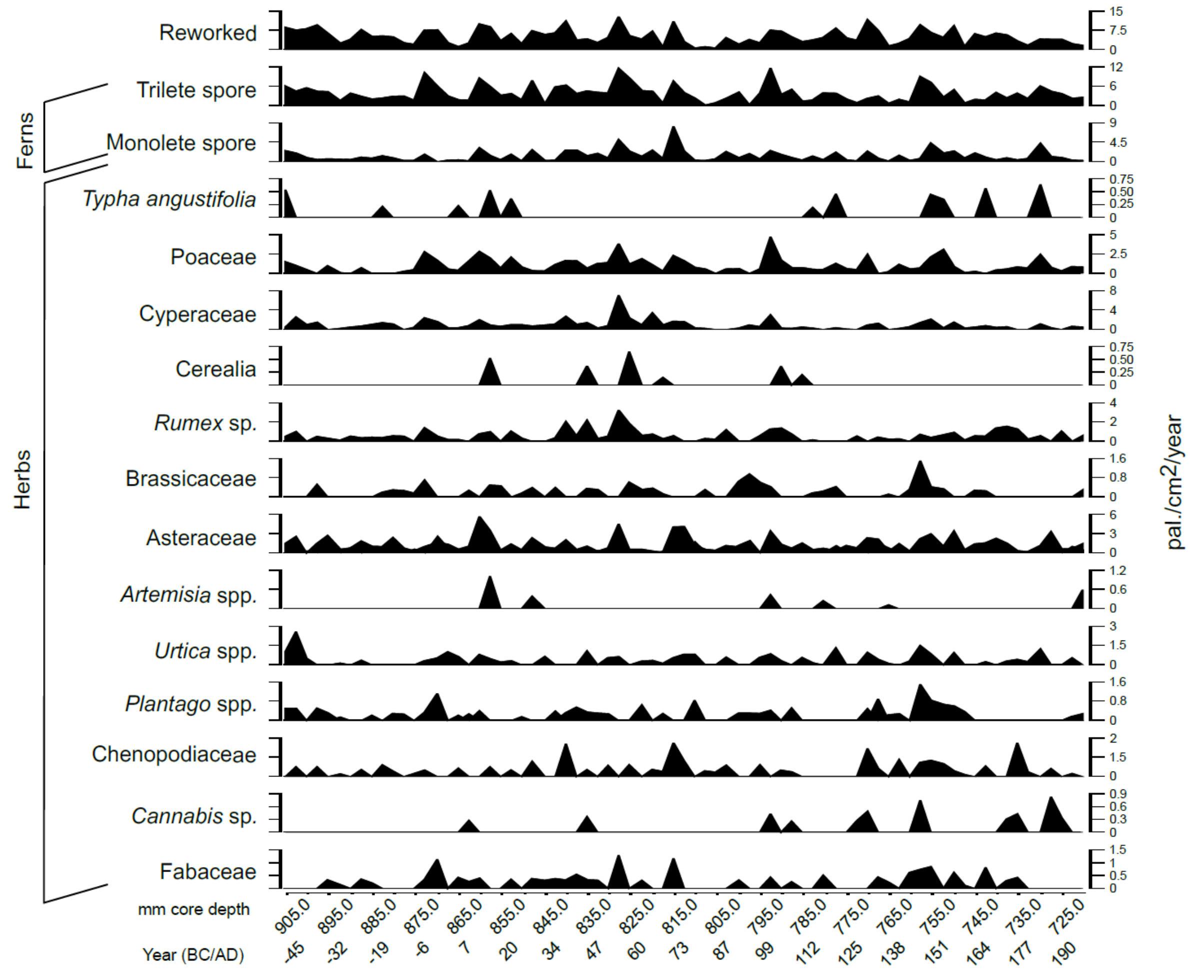
Cyperaceae
(%)
Ephedra spp.
(%)
Plantago spp.
(%)
Rumex sp.
(%)
Typha spp.
(%)
Urtica spp.
(%)
46°N
44°N
42°N
40°N
10°E
15°E
20°E
Ocean Data View / DIVA

A-2 List of stations with similar species composition - Manuscript 1

Code	Characteristic	Samples
A	Po River signal	Moccha 1, Moccha 3, F8CH64, F1CH55, F27CH37, F20CH5, GeoB 17217-2, F44AN33, F51AN35, GeoB 15417.
B	Apeninne river signal	Moccha 5, Moccha 8, Moccha 10, Moccha 13, F36CH50, GeoB 15413, GeoB 17210, GeoB 17211, GeoB 15410, GeoB 15411, GeoB 17207, GeoB 17222, GeoB 17223, GeoB 17216, GeoB 15420, GeoB 10742.
C	Mixed samples	Moccha 22, GeoB 15416, GeoB 17206, GeoB 15412, GeoB 15418, GeoB 15410, GeoB 17203, GeoB 15419, GeoB 10724, GeoB 10725, GeoB 10732, GeoB 10733, GeoB 10739, GeoB 10743, GeoB15401, GeoB 17236, GeoB 15406, GeoB 10707, GeoB 10703, GeoB 10704.
D	Mediterranean/ Sclerophile signal	Moccha 25, GeoB 10726, GeoB 10730, GeoB 10731, Geo B10734, GeoB 15421, GeoB 17201, GeoB 10736, GeoB 10738, GeoB 10741, GeoB 10741, GeoB 10744, GeoB 10713, GeoB 10714, GeoB 10717, GeoB 10718, GeoB 10705, GeoB 10706, GeoB 10702.

A-3 Palynomorph flux - Manuscript 2




Reworked
Ferns
Trilete spore
Monolete spore
Herbs
Typha angustifolia
Poaceae
Cyperaceae
Cerealia
Rumex sp.
Brassicaceae
Asteraceae
Artemisia spp.
Urtica spp.
Plantago spp.
Chenopodiaceae
Cannabis sp.
Fabaceae
mm core depth
Year (BC/AD)
pal./cm2/year

A-4 List of pollen types found in each project

Manuscript 1	Manuscript 2	Manuscript 3
Abies	*Abies*	*Abies*
Acer	*Acer*	*Acer*
Alnus	*Alnus*	*Alnus*
Ambrosia	Apiaceae	*Ambrosia*
Apiaceae	*Artemisia*	*Armeria*
Artemisia	Asteraceae liguliflorae	*Artemisia*
Armeria type	Asteraceae tubiliflorae	Asteraceae liguliflorae
Asteraceae liguliflorae	*Betula*	Asteraceae tubiliflorae
Asteraceae tubiliflorae	Borraginaceae	*Betula*
Betula	Brassicaceae	Brassicaceae
Borraginaceae	*Cannabis*	*Buxus*
Brassicaceae	*Carpinus*	*Cannabis*
Cannabis	Caryophyllaceae	*Casuarina*
Carpinus	*Castanea*	*Carpinus*
Caryophyllaceae	*Cedrus*	Caryophyllaceae
Castanea	*Cerealia*	*Castanea*
Cedrus	Chenopodiaceae	*Cedrus*
Centaurea	*Corylus*	*Centaurea*
Cerealia	Cupressaceae	*Cerealia*
Chenopodiaceae	Cyperaceae	Chenopodiaceae
Corylus	*Ephedra fragilis*	*Citrus*
Cupressaceae	Ericaceae	*Corylus*
Cyperaceae	Fabaceae	Cupressaceae
Ephedra dystachia	*Fagus*	Cyperaceae
Ephedra fragilis	*Filipendula*	*Ephedra dystachia*
Ericaceae	*Fraxinus*	*Ephedra fragilis*
Fabaceae	*Hedera*	Ericaceae
Fagus	*Junglans*	Fabaceae
Filipendula	Monolete spore	*Fagus*
Fraxinus	*Olea*	*Filipendula*
Hedera	*Ostrya*	*Fraxinus*
Junglans	Papaveraceae	*Junglans*
Larix	*Phillyrea*	*Larix*
Malvaceae	*Picea*	Monolete spore
Monolete spore	*Pinus*	*Olea*
Myrtaceae	*Pistacia*	*Ostrya*
Olea	*Plantago lanceolata*	*Persicaria*
Oleaceae spp.	*Plantago mayor*	*Phillyrea*
Ostrya	Poaceae	*Picea*
Phillyrea	*Quercus ilex*	*Pinus*
Picea	*Quercus robur*	*Pistacia*
Pinus	*Rumex*	*Plantago lanceolata*
Pistacia	*Tilia*	*Plantago mayor*
Plantago lanceolata	Trilete spore	Poaceae
Plantago mayor	*Tsuga*	*Quercus ilex*
Poaceae	*Typha angustifolia*	*Quercus robur*
Potamogeton	*Ulmus*	Rosaceae
Quercus ilex	*Urtica*	Rubiaceae
Quercus robur	*Vitis*	*Rumex*

Rosaceae		*Salix*
Rubiaceae		*Tilia*
Rumex		Trilete spore
Salix		*Typha latifolia*
Tilia		*Ulmus*
Trilete spore		*Urtica*
Tsuga		
Typha angustifolia		
Typha latifolia		
Ulmus		
Urtica		
Valeriana type		
Vitis		
Xantium type		

A-5 Additional publication resulting from research during the PhD studies

This collaboration paper, in which pollen were used to establish an age model, is included as an additional publication from research during my PhD studies. For this paper, I processed the sediments for palynological analysis, analysed the slides and the data, wrote the palynological part and helped to establish a reliable age-model based on pollen types across the studied period.

"Historic Development of Heavy Metal Contamination into the Firth of Thames, New Zealand"

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Submitted, *Geo-Marine Letters*

Abstract

Near-coastal marine sediments often provide high-resolution records of various anthropogenic influences such as the release of heavy metals, which pose a potentially negative influence on aquatic ecosystems because of their toxicity and persistence. In places, the gradual onset of man-made heavy metal emission dates back to ~ 4,500 years BP and is difficult to distinguish from potential natural sources. New Zealand offers a perfect playground for studies on anthropogenic impact due to its well-defined three-step development: pre-human era (until ~1300 CE), Polynesian era (~1300-1800 CE) and European era (since ~1840 CE). However, hardly any information exists about the degree of heavy metal input to New Zealand's coastal areas and the 'pristine' natural background values.

This study determines the natural background contents of lead (Pb) and zinc (Zn) in marine sediments of the Firth of Thames, a shallow marine embayment on New Zealand's North Island, and investigates anthropogenic inputs in historic times. Eight sediment cores were analysed by X-ray fluorescence (XRF) for their element composition and temporally resolved by a pollen and radiocarbon-based stratigraphic framework. Sharp increases in Pb and Zn contents occurred simultaneously with the onset of goldmining activities (1867 CE) in the nearby catchment area. The contents of Zn (Pb) increase from very stable values around 60 (13) ppm in the older sediments, interpreted to reflect the natural background values, to an average maximum of 160 (60) ppm near the core top, interpreted to reflect a significant

anthropogenic input. Our findings unravel the history of contamination in the Firth of Thames and provide an urgently needed data base for the assessment of its current ecological state.

Introduction

Anthropogenic influences on coastal marine ecosystems can date back several centuries or even millennia, however, with severely increasing impacts during the last two centuries. This is mainly accompanied by the rising release of various pollutants into the environment. A prominent example is the increasing release of heavy metals by mining and metallurgy, with the beginning traced back to the Copper and Early Bronze Age ~4,500 yrs BP (e.g. Leblanc et al., 2000; Nocete et al., 2005; Cortizas et al., 2016). To understand the sources, the causes and the temporal development of enhanced inputs of contaminants, such as heavy metals, to the marine environment, near-coastal sediment depocentres can provide high-resolution sedimentary archives. These have a great potential for the reconstruction of heavy metal inputs through time and to differentiate between anthropogenic or natural causes.

Pollution history has been decoded in marine sediments (e.g. Irion et al., 1987; Fukue et al., 1999; Hebbeln et al., 2003; Badr et al., 2009; Seshan et al., 2010) from many places in the world. However, only a few studies exist about anthropogenic heavy metal enrichments in sediments from New Zealand. These are mainly concerned with streams (Webster, 1995; Craw & Chappel, 2000; Sheppard et al., 2009; Clement et al., 2017) and more recent releases by urban runoff near major cities (Dickinson et al., 1996; Abrahim & Parker, 2002; Abrahim & Parker, 2008).

New Zealand offers an exceptional setting to study the historical development of human impact, as it was the last main landmass that was colonised by people (e.g. McGlone et al., 1994). Three different stages can be distinguished: (i) the pre-human era until ~1300 CE, with pristine flora and fauna influenced by volcanic activity and natural wildfires, (ii) the Polynesian era with first human settlements and slash-and-burn deforestation starting ~1300 CE (e.g. McFadgen, 1994; Ogden et al., 1998; McGlone and Wilmshurst, 1999; Horrocks et al., 2001), followed by (iii) the onset of the European era due to the "discovery" of New Zealand by James Cook in 1769 with the successive European settlement and the local onset of land-use changes and gold mining at about the same time as the industrial revolution. This very short history of human influences is a suitable precondition to define the regional natural background conditions. In the course of such a short anthropogenic history, the obtained pristine background values are less prone to reflect major environmental changes as e.g. triggered by strong climatic variations or tectonic activities.

Abrahim & Parker (2008) evaluated the heavy metal pollution in the Tamaki Estuary (New Zealand) and found elevated levels of Cu, Pb, Zn and Cd in the uppermost 10 cm compared to 'pristine' values in the older sediments. They linked this contamination to the development of the catchment urbanization and industrialization especially over the past 50 years. Another study in the Wellington Harbour, by

Dickinson et al. (1996), showed post-1900 anthropogenic increases in the elements Cu, Pb and Zn. However, neither of these studies covers the pre-human era and, thus, it is not clear, if the baseline presented shows the actual pristine (pre-human) values, or if the presented values already reflect an impact triggered by the Maori's widespread slash-and-burn deforestation (pre-European baseline). Secondly, both studies concentrate on highly urbanised areas with multiple sources for heavy metal input (e.g. industrial areas, port development, dredging, boat yards and yacht anchorages). Thus, the degree of pollution of New Zealand's coastal areas and 'pristine' heavy metal background contents are still only poorly known.

In this study, we use XRF data obtained on subtidal sediment cores from the Firth of Thames (i) to reconstruct past heavy metal inputs, here Pb and Zn, to the shallow marine environment, (ii) to identify the source for human-induced inputs and (iii) to quantify the impact of anthropogenic change. This is especially important to validate a truly pristine baseline for Pb and Zn in order to put past, current and future heavy metal contaminations in relation to the natural background.

Background

Regional setting

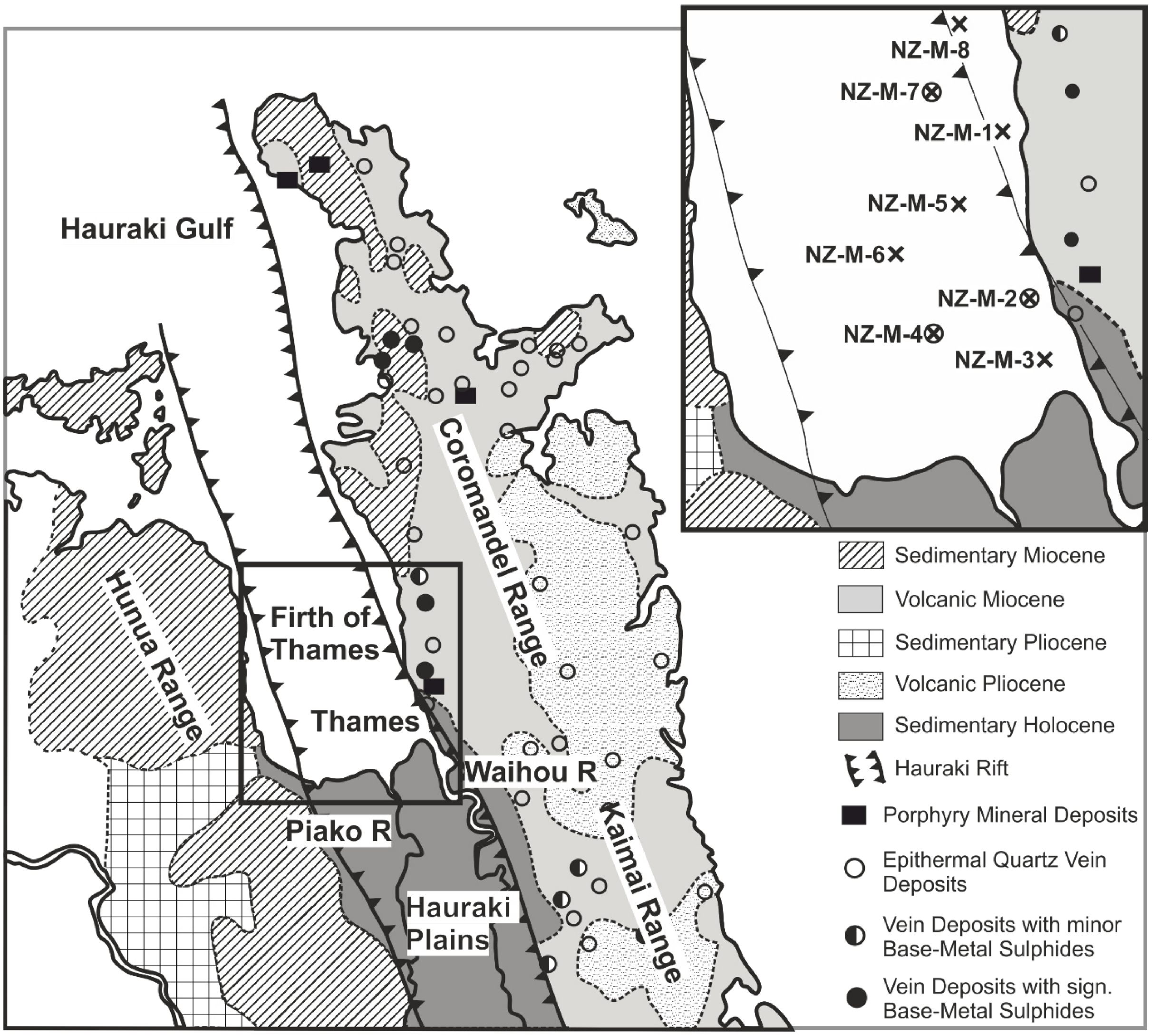


Figure 9.1 Geological map of the Coromandel region, black square indicates the study area (after Livingston 1987 and Dredge 2014). Crosses in insert map mark the core locations, circled crosses indicate the 3 high resolution principal cores NZ-M-2, NZ-M-4 and NZ-M-7.

The Firth of Thames is a mesotidal, shallow estuarine embayment (Naish et al., 1993), located at the north-eastern North Island of New Zealand (Figure 1). It is part of the north-south striking Hauraki Rift, bounded by the Coromandel-Kaimai Range in the east and Hunua Range in the west (Hochstein and Nixon 1979). The rift continues to the south with the Hauraki Plains. To the North, the Firth is bordered by the Hauraki Gulf. Hydrodynamically it is a partially to well mixed, tidally dominated estuary with low-wave energy. On average, tidal currents are ≤0.3 m s^{-1} (Black et al., 2000).

Major sediment supply into the Firth of Thames derives from the Waihou River (160 kt/yr sediment) and Piako River (30kt/yr sediment) (Hicks et al., 2011), which enter the Firth from the South. With a catchment area of 1984 km^2 (Hicks et al., 2011) the Waihou River is the largest river in the Coromandel Region, with several tributaries draining goldmining districts (e.g. Golden Cross, Tui mine) (Webster, 1995).

Human impacts on vegetation

The first major human impact on New Zealand resulted from the widespread forest clearance by the Polynesian settlers. Even though this vegetation change is well recorded in pollen-based paleoenvironmental reconstructions, the exact timing is still under debate and location dependent (McGlone, 1983; Newham et al., 1998; Odgens et al., 1998; McGlone & Wilmshurst, 1999; Byrami et al., 2002, Newham et al., 2018). South of the Coromandel Peninsula (Waihi and Kopouatai), the onset of deforestation is dated to c. 1200-1300 CE (Newnham et al., 1995a, b) and in the following we will use 1300 CE. Byrami et al. (2002) gives a detailed reconstruction of the vegetation composition from pollen records in the Kauaeranga valley, Coromandel Peninsula: (i) in pre-human times, the native forest was dominated by tall trees (e.g. *Podocarpus s*) and no major changes in floral makeup could be detected over a period of 1000 years, (ii) the onset of deforestation during the Polynesian era is indicated by a significant increase in *Pteridium* and charcoal as well as a decrease in forest taxa (tall trees), and (iii) the European era is marked by the occurrence of the exotic *Pinus s* pollen and losses in *Agathis australis*, the New Zealand Kauri tree.

Human impact during European era

With the establishment of European settlements at c.1840 CE (Augustinus et al., 2006), there was a marked change in land use including further deforestation for agriculture, construction works, Kauri logging and gum digging. Kauri logging posed a special threat for the environment, as it not only meant clearance of native forest, but also damming rivers and regularly flush them in order to transport the logs to the coast in the rough terrain.

Another major human impact were the gold mining operations in the Coromandel Range (Thames started in 1867 CE). The Coromandel Peninsula consists of Mesozoic greywacke capped and intruded by Neogene andesites and rhyolites (Williams, 1974; Brathwaite et al., 1989; Adams et al., 1994). Hydrothermal quartz veins cut through these host rocks and scatter small discrete alteration zones with variable gold and base metal content throughout the Coromandel range (Figure 1) (Williams, 1974; Livingston, 1987; Brathwaite et al., 2001).

Materials and Methods

Coring

The sampling campaign was conducted from 20 March to 22 March 2017 on the crane barge *Quest* hired from the company *Bay Marine Works Limited*. A set of 8 sediment cores was collected in the south-eastern part of the Firth of Thames (Figure 9.1 and Table 9.1) using a QR 300 Vibracorer with a total weight of ~250 kg and a tube diameter of 76 mm. The QR 300 Vibracorer was configured to take up to 3 m long cores of unconsolidated or semi-consolidated sediment in a wide range of geological settings. Due to its weight and the very soft sediments in the Firth of Thames, we deployed the QR 300 Vibracorer in a gravity corer mode in order to recover undisturbed sediments from the very top layers.

After cutting the 194-258 cm long sediment cores into 1 m long core segments, they were all split along the c-axis into working and archive halves. All cores were logged immediately after opening with a handheld XRF device (see below). Further analyses focused on three cores (‘principal cores’: NZ-M-2, -4, and -7) from which samples from the working and archive halves were taken at discrete depths for radiocarbon dating, WD-XRF, particle size and pollen analysis (195 samples in total). Archive halves of these principal cores have also been rescanned (prior sampling) with a XRF core scanner at a later stage.

Core no.	Latitude	Longitude	Water Depth (m)	Core length (m)
NZ-M-1	37°02.6894'S	175°30.5168'E	2.6	2.47
NZ-M-2	37°07.1123'S	175°30.6993'E	1.0	2.41
NZ-M-3	37°08.9551'S	175°30.8024'E	0.4	1.94
NZ-M-4	37°08.3227'S	175°26.6615'E	2.2	2.50
NZ-M-5	37°04.6947'S	175°27.9617'E	5.6	2.44
NZ-M-6	37°06.2822'S	175°25.7361'E	4.0	2.58
NZ-M-7	37°01.4124'S	175°27.1250'E	8.5	2.46
NZ-M-8	36°59.6255'S	175°27.7667'E	7.8	2.51

Table 9.1 Information on the eight sediment core locations in the Firth of Thames including mean water depth and recovered core length.

Age determination

Radiocarbon dating

Accelerator mass spectrometry (AMS) radiocarbon dating was performed on bulk sediment organic matter (n=18), unclassified marine shells (n=3), and mixed benthic foraminifera (n=12; Table 2) at the Atmosphere and Ocean Research Institute, University of Tokyo, and at the MICADAS Laboratory, Alfred-Wegener-Institute in Bremerhaven. Single stage accelerator mass spectrometry was used to obtain radiocarbon ages on bulk organic matter (Hirabayashi et al., 2017), with graphitisation completed using the protocol described by Yokoyama et al. (2007, 2010). Sediment samples are pretreated in 1 M HCl for 1 h to remove the calcium carbonate. The sample was heated in a muffle furnace to 850 °C for 2 h. Then carbon that had been incorporated in the sample was recovered as CO_2 (Ishizawa et al., 2017). Pretreatment of marine shell samples followed Yokoyama et al. (2007).

Radiocarbon ages were converted to calibrated calendar ages using CALIB 7.1 (Stuiver et al., 2018) based on the Marine13 dataset (Reimer et al., 2013) and no local reservoir correction ($\Delta R = 0$).

Palynological analysis

19 samples from core NZ-M-4 were prepared for palynological analysis using the standard laboratory procedures by Faegri and Iversen (1989). Sediment (2 cm³) was weighed and dried overnight in a stove at 62°C. Dry sediment was again weighed before decalcification with diluted HCl (10%) and then treated with HF (40%) to remove silicates. One tablet of exotic *Lycopodium* spores (18.583 ± 1.708 spores per tablet) was added to the samples previous to decalcification to allow the calculation of pollen grain abundance. After chemical treatment, samples were wet sieved over a 10 µm nylon mesh using an ultrasonic bath to disaggregate organic matter. An aliquot (50 µL) was mounted on a permanent glass

slide using glycerine jelly and the slide sealed with paraffin wax. Samples were checked for *Pinus, Olea europaea* and *Podocarpus* pollen only.

Grain size analysis

Grain-size measurements on 115 samples taken from the three principal cores were performed in the Particle-Size Laboratory at MARUM, University of Bremen, with a Beckman Coulter Laser Diffraction Particle Size Analyzer LS 13320. Prior to the measurements, the terrigenous sediment fractions were isolated by removing organic carbon, calcium carbonate, and biogenic opal by boiling the samples (in about 200 ml water) with 10 ml of H_2O_2 (35%; until the reaction stopped), 10 ml of HCl (10%; 1 min) and 6 g NaOH pellets (10 min), respectively. After every preparation step the samples were diluted (dilution factor: >25). Finally, remaining aggregates were disaggregated prior to the measurements by boiling the samples with ~0.3 g tetra-sodium diphosphate decahydrate ($Na_4P_2O_7 * 10H_2O$, 3 min) (see also McGregor et al., 2009). Sample preparation and measurements were carried out with deionized, degassed and filtered water (filter mesh size: 0.2 µm) to reduce the potential influence of gas bubbles or particles within the water. The obtained results provide the particle-size distribution of a sample from 0.04 to 2000 µm divided in 116 size classes. The calculation of the particle sizes relies on the Fraunhofer diffraction theory and the Polarization Intensity Differential Scattering (PIDS) for particles from 0.4 to 2000 µm and from 0.04 to 0.4 µm, respectively. The reproducibility was checked regularly by replicate analyses of three internal glass-bead standards and is found to be better than ±0.7 µm for the mean and ±0.6 µm for the median particle size (1σ). The average standard deviation integrated over all size classes is better than ±4 Vol-% (note that the standard deviation of the individual size classes is not distributed uniformly). All provided statistic values are based on a geometric statistic.

Heavy metal analysis

Portable X-Ray fluorescence analyzer (pXRF, energy-dispersive)

An Olympus Innov-X Delta 50 keV Handheld XRF Analyzer gun (Olympus Innov-X 50 KV DP4050CX) manufactured by Olympus was set to "Soil mode" and mounted directly on the smoothed core surface of all collected sediment cores. Sample spots were covered with a thin polypropylene film LS-240-2510 (from Premier Lab Supply Ltd).

Contents of major and trace elements were recorded by Innov-X Delta Advanced PC software. Limits of detection for elements analysed by pXRF are in the ppm range. From a range of elements analysed, here we only use Pb and Zn. Each scan took 60 seconds with calibration checks made every 30 samples on a reference material (Stainless Steel Calibration Check Reference Coin provided by Olympus) for evaluation of accuracy and precision. Expecting anthropogenic impact only in the youngest sediments, sample spacing was 5 cm for the upper 21 cm of the cores and 10 cm further downcore.

X-Ray fluorescence core scanner (energy-dispersive)

XRF Core Scanner data were collected on the three principal cores every 1 cm down-core over a 1.2 cm^2 area with down-core slit size of 10 mm using generator settings of 10, 30 and 50 kV, a current of 0.05 mA (10 kV) and 0.5 mA (30 and 50 kV) respectively. The sampling time was 10 seconds directly at the split core surface of the archive half with XRF Core Scanner III (AVAATECH Serial No. 12) at the MARUM - University of Bremen. The split core surface was covered with a 4 micron thin SPEXCerti Prep Ultralene1 foil to avoid contamination of the XRF measurement unit and desiccation of the sediment. Our data were acquired by a SGX Sensortech Silicon Drift Detector (Model SiriusSD® D65133Be-INF with 133eV X-ray resolution), the Topaz-X High-Resolution Digital MCA, and an Oxford Instruments 100W Neptune X-Ray tube with rhodium (Rh) target material. Raw data spectra were processed by the analysis of X-ray spectra by Iterative Least square software (WIN AXIL) package from Canberra Eurisys and interpreted for Pb and Zn.

X-Ray fluorescence spectrometer (wavelength-dispersive)

28 discrete samples of 20 g wet sediment were taken from the three principal cores at positions that cover the full range of Zn and Pb values according to the XRF core scanner measurements. For calibration of the semi-quantitative XRF core scanning data, samples were kept in the drying cabinet at 40°C for 48 hours and ground using a ring mill. Prior to the analysis, 700 ± 0.6 mg per sample were weighted into a ceramic crucible, 4200 ± 1 mg of a di-lithium tetraborate fusion flux (SpectromeltR A10, Merck) added and mixed with about 1000 mg of ammonium nitrate. Following the pre-annealing at 500 °C overnight, the samples were fused into glass beads and analysed via wavelength dispersive XRF-spectrometry (WD-XRF) at the ICBM, Institut für Chemie und Biologie des Meeres, at the University of Oldenburg. For the determination of precision and accuracy of the method, the in-house standard PS-S was used (average error RMS for Zn (Pb) 9ppm (5ppm); precision in rel-% for Zn (Pb) = 1 (7)).

Calibration of X-Ray fluorescence data

The net intensities for Pb and Zn in counts per second (cps) measured with the XRF Core Scanner were divided by the total cps from the according 30kV run to normalise the data (i.e. for deviations resulting from varying water content, grain size variations and others). These normalised net intensities from XRF Core Scanner data were converted to element contents using linear regressions between XRF scanner-derived intensities and element contents measured on the corresponding 28 discrete samples measured quantitatively (e.g. Jansen et al., 1998; Kido et al., 2006; Tjallingii et al., 2007). All normalised net intensities for Pb and Zn obtained from the XRF Core Scanner analysis were then calibrated using the following linear equation:

$$W_{ij} = a_j\, I_{ij} + b_j$$

In which Wij represents the weight proportion (content) of element j in sample i. Iij denotes the normalised net intensity of the raw spectrum gathered from the XRF Core Scanner analysis. Coefficients *aj* and *bj* are empirical constants specific to the data set and element under consideration derived from the linear correlation. The obtained data of the pXRF were calibrated using the same equation, but normalisation was not conducted as values given by the instrument were already in ppm.

Results

Radiocarbon dating

For all three principal cores (include which ones), the 33 obtained 14C dates show with the exception of a few samples consitent trends to become younger towards the top (Table 9.2, Figure 9.6). 'Outliers' are restricted to either bulk organic matter (blue circles) or not classified shell dates. In the upper 150 cm of the sediment column cores NZ-M-2 and NZ-M-4 comprise ~1,500 yrs and ~1,660 yrs, respectively. Core NZ-M-7 shows slightly older ages and represents 2,380 yrs in 141 cm sediment depth. Therefore, the pre-human era is covered in all three cores. NZ-M-2 is the core with the most radiocarbon dates on foraminifere (n = 8) and all ages above 56 cm show values lying within the last ~250 years. Taking the 1σ into account, no age reversals can be confirmed and all dates potentially reach the time past 1840 CE. Samples below 56 cm become successively older and date back to ~405 yrs CE (maximum 1σ age) in 141 cm depth. The cores NZ-M-4 and NZ-M-7 contain only two radiocarbon dates on foraminifera with no age reversals. In both cases, the deeper sample represents the Polynesian Era (NZ-M-4 at 56 cm = 1438-1508 CE; NZ-M-7 at 26 cm = 1611-1722 CE), whereas the upper sample falls into the European Era (NZ-M-4 at 31 cm = 1692-1822 CE; NZ-M-7 at 16 cm = modern CE). Considering the 14C data from mixed benthic foraminifera (grey circles) only, the obtained ages are generally younger than the 14C ages obtained on bulk organic matter. Radiocarbonages too young for a calibration with CALIB 7.1 were assumed to be 1950 ± 60 CE and marked in Table 9.2 with an asterix. For verification of the general trend, these dates have been compared to data from Naish et al. 1993 (orange circles) from cores very close to our sampling locations (within 5 km distance).

Table 9.2 Radiocarbon dates for the 3 principal cores NZ-M-2, NZ-M-4 and NZ-M-7. Rows in bold are considered as the most reliable 'hard constraints'. [1] - The 14C ages were calibrated using CALIB 7.1 [Stuiver et al., 2018] and the Marine13 data set [Reimer et al., 2013] without a further adjustment for a regional 14C reservoir age (DR = deviation from the average global reservoir age of 400 years); Organic – bulk organic matter; foraminifera – mixed benthic foraminifera; [2] - Here 1 σ enclosing 68.3% of probability distribution [Stuiver et al., 1998]. Values in parentheses are the relative area under probability distribution. - too young for calibration, therefore we assumed CE 1950±60.*

Laboratory Code	Core	Sample Depth (cm)	Sample Material	^{14}C age (yrs BP)	Calendar Age CE [1]	1σ Calendar Age range CE/BCE [2]
YAUT-034228	NZ-M-2	20	Organic	2100 ±28	273	cal CE 232-331 (1.000)
AWI-1529.1.1	**NZ-M-2**	**26**	**Foraminifera**	**563±112**	**1756**	**cal CE 1659-1890 (0.987), 1946-1950* (0.013)**
AWI-1529.1.2	**NZ-M-2**	**26**	**Foraminifera**	**557±103**	**1764**	**cal CE 1667-1885 (0.996); 1949-1950* (0.004)**
YAUT-034229	NZ-M-2	40	Organic	962±30	1393	cal CE 1353-1374 (0.248); 1381-1430 (0.752)
AWI-1729.1.1	**NZ-M-2**	**41**	**Foraminifera**	**modern**	**1950***	**CE 1890-2010***
AWI-1730.1.1	**NZ-M-2**	**56**	**Foraminifera**	**459±104**	**1828**	**cal CE 1760-1787 (0.123); 1803-1950* (0.877)**
YAUT-034231	NZ-M-2	60	Organic	1041±27	1334	cal CE 1300-1356 (0.889); 1372-1383 (0.111)
YAUT- 034702	NZ-M-2	90	Organic	5020±30	-3423	cal BCE 3467-3369 (1.000)
AWI-1530.1.1	**NZ-M-2**	**91**	**Foraminifera**	**1110±105**	**1274**	**cal CE 1194-1386 (1.000)**
AWI-1530.1.2	**NZ-M-2**	**91**	**Foraminifera**	**967±105**	**1385**	**cal CE 1307-1458 (1.000)**
YAUT-034232	NZ-M-2	120	Organic	1366±27	1035	cal CE 1002-1063 (1.000)
YAUT- 034723	NZ-M-2	120	Shell	935±37	1414	cal CE 1387-1454 (1.000)
YAUT-032409	NZ-M-2	139,5	Organic	2536±29	-265	cal BCE 322-214 (1.000)
YAUT-032403	NZ-M-2	139,5	Shell	28854±95	-30465	cal BCE 30755-30222 (1.000)
AWI-1531.1.1	**NZ-M-2**	**141**	**Foraminifera**	**1893±103**	**501**	**cal CE 405-623 (1.000)**
AWI-1531.1.2	**NZ-M-2**	**141**	**Foraminifera**	**1850±112**	**541**	**cal CE 436-660 (1.000)**
YAUT- 034703	NZ-M-4	20	Organic	930±28	1420	cal CE 1401-1450 (1.000)
YAUT- 034724	NZ-M-4	20	Shell	193±38	1950*	CE 1890-2010*
AWI-1731.1.1	**NZ-M-4**	**31**	**Foraminifera**	**550±48**	**1771**	**cal CE 1692-1822 (1.000)**
YAUT- 034704	NZ-M-4	40	Organic	1143±31	1259	cal CE 1234-1291 (1.000)
AWI-1732.1.1	**NZ-M-4**	**56**	**Foraminifera**	**848±48**	**1474**	**cal CE 1438-1508 (1.000)**
YAUT- 034705	NZ-M-4	60	Organic	1041±31	1336	cal CE 1300-1358 (0.867); 1371-1384 (0.133)
YAUT- 034706	NZ-M-4	90	Organic	2351±31	-21	cal BCE 69-cal CE 31 (1.000)
YAUT- 034709	NZ-M-4	120	Organic	1733±35	664	cal CE 632-697 (1.000)
YAUT-031531	NZ-M-4	148,5	Organic	2085±53	289	cal CE 224-368 (1.000)
YAUT- 034713	NZ-M-7	15	Organic	1574±29	813	cal CE 773-866 (1.000)
AWI-1733.1.1	**NZ-M-7**	**16**	**Foraminifera**	**426±48**	**1950***	CE 1890-2010*
AWI-1734.1.1	**NZ-M-7**	**26**	**Foraminifera**	**629±48**	**1674**	**cal CE 1591-1609 (0.063); 1611-1722 (0.926); 1793-1797 (0.011)**
YAUT- 034715	NZ-M-7	30	Organic	1023±36	1352	cal CE 1316-1389 (1.000)
YAUT- 034716	NZ-M-7	55	Organic	1521±41	873	cal CE 806-926 (1.000)
YAUT- 034717	NZ-M-7	80	Organic	1508±36	889	cal CE 833-948 (1.000)
YAUT- 034718	NZ-M-7	110	Organic	1607±32	773	cal CE 722-813 (1.000)
YAUT-031536	NZ-M-7	141,5	Organic	2674±65	-433	cal BCE 521-345 (1.000)

Palynology

Core NZ-M-4 has been analysed for the occurrence of pollen from *Podocarpus, Pinus* and *Olea europaea* with 19 samples taken from 17 cm down to 230 cm core depth (Figure 9.2). For the amount of *Podocarpus s* pollen a general decrease in the number of pollen per gram from core base towards core top was seen, with a slower decline from 230 cm to 72 cm and a sharp drop above 72 cm. The first specimens of *Pinus* pollen occurred in a depth of 53 cm with constant increase to a depth of 22 cm. In the very top sample at 17 cm depth an extensive increase in *Pinus s* is recorded. Additionally, this is the only sample were a specimen of *Olea europaea* was found, another exotic plant to New Zealand, naturalised in 1977.

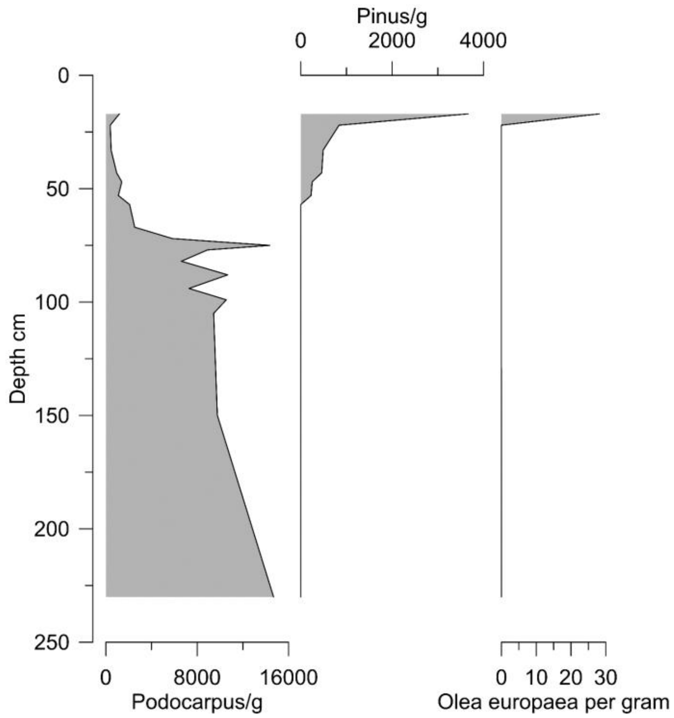


Figure 9.2 Pollen diagram of core NZ-M-4 showing the relative abundances (number per grams) of the counted pollen types (Podocarpus, Pinus, Olea europaea),

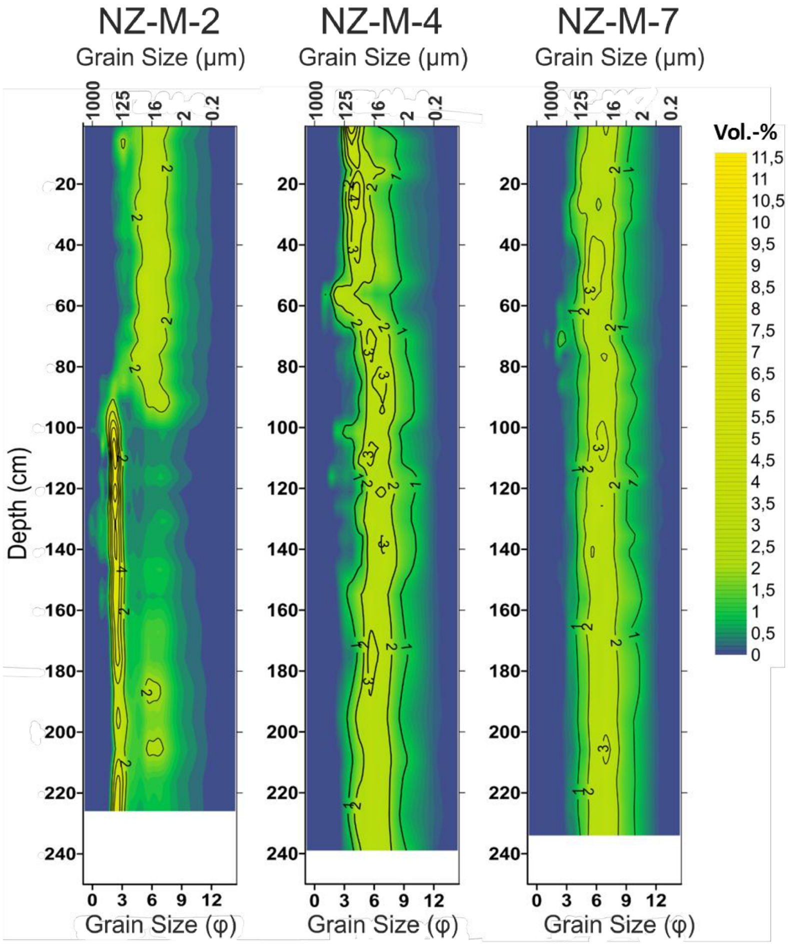
Core description

A strong H_2S smell was obvious after splitting of the cores NZ-M-1, NZ-M-4, NZ-M-5, NZ-M-6, NZ-M-7 and NZ-M-8. These cores consist of a dark greenish mud (Munsell 5Y colour chart) with only minor internal textures, which include thin layers of fine sand or shell debris. No lamination, burrowing or change in colour was observed. Furthermore, no changes in grain size were apparent. An exception are the cores NZ-M-2 and NZ-M-3. Core NZ-M-2 is composed of fine-sandy material in the core section below 96 cm that gradually changed to the same dark olive coloured, homogeneous mud as described before with a minor sand content (<5%) that makes up the uppermost 90 cm of the core. Core NZ-M-3 (near Waihou river mouth) is dominated by black middle to coarse sand with fine shell debris and had a smell of rotten fish. In the sediments below 120 cm depth, the sand alternates with black muddy layers similar to those described from the other cores, but very compact.

Grain size analysis

Core NZ-M-2 displays the already macroscopic observed change in grainsize. From 226 to 91 cm the mean grain size stays constant around 70 µm, fine sand dominated with minor clay content, whereas from 91 cm to the core top the mean grain size values are ± 13 µm (Figure 3). The cores NZ-M-4 and NZ-M-7 consist dominantly of silt and show no changes in grain size with means of ± 14 µm for core NZ-M-4 and a slightly finer mean grain size of ± 9 µm for core NZ-M-7 retrieved somewhat further offshore.

Figure 9.3 Grain size distribution of the 3 principal cores NZ-M-2 (near river, coastal), NZ-M-4 (near river, offshore) and NZ-M-7 (far from river), colour coding gives percentage of each grain size class, varying from 11.5% (yellow) to 0% (blue)



Calibration of pXRF and XRF scanner data

To quantify the pXRF and XRF Scanner data, we calibrated them with WD-XRF data. The correlation plots (Figure 4) with the 28 discrete samples analysed with WD-XRF show that the scanner data have better correlation factors than pXRF data. For the XRF Scanner data Pb (R^2=0.97) generally has a higher correlation than Zn (R^2=0.87), but both show a good fit. For pXRF the correlations with the discrete samples are less significant, but still show a clear positive correlation for Pb (R^2=0.81) and Zn (R^2 = 0.72). The correlation between the XRF Scanner and the pXRF was used as additional verification for data reliability. Correlation factors for the two elements are good and show no difference between Pb (R^2=0.89) and Zn (R^2=0.89). Using the corresponding correlation equations gives higher quantitative Pb and Zn contents for calibrated XRF Scanner data than for pXRF data.

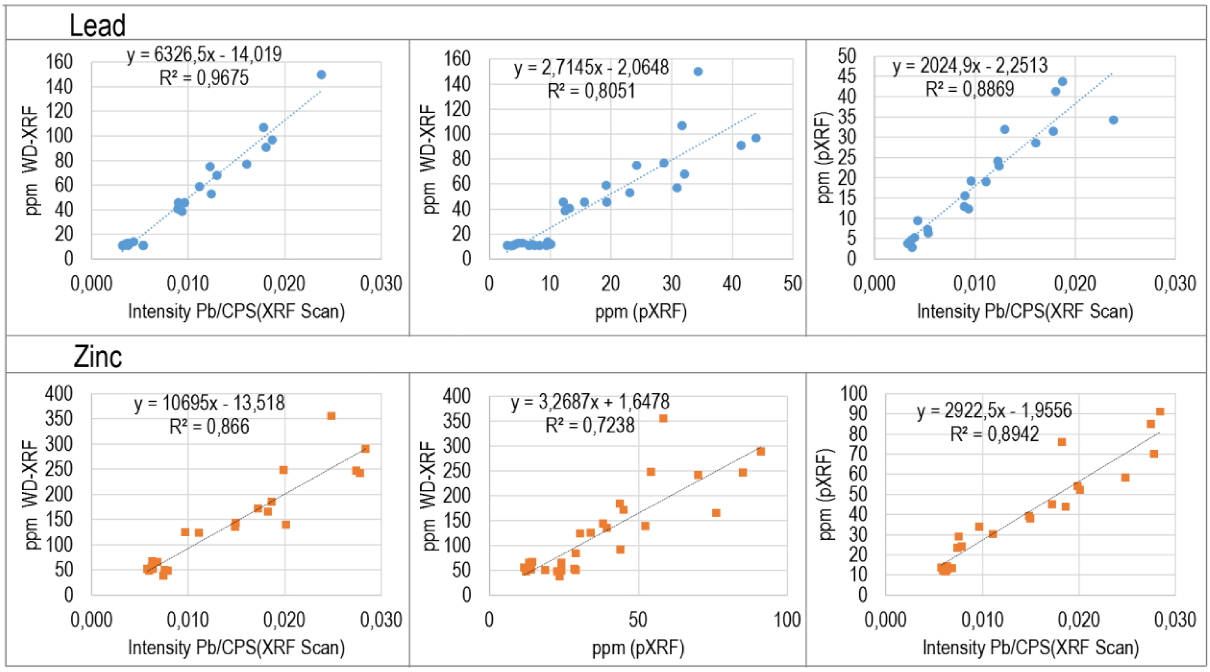


Figure 9.4 Correlation diagrams of the different XRF methods (XRF Scan, pXRF, WD-XRF) and empirically established linear equations for calibrating the XRF Scanner and the pXRF data with the quantitative WD-XRF results.

Calibrated pXRF data

Calibrated pXRF data show a distinct and sudden peak in Pb and Zn in the upper part for 6 of the 8 cores with varying contents and depth of onsets (Figure 9.5). Values below the peaks are very constant within the cores as well as between the cores with values around 12 ppm for Pb and 50 ppm for Zn. Exceptions are cores NZ-M-2 and NZ-M-3 with slightly higher values of 18 ppm for Pb and 80 ppm for Zn in the lower core sections.

Cores NZ-M-2 and NZ-M-4 show very distinct peaks in their uppermost 56 cm and 54 cm, which also show the highest Pb and Zn contents of all cores (Pb up to 90 ppm resp. 120 ppm; Zn up to 190 ppm and 300 ppm). The cores NZ-M-7 and NZ-M-8 located furthest north have less pronounced peaks with lower contents and onsets of the peaks higher up in sediment column. Nevertheless, all these four cores show the same pattern in peak structure with a pronounced increase and a following decrease in Pb and Zn content towards the core top. The Pb and Zn contents of cores NZ-M- 1, NZ-M-3 and NZ-M-5 increase only very close to the core tops (11-16 cm) and no increase at all was detected in core NZ-M-6.

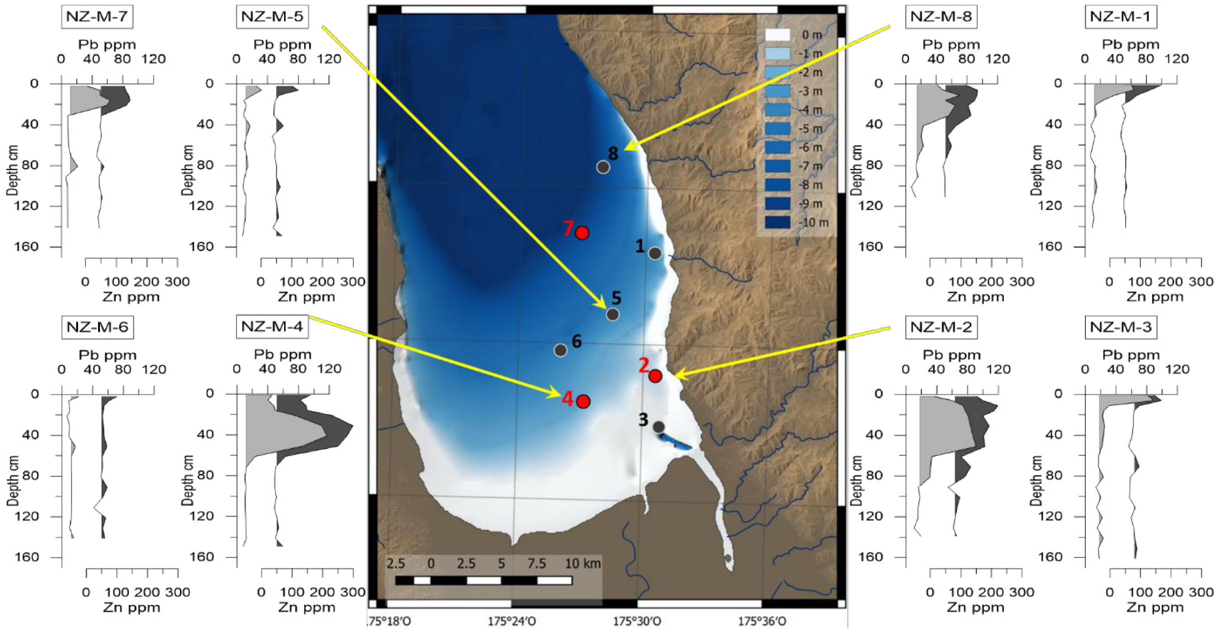


Figure 9.5 Regional and vertical distribution pattern of Pb and Zn contents in the Firth of Thames sediments obtained by calibrating portable XRF data. Circles represent core locations, red circles - 3 high-resolution principal cores, black circles - other cores.

Calibrated XRF-scan data (principal cores)

The temporal and spatial patterns of the pXRF data were confirmed by the calibrated XRF Scan results of the three cores NZ-M-2, NZ-M-4 and NZ-M-7, which, however, provide a much higher temporal resolution (Figure 9.6). In the lower part of core NZ-M-4 rather constant values around 12 ppm for Pb and 57 ppm for Zn can be observed. At 54 cm core depth both values increase steeply to a maximum at 34 cm depths and show from there a general declining trend towards the top of the core. The heavy metal content at the sediment surface are still clearly above the background values found in the lower part of the core. The average content of this section is 82 ppm for Pb and 236 ppm for Zn respectively. Core NZ-M-7 shows a very similar pattern, but with generally lower values and the onset of the strong increase higher up in the core at 26 cm.

Core NZ-M-2 is exceptional, as we see a two-step increase in heavy metals in both independent XRF data sets. Heavy metal values stay very constant at 14 ppm (Pb) and 65 ppm (Zn) from the core bottom up to 93 cm depth. Above this boundary, matching with the change in grain size, the level of Pb (44 ppm) and Zn (101 ppm) is elevated abruptly. This step is followed by a second, even sharper increase to average contents of 100 ppm for Pb and 235 ppm for Zn at 56 cm. Towards the core top, also in this core, Pb and Zn contents decline again but stay the above lower core section values.

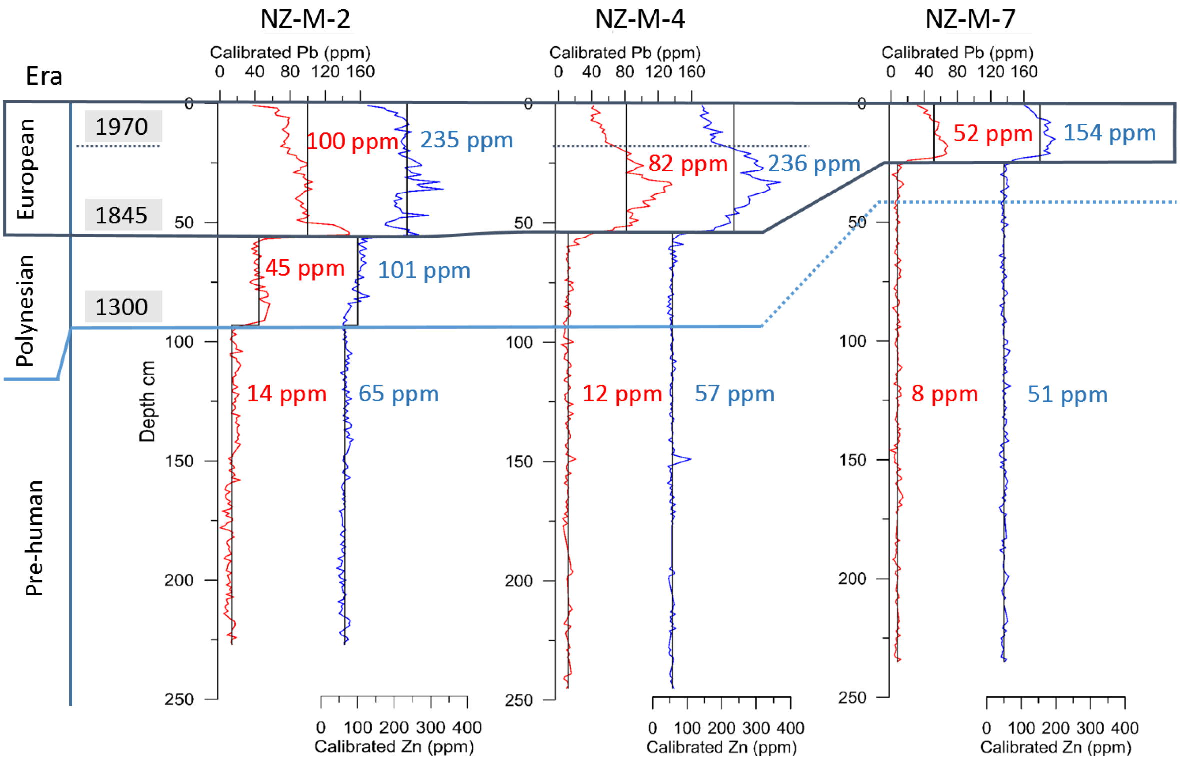


Figure 9.6 High resolution calibrated XRF Scanner data for the 3 principal cores with the interpreted age zonation of the Pre-human, Polynesian and European Era. Averaged Pb and Zn contents were calculated from all values above the peak onset and below the peak onset respectively. Solid lines are based on the stratigraphic frameworks. Dotted lines extrapolated under the assumption of similar relative changes in sedimentation rates between the cores.

Discussion

Development of the stratigraphic framework

Despite the high number of available radiocarbon dates (n=33), the overall scatter of the data makes the development of a stratigraphic framework for the three principal cores focusing on the delineation of the three eras discussed above (Figure 9.7) rather complex. Here, the resulting stratigraphic frameworks are built on three premises: (i) mixed benthic foraminifera give the most reliable ages of the different materials dated and are therefore taken as hard constraints, (ii) the first occurrence of exotic pollen is a distinct marker for the beginning of the European era, and (iii) radiocarbon dates based on organic matter can serve only to constrain maximum ages (soft constraints). Ages obtained from bulk organic matter are known to be generally older than the time of actual deposition, as they have long residence times, depending on the size, morphology and climate of the catchment area (Blaauw et al., 2011). Shells were only regarded if their dates fitted into the overall pattern, as especially in the very shallow setting investigated here these are prone to redeposition. Thus, in a first step the age-depth relationship attained from radiocarbon dating on Foraminifera by interpolation is accepted as the most reliable (solid line, Figure 9.7). Minor age reversals covered by the 1□ range could be influenced by bioturbation or some disturbance related to the shallow water depth and are not considered here.

With respect to premise (ii) two additional palynological depth levels were used for core NZ-M-4: (1) at 53 cm, based on the first occurrence of *Pinus s* pollen (1845±25 yrs CE), and (2) a shell at 20 cm depth. The occurrence of *Pinus* pollen in New Zealand lake and coastal sediments is widely used as stratigraphic event that marks the onset of the European era in c. 1845 (Augustinus et al., 2006). Furthermore, the drastic increase in the content of *Pinus* pollen in the sediment at 17 cm depth is interpreted as the first evidence of pine plantations, which were established in the 1970's (Hume & Dahm, 1992). This interpretation is supported by the occurrence of the only *Olea europaea* pollen found in the core, as *Olea Europaea* was naturalised in 1977. This allows for core NZ-M-4 to differentiate the European Era into an historic and recent part (Figure 9.5).

All stratigraphic frameworks, interpreted here solely to differentiate between the European, the Polynesian, and the pre-human eras, were forced to go through 2010 CE at the sediment surface, as this assumption is supported by a modern radiocarbon date from Naish (1990) at the sediment surface of a core taken in less than 5 km distance from our core NZ-M-7. A comparison of our stratigraphic frameworks to published age-depth relationships based on shells and wood fragments for two cores taken within less than 5 km from the cores NZ-M-2 and NZ-M-7, respectively (orange circles in Figure 9.7; Naish 1990) shows a reasonable well agreement of the general sedimentation rates.

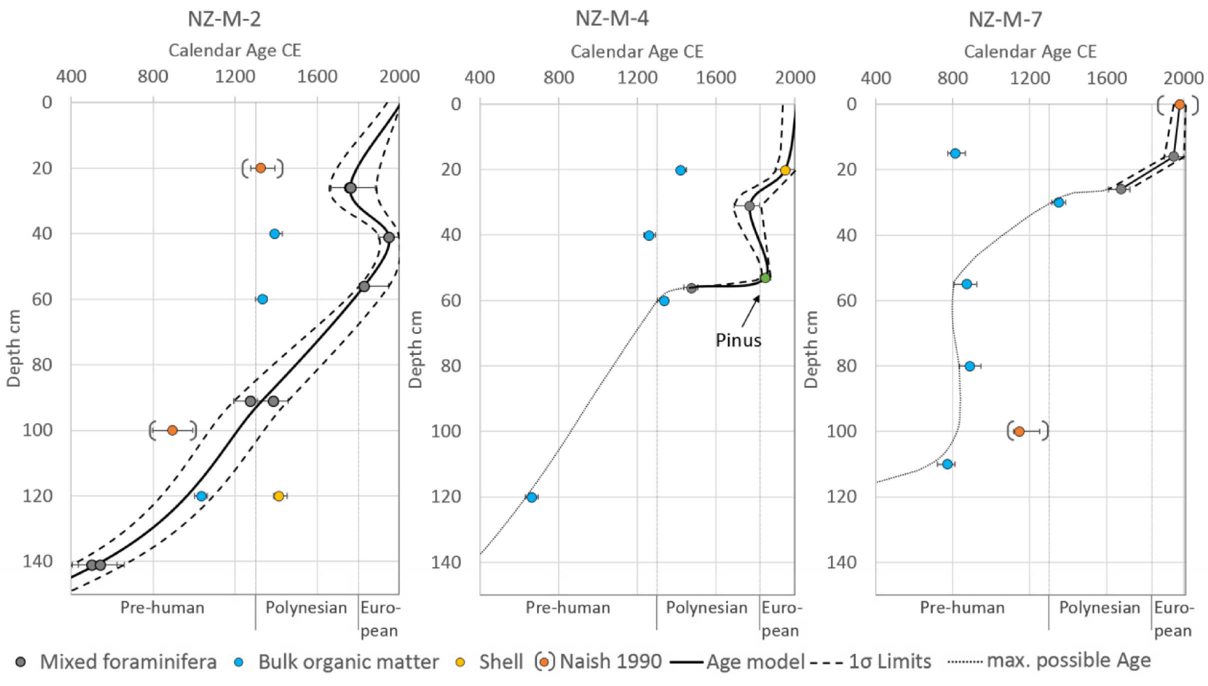


Figure 9.7 Stratigraphic frameworks for the 3 principal cores NZ-M-2, NZ-M-4 and NZ-M-7. Solid black line is the interpolation between the calibrated ages of the values considered as reliable. Dashed black line is the upper and lower limit based on the 1σ range of the reliable data. Dotted line represents the maximum possible age for the lower sections of NZ-M-4 and NZ-M-7. Orange points in () are dates on previous cores in less than 5 km distance from the study by Naish (1990).

Sedimentation rates

A comparison of the resulting sedimentation rates for our principal cores with rates obtained from the same area (Naish, 1990; Hume & Dahm, 1992), shows that all rates are in the same order of magnitude (Table 4). Even though our sedimentation rates are higher than those previously reported, all three data sets document that sedimentation rates were significantly lower during the Pre-human and Polynesian era (< 0.65 mm/yr) and increased with the onset of the European era (up to 3.2 mm/yr). On average, all data indicate an approximately fourfold increase in sedimentation rate upon the beginning of the European era. Additionally, we have indications in core NZ-M-4, that the sedimentation rates in the recent European era are even higher than in the historic one based on the pollen data. Acceleration of sedimentation rates during European times was already suspected by Hume & Dahm (1992) and suggested to be additionally accelerated by the establishment of pine plantations in the 1970's. The sparse sampling resolution in this study does not allow us to differentiate between Pre-human and Polynesian era and, therefore we cannot tell whether the Polynesian deforestation had an impact on sedimentation rates.

Table 9.3 Comparison of sedimentation rates in the southeastern Firth of Thames in mm/yr.

		This study			**Naish (1990)**	**Hume & Dahme (1992)**
		NZ-M-2	NZ-M-4	NZ-M-7		
European	Recent		3.8			
	Historic		2.8			
	Overall	3.2	3.1	1.5	1.9-2.1	0.5 & 1.3-1.5
Polynesian		0.65	n.a.	n.a.	0.35-0.45	0.1-0.4
Pre-human		n.a.			n.a.	0.1-0.2

Timing of increase in heavy metal inputs

For the older parts of the cores, a well-defined stratigraphic framework only exists for core NZ-M-2. Two radiocarbon ages in 91 cm core depth give an age of ~1350 CE, which slightly postdates the arrival of the Polynesians. This is close to the depth level of 96 cm where the sediments get significantly finer and enriched in heavy metals. Interestingly, the two-step increase in heavy metal contents in core NZ-M-2 neither is observed in the other cores (Fig. 9.5, 9.6) nor is the shift in grain size recorded in the other two principal cores (Fig. 3). However, assuming a sedimentation rate for the Polynesian era of ~25% of that corresponding to the European era, all of these cores should cover the change from the pre-human to the Polynesian era (see correlation in Fig. 8 for the principal cores). Consequently, the two-step increase in core NZ-M-2 is interpreted to reflect a local sedimentological change at this near-coastal site (e.g., a meandering tidal channel) that primarily affected the grain size distribution. The accompanying shift towards higher heavy metal contents probably reflects a simple grain-size effect caused by higher loadings of heavy metals of finer particles due to their larger capacity for adsorption (Förstner & Wittmann, 1981).

The coherent pattern with continuously low heavy metal contents through the pre-human as well as the Polynesian era in all the cores (except NZ-M-2) indicates that the Polynesians did not leave any sizable impact on the heavy metal budget in the Firth of Thames. Instead, the sudden increase in heavy metal contents in 6 out of 8 investigated cores (Fig. 9.5) can be correlated with the European era based on the stratigraphic information available for the three principal cores (Fig. 9.7).

In cores NZ-M-2 and NZ-M-4, both located in the inner Firth of Thames, the increase in heavy metal contents occurs in 56 cm and 54 cm core depth, respectively, corresponding to a radiocarbon age of ~1830 CE (NZ-M-2) as well as to the onset of *Pinus s* pollen deposition (NZ-M-4) corresponding to ~1845 CE (Augustinus et al., 2006). In core NZ-M-7 from a more distal position in the central Firth of Thames, Pb and Zn contents increase in 26 cm core depth, radiocarbon dated to ~1670 CE. Despite this somewhat older age, linking the increase in heavy metal contents (Fig. 9.6) with the increase in sedimentation rate (Fig. 9.7) clearly reflects the same pattern as in the other two cores and, thus, points to the impact of the European settlers causing the enhanced deposition of heavy metals also at this site. The comparably old radiocarbon age might be affected by the significantly lower average sedimentation rate at this site that might allow for a larger bioturbation bias in the dating and/or 'smear' the onset of the Pb and Zn signal to some degree into the deeper sediment.

As the grain-size data show no change associated with the increase in heavy metals, any grain size effect can be excluded pointing to a real input signal. Thus, based on the similar pattern of the Pb and Zn contents in the cores, the highest values and thickest affected sediment packages close to the main sediment source, the Waihou River (not considering core NZ-M-3 due to its very different sediment texture, see above), from where they decrease further downstream, and the fit between the radiocarbon and the pollen data defining the onset of the increase, we conclude that the increase in heavy metal input to the Firth of Thames correlates with the onset of the European era.

Furthermore, all three principal cores show a decline in heavy metal content towards the core top (Fig. 6), most pronounced in core NZ-M-4 above maximum Pb and Zn contents at 34 cm. In the same core, we have another time marker, the rapid increase in the number of *Pinus s* pollen and the occurrence of *Olea europaea*, at 17cm depth, reflecting roughly the year ~1970 CE. This indicates, that in the second half of the 20th century the input of heavy metals into the Firth of Thames already subsided, coinciding with a decline in mining activity in the Coromandel region with main operations between 1890's and 1930's (Kidd, 1988).

Further downcore, the boundary of the Pre-human/Polynesian era is covered in NZ-M-2 by foraminifera radiocarbon dates. As there are no reliable ages for the other cores exist, we extrapolated for core NZ-M-4 assuming same sedimentation rates (as it is the case for the European Era). We acknowledge this assumption is rather speculative as there are radiocarbon dates on bulk organic matter indicating the boundary higher up in the sediment core, but argue that they are only a reliable indicator for absolute maximal ages. Extrapolation for NZ-M-7 (dashed line in Figure 9.6) is based on the assumption, that sedimentation rates in this distal location were constantly half of the sedimentation rates close to the Waihou River, as it is the case for the European era. As cores NZ-M-4 and NZ-M-7 show no changes neither in XRF nor grain size, the precise determination of the boundary between the pre-human and Polynesian era is of secondary importance.

Cause of elevated heavy metal levels

The main evidence for the source of pollution is provided by the stratigraphic framework, placing the marked increase in heavy metal contents in the Firth of Thames sediments clearly into the European era. As sedimentary Pb and Zn contents were rather low and constant during the pre-human and Polynesian eras, the increase in heavy metal inputs points to a cause unique to activities of the European settlers.

The prime candidate here is mining. During the early gold mining operations (1850's to 1950's) along the Coromandel Peninsula substantial volumes of mine waste were dumped directly into the Firth of Thames and discharge of tailings into the Waihou River and its tributaries occurred (e.g. Webster 1995, Craw & Chappell 2000). Typical contaminants in these mine tailings are Pb and Zn as well as Cu and As (Hume & Dahm 1992; Christie et al., 2007). As in this region the gold is associated with sulphide-rich sections of hydrothermally altered host rock, these elements were probably released to the Firth of Thames through oxidation and leaching of the mine waste (Garrels & Thompson 1960; Livingston, 1987; Craw and Chappell, 2000). Elevated Cu and As contents were neither detected in the XRF Scan nor in the pXRF data, but there are indications from the WD-XRF that these elements show similar enrichments as Pb and Zn in the Firth of Thames sediments. Most certainly, the Cu and As contents are too low to be detected by the energy dispersive methods (Schnetger, pers. communication).

The strongest enrichment in heavy metals was recorded near the Waihou River (cores NZ-M-2 and NZ-M-4). The Waihou River, and especially its tributaries the Tui Stream and Ohinemuri River are known to be strongly influenced by mining operations (Webster, 1995). In addition, elevated Zn and Pb levels also have been found near the coast of the Coromandel Peninsula, where small streams drain the mining areas. Also the sediments from these streams are characterised by enhanced Zn (106-720 ppm) and Pb (18-85 ppm) contents (Sheppard et al., 2009).

The main period of gold mining operations in the Coromandel range ended with the closure of the Martha mine in 1952 (Christie et al., 2007), although most of the mines were already closed by the end of the nineteenth century (Fraser, 1910; Downey 1935; Sheppard et al., 2009). This is a likely explanation for the observed decrease in Pb and Zn contents in the uppermost sections of the principal cores. While some mines re-opened in a recent mining phase (post-1960), these days' tailings are confined by dams (Craw and Chappell, 2000).

Although these observations clearly point to mining as the most likely trigger for the elevated Zn and Pb contents in the Firth of Thames sediments, it also might be impacted by other processes. These include volcanic eruptions, changes in source area, enhanced erosion in the catchment and urban runoff. As New Zealand, part of the Pacific ring of fire, is characterised by frequently occurring volcanic eruptions also over the last millennium (Lowe et al., 2002; Lowe, 2008), the sudden increase in our Pb and Zn records after low and rather constant inputs for seemingly centuries (Figure 8) does not fit to a volcanic source of the heavy metals.

To link the enhanced Pb and Zn inputs to a change in source area would most likely also require changes in the sediment transport and deposition processes that should be reflected in the grain-size data. However, in the principal cores (except NZ-M-2) the grain-size compositions show no change corresponding to the increase in heavy metal contents (Figure 3). This is further supported by the macroscopic lithological core description of the remaining five cores, with fine sand sedimentation near the Waihou River mouth (NZ-M-3) and homogeneous silt grain size for all others (NZ-M-1, NZ-M-5, NZ-M-6, NZ-M-8).

Interestingly, the lowest Pb and Zn contents in the sand dominated lower part of core NZ-M-2 are comparable or even slightly above the 'background' values observed in the two other fine-grained principal cores (Figure 3), despite of any grain-size effect. However, the coarser grained deposits in this core might derive from a different source area. Still, natural background values for Pb (45 ppm) and Zn (101 ppm) recorded in NZ-M-2 for the Polynesian era are much higher than for cores NZ-M-4 and NZ-M-7 collected further offshore (Pb: 8-12 ppm; Zn: 51-57 ppm). This offset might be due to the close proximity of core NZ-M-2 to the Thames gold deposits immediately onshore. A comparable short transport distance, thus, might have resulted in higher natural Pb and Zn inputs to this site originating from natural erosion.

The discussion, if erosion in New Zealand has increased after arrival of the first humans, is controversial (e.g. Grant, 1985; McGlone, 1989; Wilmshurst, 1997; Ogden et al., 2003). Both, the Polynesian and the European settlers cleared vast stretches of land for agriculture. Nevertheless, on a regional scale around the Firth of Thames the arrival of the Polynesians left no traces in the sediment composition (grains size, Pb and Zn contents) albeit sedimentation rates slightly increased (Table 9.4). Thus, deforestation by the Polynesians probably just mobilised more sediments from the same source area representative for the regional lithology by the same transport processes. The same can be assumed for the effects of forest clearances during the European era done in the context of farming, construction works, kauri logging, and gum digging (Sale & Edmund, 1978; Hume & Dahm, 1992). Consequently, these activities probably increased the quantity of the eroded sediments, but did not affect the composition towards containing more heavy metals. Thus, deforestation probably also had no major impact on the sudden increase of Zn and Pb inputs since the 1840's. Still, we have to acknowledge that there is conflicting evidence in core NZ-M-2, where a distinct natural background value cannot be determined with absolute certainty due to a change in grain size.

Finally, also significant urban runoff from the major settlement areas on New Zealand's North Island was identified as a source for heavy metals. However, being documented to begin in the mid-1920's for Wellington Harbour (Dickinson et al., 1996) and post-1945 in the Tamaki Estuary (Auckland area;

Abrahim & Parker, 2002), urban runoff is not very likely to cause the sudden increment in Pb and Zn contents at c.1845 in the Firth of Thames that is surrounded by only lightly populated areas. In addition, by pinpointing the years around 1970 in our records by increasing *Pinus s* pollen numbers and by the occurrence of *Olea europaea*, heavy metal inputs to the Firth of Thames were already decreasing shortly after urban runoff began to affect the Auckland region. Based on these considerations, gold mining in the Coromandel Peninsula is most likely the dominant cause for an increased release of Pb and Zn during the European era.

Assessment of human impact

Based on the dated cores, we conclude that the onset of enhanced heavy metal input clearly observed in 6 out of the 8 investigated cores from the Firth of Thames occurred at all sites simultaneously approximately at 1845 CE. Since then, enhanced heavy metal input most likely due to historic gold mining in the region, had a widespread effect on sediment chemistry in the Firth of Thames.

Table 9.4 XRF Scanner contents of Pb and Zn in ppm of the surface sediments (topmost value obtained in the sediment core), the averaged peak section (all values from peak onset to core top) and the averaged background section (all values below peak onset) measured on the three principal cores. Additionally Pb and Zn values for the Tamaki Estuary (Abrahim & Parker 2008) and for Wellington Harbour (Dickinson et al. 1996) are listed for comparison. Values in parentheses present the conflicting potential natural baseline which are not comparable because of grain size differenced. EF = enrichment factor comparing peak average and background values.

	NZ-M-2		NZ-M-4		NZ-M-7		Abrahim & Parker (2008)		Dickinson et al. (1996)	
	Pb	Zn	Pb	Zn	Pb	Zn	Pb	Zn	Pb	Zn
Surface Sediment	38	127	42	143	32	109				
Peak average	100	235	82	236	52	154	73	207	104	221
Background	45 (14)	101 (65)	12	57	8	51	22	72	37	128
EF	2.2	2.3	6.8	4.1	6.5	3.0	3.3	2.9	3	1.7

In our principal cores, the baseline contents for Zn and Pb are extremely stable in the pre-European era, suggesting that these reflect pristine natural background values. Except for NZ-M-2 where the natural baseline is not absolute certain, our Firth of Thames background values are significantly lower than those reported for Wellington Harbour (Dickinson et al., 1996) and for the Tamaki Estuary near Auckland (Abrahim and Parker, 2008) (Table 9.5). These differences in background values underline the need for baseline studies in different regions with different hydrological and geological settings.

Table 9.5 ANZECC Sediment Quality Guideline Values for total content of lead and zinc

	Pb	Zn	Effects
Category 1	< 50 ppm	< 200 ppm	Negligible
Category 2	50-220 ppm	200-410 ppm	Possible
Category 3	> 220 ppm	> 410 ppm	Expected

According to the Australian and New Zealand Environment and Conservation Council (ANZECC) Sediment Quality Guidelines (Simpson et al., 2013), the Pb and Zn contents of the surface sediments are below the Sediment Quality Guideline Values (SQGV) for Category 1 indicating negligible effects (Table 9.6). However, as all three cores show Zn and Pb peaks in the subsurface and decrease again towards the core top, Category 1 thresholds have been passed in the past. Taking the average peak values (Fig. 9.6), the Pb contents of all three principal cores and the Zn contents of two of them fall into category 2, meaning ecological effects are possible. Even though Pb and Zn contents in the surface sediments are below the threshold for Category 2, the higher contents in the subsurface imposes potential threats for the ecosystem in case of resuspension (e.g. dredging, bioturbation). As we consider the pXRF data as less reliable, we will not evaluate the heavy metal contents in the other cores with regard to the ANZECC Sediment Quality Guidelines, but remark that also for these cores Category 2 thresholds might be passed.

Conclusion

From the results presented we draw the following conclusions:

- A significant increase of Pb and Zn contents occurred with the onset of the European Era and was caused by mining.
- The sedimentation rates during this time period increased simultaneously, but might not only be related to the historic gold mining operations.
- The impact on the Pb and Zn contents of the bulk sediment is widespread in the southeastern Firth of Thames.
- Most Pb and Zn surface sediment contents are according to ANZECC below the guideline values, but exceed them in the subsurface.

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