

PROVENANCE RESEARCH ON 14TH-CENTURY GREENWARES FOUND IN SINGAPORE



Greenware sherds recently recovered by the NSC Archaeology Unit at the Victoria Concert Hall excavation site

Roeland Stulemeijer



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Provenance Research on 14th-Century Greenwares Found in Singapore

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1. Introduction

This research emerged from the idea that there must be a more scientific way to assess provenance of ancient ceramics than the normal method of observation with the naked eye employed by most experts. These thoughts developed over nearly fifteen years of object conservation work in Singapore and were amplified by the volunteer work I have done with the Singapore Archaeological Laboratory. This research is meant to strengthen the knowledge of an active port-of-trade in the 14th century on the island of Singapore, not much different from present-day Singapore.

This research explores the possibilities of employing EDXRF (Energy-Dispersive X-ray Fluorescence Analysis) to obtain an enhanced insight into the greenware ceramic assemblage uncovered from the excavated 14th-century layers at Empress Place and the Singapore Cricket Club, Singapore. The scientific testing conducted for this thesis is aimed to provide repeatable tests with a dataset, which can be analysed to investigate provenance questions. The 130 tests conducted for this research have been compared with each other in order to find commonalities or differences in chemical fingerprints. The 130 objects have been put in a sorting system; though this system is subjective, no pre-testing sorting system has been described in previous literature on the chemical analysis of ancient Chinese porcelain. This sorting system will allow us to compare the different test results with each other in unique ways. Test results show great promise when comparing chemical elements and displaying the results in graphs. Though more research will be needed to draw absolute conclusions, results of this research show that employing EDXRF for provenance research to solve problems regarding ancient trade in Singapore is a viable option.

2. Context of the Research

In order to place this research within its context, it is necessary to define its geographical location, chronological period, and scientific genre. The field of this research is multi-disciplinary; thus, its intellectual boundaries must be defined in conformity with the questions which this thesis proposes to address.

3. Research Questions

Is it possible to distinguish chemical fingerprints of Chinese Greenware of the late Yuan Dynasty (roughly 1300-1368) with the use of Energy Dispersive X-Ray Fluorescence (EDXRF)? Is it possible to detect clustering of different chemical fingerprints of the celadon sherds found in Singapore? Can EDXRF be an indicative procedure for research on the specific provenance of the kiln sites where ancient Chinese celadon found in Singapore was produced?

4. Research Placement

4.1. *Geographical scope of this research*

Scientific archaeological excavations and analyses have been conducted in Singapore since early 1984. Over the last 20-plus years, ten excavations and a number of surveys have been conducted on the island of Singapore. The majority of the excavations have been executed within the boundaries of what is known as the “civic district” of Singapore. To be more precise, the excavations have been conducted within the area bounded by Stamford Road in the northeast, Fort Canning Park in the northwest, the Singapore River in the southwest and the Esplanade in the

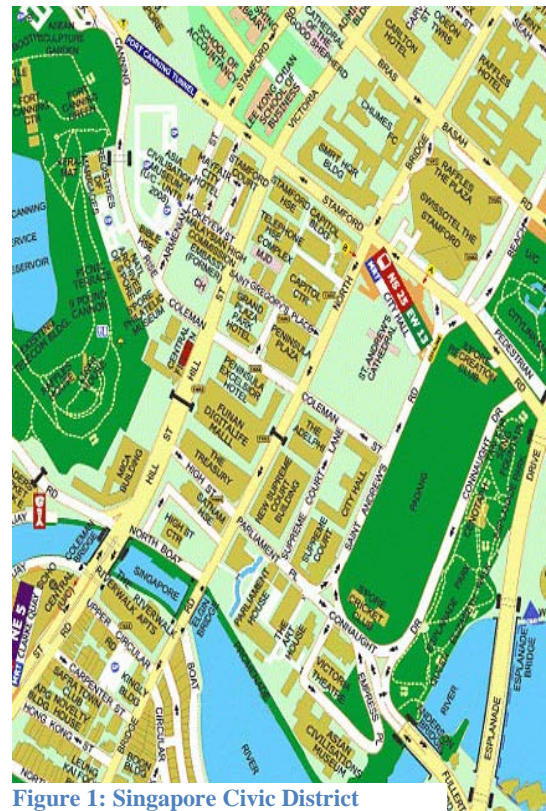


Figure 1: Singapore Civic District
streetdirectory.com

southeast. This land area of approximately eight-five hectares (Miksic, 1985) is likely the same area where in the 14th century a settlement existed. Ancient Singapore was not unlike present-day Singapore; it was an important trading post for likely regional and but certainly long-distance trade.

4.2. *Historical Context*

The archaeological data currently available at the Singapore Archaeological Laboratory (S. A. L.) situated on Fort Canning suggest that within the boundaries of the “civic district” on the island of Singapore, there was a well-used port of trade in the 14th century. According to Miksic (1985), Shah Alam bin Zaini (1997), and Heng (2005), archaeological data point

towards the likelihood of a significant settlement in this area. Miksic and Low (2004) describe the historic data and references to Singapore from 1300 to 1819. This collection of historic articles is to date (2008) the most complete published work on the ancient history of Singapore.

In the text on Singapore by the Chinese traveller Wang Dayuan, interesting occurrences are mentioned. One of these is that during the Yuan dynasty (1280-1368) a diplomatic or commercial mission from Long-ya-men was sent to China in 1320. Interestingly later in his texts, Long-ya-men is described as a place where the people are addicted to piracy. Wang Dayuan mentions that: "The natives and Chinese dwell side by side." A note that a Chinese envoy was dispatched to that same location to buy tamed elephants creates an interesting combination of information, which is open to a range of conclusions. To send a diplomatic or commercial mission to China must mean that the people at Long-ya-men were sufficiently well-organised to send a committee to travel afar to enhance the local community, and ensure long-term relationships with a powerful foreign neighbour. With piracy they assured themselves of a short-term income. However, as this research focuses on the other inhabited area, 15 kilometres to the east, the conclusions that can further be drawn about this site are left to the reader.

The other settlement, of which the location is illustrated above (page 8), was described by Wang Dayuan as Pan-Tsu (Wheatley 1961) where the people were honest. As Rockhill wrote (1915: 129-132, in his translation of the *Daoyi Zhilue*).

This locality is the hill behind *Lung-ya-men*. It resembles a truncated coil. It rises to a hollow summit, [surrounded by] interconnected terraces, so that the people's dwellings encircle it. The soil is poor and grain scarce. The climate is irregular, for there is heavy rain in summer, when it is rather cool. By custom and disposition [the people] are honest. They wear their hair short, with turbans of gold-brocaded satin, and red oiled-cloths [covering] their bodies. They boil seawater to obtain salt and ferment rice to make spirits called, *ming-chia*. They are under a chieftain. Indigenous products include very fine hornbill casques, lakawood of moderate quality and cotton. The goods used in trading are green cottons, lengths of iron, cotton prints of

local manufacture, *ch'ih chin* (already obsolete coinage, when Dayuan wrote his remarks), porcelain ware, iron pots, and suchlike.

These historical descriptions are thus far the only direct descriptions of the two known inhabited locations in ancient Singapore. Both locations seem to have thrived on trade, either regulated or unregulated. Important is the mention of porcelain ware being used in trade. Greenware is in Chinese a porcelain ware; in Chinese there are only words for Earthenware (Taoqi 陶器), and Porcelain (Ciqi 瓷器). In the Modern Chinese language, there is another term for the modern concept of "stoneware" (shiqi 炆器)¹.

It is unknown whether there was one or there were several production centres exporting their Greenware products to Singapore in the fourteenth century. This research will likely be able to identify if the Greenware objects found in Singapore were produced in several areas in China or only one. If there was one production centre trading its wares through and in Singapore, it is likely that the other products named by Wang Dayuan were also from or traded with one area in China.

14th-century Singapore seems to have been unique in other ways in addition to its geographical location. There seem to be no similar ports-of-trade in the region fulfilling the same purpose as Singapore. There were vaguely similar locations earlier and later but none at the same time as Singapore. Kota Cina in North Sumatra played the same role around the turn of the first millennium, as did Palembang in South Sumatra, though more up river, not so near the coast in the 7th through 12th Century, and then later there was Malacca in the late 14th early 15th Century. Nor were there similar places in Vietnam, the Philippines, Siam (Thailand), or Majapahit (Indonesia).

For local trade, by which I mean trade within the Southeast Asian region, little is known. As A. B. Lapian (1985) mentions, the Orang Laut, though not a term preferred by these 'people of the sea', were an extensive group of people who lived their lives on or next to the sea. They could be found from Thailand to Malaysia, Sumatra, Java, Singapore, Kalimantan,

¹ Information and Chinese characters provided by Sharon Wong Wai Yee PhD.

Sulawesi, the Southern Philippines, and further east to the Moluccas. He further says that most of these people show a certain cultural homogeneity and seem to share similar linguistic characteristics; one cannot yet determine the specific or general nature of their ethnic affinities. This makes it difficult to earmark the Orang Laut as 'one people' or a single ethnic group.

The Southeast Asian chiefdoms, and likewise organised communities, produced few commodities that have stood the test of time; hence little archaeological evidence of trade within the region has been found (see Miksic and Yap 1992.). Until more archaeological research has been conducted, and analysis using provenance testing has been employed, information on inter-regional trade will have to come from written sources; to date only Chinese and Vietnamese ancient texts shed a dim light on this trade. Thus far, the conclusions drawn on trade within Southeast Asia have been derived indirectly from these texts.

4.3. Archaeological background

The artefacts used for this research were all found in the archaeological strata defined by the archaeological data as dating from the 14th century on Fort Canning, and continuing into the late 16th century at the sites found on the flatland between the hill and the former shoreline. This era in Chinese history is defined as the dynasties of late Yuan (1280-1368) and early Ming (1368-1644). It is probable that the sherds recovered from these strata were all produced in this period. The conclusions of this thesis will be presumed to refer to that period, during which similar chemical fingerprints and a narrowly defined cluster of dates should logically indicate common places of production for the ceramics, which form the basis of this research. The chemical fingerprint analysis might be able to provide us with a better insight into archaeological site specific demand within the 14th-Century Singapore context.

Does the material excavated at the Empress Place (EMP) site have a large scatter of chemical fingerprints in the graphs compared to that obtained at the Singapore Cricket Club (SCC) site? Will the analysis of the chemical fingerprints provide us with some greater

insight into the provenance of the artefacts? Will we be able to distinguish ceramic production centres, or better yet, individual kiln sites, by the use of graph analysis?

4.4. Scientific Context

Within the geographical and historical context of this research, the scientific boundaries of the disciplines to be employed must also be specified. Although this research is not unique in its way of testing materials nor in the mode by which it conducts a search for provenance of the objects, it is however unique in the singular object type on which it is focused within the broad range of artefacts and materials from the overall site of ancient Singapore.

The scientific context of this research is defined by the use of Energy Dispersive X-Ray Fluorescence (EDXRF) as applied exclusively to ancient Chinese Greenware ceramics. Hence, conclusions drawn from it do not enable us to compare the results with other types of ceramics. My research is focussed on the analysis of Greenware ceramics in order to find chemical characterization clustering by acquiring the chemical fingerprints of the Greenware through the use of EDXRF.

4.5. Greenware

The S. A. L. has conducted ten excavations within the civic district of Singapore as stated above; all these excavations have yielded a diverse assemblage of artefacts and materials, including metals, ceramics, glass, and few other materials in slightly different proportions per excavation. As ceramics in Singapore are the most numerous types of the recovered artefacts, description of them requires division into more specific categories. The ceramic assemblage consists of Earthenware, which can be further subdivided between coarse and fine earthenware, Stoneware, which is subdivided into Storage jars and mercury jars, and then Greenwares, and Porcelain. These categories can in turn be further broken down into yet smaller types, based on material, shape, size, colour, and numerous other relevant traits.

The research objects studied in this thesis are usually described by scholars with one of two different words, Greenware or Celadon. Before distinguishing between these terms it is

prudent to provide a more in-depth identification of the type of ceramic object, confusingly described by both words.

The Greenware clay consists of a gradient of mixtures of porcellaneous kaolin clay and iron-rich stoneware clay. The combination ratio differs along a spectrum producing a wide range of light whitish grey to a dark grey and even to a pinkish brown clay colour. On page 41 of this thesis the range of colours is illustrated.

The glaze colours described as either Greenware or Celadon consist of an Iron-rich glaze. The objects were fired in a reducing kiln environment, which means there is no oxygen added during the firing process, creating a chemical reaction causing the glaze to turn into a variety of green colours. The green ranges from a light bluish green to a deep dark green and to a light yellowish green.

The term Greenware is the preferred term as it is all-inclusive whereas the name Celadon refers only to the pale green glaze colour. For the origins of the name Celadon, there are two distinct theories. The first is that the name has a French origin. Honoré d'Urfé's romantic play or novel *L'Astrée* (1627) depicted a shepherd, named Celadon, wearing light green clothes, which resembled the colour of the ceramic objects. Honoré d'Urfé had likely used the shepherd's character from a previous Roman work by Ovid, named 'Metamorphoses' (Hobson, 1936). The other theory is that of Savage (1954) who noted that the name celadon needs explanation, since it is inaccurately defined in some older works. He proffered that the name is probably a corruption of Salah-ed-din (Saladin), Sultan of Egypt, who sent forty pieces of this ware to Nur-ed-din, Sultan of Damascus, in 1171.

Willits (1997:33) has some additional thoughts about the name and its provenance;

The most recent instance of its use recorded in the big Oxford English Dictionary of 1933, dating from no later than 1877, is in a couplet by Henry Wadsworth Longfellow, where it still means a colour: "A sky just washed by gentle April rains/And beautiful with céladon", wherein the original accent aigu of the 'é' of the shepherd's name is retained. Had Longfellow perhaps been

reading the classical Chinese description of Ch'ai ware, "blue as the sky after rain"? He had certainly read Père d'Entrecolles, as plainly appears in his poem *Kéramos*.

The etymology by Savage seems rather difficult to correlate with the name Celadon where Wadsworth's definition seems to identify a certain colour derived from either the French or Chinese source. It shows that the name Celadon is far more confusing and limiting than the name Greenware. The all-inclusive name of Greenware is thus the preferred name to be used in the scholarly study of this beautiful ware.

The Greenware material is the most logical choice for this study since it is always represented in the excavations in Singapore. The Greenware assemblages seems to consist of several types of shapes, including small bowls, saucers, little jars, small plates, large bowls, and large plates. The large array of shapes of Greenware could mean that there were several Chinese production centres providing Singapore with Greenware products. The possibility of a number of production centres makes Greenware a good candidate for provenance research. This type of material is likely to give a picture of the widest range of links between Singapore and parts of China. Materials such as Whiteware and Blue and White porcelain are less indicative of trade, as they were produced in few locations. Little research has been conducted on Whiteware, and thus, little is known about its production sites. As for Blue and White, much is known about this type of ware but for production sites thus far our knowledge seems to focus on Jingdezhen. Little is known about other locations, like in Yunnan province where there is thought to have been a Blue and White production centre, but no conclusive evidence has yet been brought forward.

CHAPTER 2

5. Historic Background

5.1. *The Fourteenth Century in Southeast Asian History*

During the 14th century, Southeast Asia experienced far-reaching changes: cities were larger than before, the financial sector became more organised, large Chinese overseas communities were established, and the Islamic faith expanded throughout Southeast Asia. Evidence from excavations in Singapore demonstrates that the island was occupied in the beginning of the Yuan dynasty as many pieces of Chinese ceramics from this period have

been excavated. The Mongol dynasty that ruled China until 1367, was more tolerant toward maritime trade than its Chinese predecessors were, and this is the early period of the success of Singapura. The quality and the large number of Chinese ceramics found in Southeast Asia suggest that the trade between China and Southeast Asia peaked during this period (Wheatley 1961, Wolters 1982). In addition, the likelihood of these ceramics having their provenance in several provinces is high as there are multiple port cities along the Chinese south-eastern coast. Ancient cities like Canton, Quanzhou, and Fuzhou are possibly providers of trade with Southeast Asia.

Other references naming Singapore in the 14th century are the *Sejarah Melayu* and Javanese fourteenth-century court poem Desawarnana. It has to be said that one cannot take the *Sejarah Melayu* literally (Wolters 1970) as it was revised from generation to generation (Roolvink 1967), but Raffles' conclusion that the general image of Singapura as an active port town was factual has been substantiated by references in Javanese, and Chinese references of the same era. Chinese sources of the 14th century do not use the name Singapura for the island but employ names like Pancur (Wheatley 1961), Temasek (Wolters 1982), and Longyamen ("Dragon's Tooth Strait") (Gibson-Hill 1954).

The early 14th-century Chinese explorer Wang Dayuan mentioned that there were Chinese residents in the territory of Temasek, which had a local ruler and government. In addition, he mentioned that just a few kilometres away there was a pirates' lair named Longyamen (Rockhill 1915). These observations could mean that the regional Malay community transported goods around the region. There are several shipwrecks discovered in recent years, which display characteristics of Southeast Asian shipping traditions, from the Philippines to the Malay Peninsula to the Java Sea (Manguin 1993). It is likely if there were Chinese in the 14th-century Singapore context, there would have been Chinese in other locations in Southeast Asia.

5.2. Singapura's Strategic Location

The Straits of Malacca, a waterway separating the east coast of Sumatra from the west coast of the Malay Peninsula, has been an important maritime passage throughout history, linking the Indian Ocean to the South China - and Java Seas. The pattern of monsoon winds

also made the Malacca Straits region a natural meeting place where sailing vessels could await the change of the monsoon wind direction, and merchants from East and West could exchange goods. Numerous ports have emerged along the coastlines bordering the Straits throughout history to capitalize on the shipping and trade that congregated in or passed through the region (Heng 2004).

From the middle of the first millennium CE on, the Malacca Straits region was able to capitalize on the region's advantages. Between the late 7th and the late 13th centuries, the region was under the leadership of Srivijaya, a port-polity located at Palembang and from the 11th century in Jambi near the south-eastern coast of Sumatra. The region functioned as a hub for shipping and trade between the Indian Ocean and the South China Sea, and transhipped foreign products in high demand in the Chinese, Indian, and Island Southeast Asian markets (Wheatley 1961).

John Crawford's Description of the Ruins of Ancient Singapore

From *Journal of an Embassy from the Governor-General of India to the Courts of Siam and Cochin China* (London, 1828), PP. 44-7. (Wheatley 1961)

February 3 [1822]. -I walked this morning round the walls and limits of the ancient town of Singapore, for such in reality had been the site of our modern settlement. It was bounded to the east by the sea, to the north by a wall, and to the west by a salt creek or inlet of the sea. The inclosed space is a plain, ending in a hill of considerable extent, and a hundred and fifty feet in height. The whole is a kind of triangle, of which the base is the sea-side, about a mile in length. The wall, which is about sixteen feet in breadth at its base and at present about eight or nine in height, runs very near a mile from the sea-coast to the base of the hill, until it meets a salt marsh. As long as it continues in the plain, it is skirted by a little rivulet running at the foot of it, and forming a kind of moat; and where it attains the elevated side of the hill, there are apparent the remains of a dry ditch. On the western side, which extends from the termination of the wall to the sea, the distance, like that of the northern side, is very near a mile. This last has the natural and strong defence of a salt marsh, overflowed at high-water, and of

a deep and broad creek. In the wall there are no traces of embrasures or loop-holes; and neither on the sea-side, nor on that skirted by the creek and marsh, is there any appearance whatever of artificial defences. We may conclude from these circumstances, that the works of Singapore were not intended against fire-arms, or an attack by sea; or that if the latter, the inhabitants considered themselves strong in their naval force, and therefore thought any other defences in that quarter superfluous.

Crawford's 'ancient' town of Singapore was bounded in the southeast by the sea; this has been filled in and marked with a road, Beach Road and the Padang. The town to the northeast had an earthen wall; this northern wall has been removed and its former course is now known as Stamford Road. The function of this earthen rampart, as Crawford called it, was likely a defensive wall but as Wang and Crawford both described a marsh on the outside of the wall which would flood at high tide, it could also be considered a defence against the tide not necessarily attacks. The height of the wall does suggest it to be a defence against attacks but one could ask why anyone would attack a settlement through a marsh? Low tide likely did allow for a limited attack, which could make the wall a defence against both enemy and tide.

To the southwest the settlement was bound by a salt creek or inlet of the sea; this creek is now known as the Singapore River. The above description of Singapore is likely similar to what the Yuan Dynasty traveller, Wang Dayuan, must have seen in his days in Singapore. Wang describes Crawford's 'hill of considerable extent' as having a hollow summit, with interconnected terraces on all sides with dwellings occupying them.

The terraces described by Wang Dayuan are also mentioned by Crawford who wrote that "the northern side of the mountain is covered with the remains of the foundations of buildings". It was not surprising that when Dr. Miksic started his first excavation in Singapore, he chose the north side of Fort Canning Hill.

5.3. Archaeological data

In 1984, Dr. John N. Miksic conducted the first scientific archaeological excavation in Singapore. Over the next twenty years he would lead many more teams of volunteers in unearthing parts of ancient Singapore.

At the start of my research the artefacts recovered from the Fort Canning (FTC) excavations were under post-excitation investigation and thus the excavated artefacts are not included in this study. Sherds from the two excavation sites further discussed in this paper are Empress Place (EMP) and the Singapore Cricket Club (SCC). These were used for the purpose of this research, as they were able to supply the number of sherds required to satisfy scientific statistical requirements for such research.

5.3.1. Empress Place (EMP)

This site is currently better known as the location of the Asian Civilisations Museum. The excavation site at EMP was located in between the current museum and the river. This patch of grass was excavated in 1998 and, as shown on the map in figure 2, is just inside the mouth of the Singapore River (covered by the Esplanade Bridge). During the excavation, it became apparent that there were two distinct occupational layers. The first habitation layer (counting from the top) was determined as containing artefacts from the Dutch VOC Company (Dutch East India Company) including coins and Chinese porcelain from the latter half of the 18th century. Artefacts from this era have not been retrieved from any other archaeological site in Singapore.



Figure 2:
Empress Place
Excavation Site street

The second habitation layer was dated from the late 13th to mid-15th century. This was determined by stylistic analysis of Chinese porcelain and comparison with samples of known data found in China. EMP is situated just inside the mouth of the Singapore River. It is likely that this area was a large sandbar that just rose above the water at normal high tide. From the excavation at EMP an estimated 40,000 artefacts were unearthed (Miksic & Low 2004).

5.3.2. Singapore Cricket Club (SCC)

The SCC excavation was conducted for three weeks in February and March 2003. The excavation site on the SCC side of the Padang was likely an archaeologically undisturbed area as the SCC was set up in 1852, shortly after the British settled in Singapore.² The SCC field is a well-maintained cricket pitch; criteria for this entail the requirement that the lawn be resurfaced every few years. In the case of the SCC, there was the added need to fill the pitch to a higher level as it is situated close to the seaside. A low-lying area would flood with high tide or heavy rainfall.

This infill and the years of resurfacing of the cricket pitch have preserved the underlying ancient remains. A rich deposit of artefacts has been excavated at this site, with some interesting discoveries. In the *Sejarah Melayu* it is stated that when Sri Tri Buana looked, north-northwest, toward the island of Temasik he saw the white beach sand glittering in the sunlight. During the excavation in 2003, when the excavating team had removed the habitation layers, the team discovered beautiful white beach sand that indeed glittered in the sunlight as described in the *Sejarah Melayu*. During the SCC excavation, about 40,000 artefacts were unearthed.



Figure 3:
Singapore Cricket Club
Excavation Site

² <http://www.scc.org.sg/>

PART 2

CHAPTER 3

6. Scientific Equipment:

6.1. *Introduction*

In the past, archaeological research was typically done through comparative analysis. This often means comparing the quantity of artefacts of various types found between sites, as well as studying the different kinds of finds between various sites, and the differences in stylistic attributes between artefacts. This latter type of research is usually subjective as the formation of artefact types is normally based on qualitative stylistically analysis and the previous experience of the researcher.

Over the last fifty years, scientific research has taken a prominent role in the analysis of the past. Science-based archaeology, working from the present into the past and a multidisciplinary approach to the archaeological data have been favoured. (Fagan 2003). Owing to the ready availability of technology, the search for more consistent information and the curiosity of physicists regarding the chemical composition of artefacts, research at excavation sites, and the identification of reasons underlying the differing compositions of artefact clusters are now done more frequently through quantitative or scientific means. In other words, scholarship now demands the study of chemical composition of sites and finds through analysing statistical data instead of visual examination or other forms of qualitative methods. These scientific research methods are objective and the results of their tests should be able to be reproduced, making the outcome more reliable than those produced by personal or subjective interpretations do. Bartle (et.al. 2007) has stated that “energy-dispersive X-ray fluorescence (EDXRF) spectrometry and thermoluminescence (TL) dating can result in significant damage to the artefact itself.” This is not always true. Some have noted that it is possible to obtain data on absolute concentrations of elements in an artefact, but this does entail destruction of the sample. This is the only situation in which EDXRF would result in destruction of the sample. Some scholars also argue that absolute concentrations are the ideal measure to use for comparative studies such as that discussed in this thesis. Not all experts agree on this. In fact, it is possible to argue that reliance on absolute concentrations can lead to significant errors. The measurement of absolute

concentrations can increase either the damage to an artefact or the possibility of impurities being measured during the testing. When impurities are included in destructive testing one is forced to redo the test, thereby damaging the artefact yet again or destroying the object. The latter option then could be considered vandalism as the objective should always be to avoid causing damage to any artefact. The use of non-absolute concentrations should be preferred as the tests sample a larger area, which will average out any impurities and are completely non-destructive. Thus some scholars (such as Miksic and Yap) have chosen to use relative rather than absolute concentrations for their analysis.

Thermoluminescence, C14 testing, and petrography are examples of scientific tests that have been employed to identify different aspects of artefacts. However, none of these scientific methods produces chemical fingerprints, which makes them obsolete for provenance research. The first two tests cannot be done in Singapore, as there is no such equipment available. Petrography is the science of identifying different mineral inclusions in the clay/rock. This is of no use for this research. Chinese porcelain contains no discernible inclusions; it is made almost entirely of kaolin, Al_2SiO_3 .

Provenance research has become an active field of exploration; the development of scientific instrumental research methods has in addition created mathematical applications to calculate the collected data. These new methods and applications make it possible to better define the potential and/or the limitations of researching the origin of archaeological material. Renfrew & Bahn (2000) discuss the different methods that can be used for artefact characterisation. The method used will depend on the type of artefact that is to be tested, the operational cost of the instruments, and the condition in which the sample is tested (for example, whole or crushed). These scientific methods are broadly: Optical Emission Spectrometry (OES), Atomic Absorption Spectrometry (AAS), X-Ray Fluorescence Techniques (XRF), Scanning Electron Microprobe Analysis (SEM), Proton Induced X-Ray Emission (PIXE) and Neutron Activation Analysis (NAA). Other scientific tests can also be done, like petrography or thermoluminescence (TL), but they do not identify the chemical fingerprint of the test subject. Petrography analysis is the microscopic analysis of materials using thin sections or polished surfaces. Petrography can identify specific mineralogical similarities. The technique requires specialist equipment and experienced staff with

appropriate qualifications.³ It is not appropriate for Chinese porcelain for the reason stated above.

A small sample of ancient pottery will emit a faint blue light when heated to a sufficiently high temperature. This faint blue light is known as thermoluminescence or TL and is over and above the background red glow that is emitted from all materials. The TL can be measured using a sensitive detector known as a photomultiplier tube. The intensity of the TL signal is proportional to the amount of time which has elapsed since the clay was last heated, normally since the kiln firing, and can be used to date when the object was last fired.⁴ As a TL test cannot be repeated, and it does not identify the chemical fingerprint of the subject, it will further be ignored in this research. Additionally, the method is still imprecise and it is a dating technique rather than a provenance technique. For this research, I will be focusing on chemical fingerprinting in order to identify chemical similarities. These chemical similarities can possibly help identify similar chemical fingerprints of the different test subjects which in turn can be analysed for provenance.

6.2. *Optical Emission Spectrometry*

OES works through heat stimulation of the electrons in the outer shell of the atom. The electrons thus emit a light wavelength that has its own colour. The result of these tests are expressed in percentages for the more common elements and in parts per million (ppm) for trace elements. With an accuracy of only about 25%, Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) (Sporto 2003) has superseded this method.

6.3. *Inductively Coupled Plasma Atomic Emission Spectrometry*

ICP-AES follows the same principle as OES except that the sample must be placed in a solution. By exciting the sample to a very high temperature, interference between element recognition is minimised. This is suitable for analysis of major and trace elements in most inorganic materials. The more expensive but sensitive version of the OES method is Inductively Coupled Plasma Mass Spectrometry. This method of testing gives a more detailed breakdown of the elements present in the sample.

³ <http://www.geomaterials.co.uk/index.php?page=petrography> (last visited 5-3-2008)

⁴ <http://www.oxfordauthentication.com/process.htm> (last visited 5-3-2008)

However, these methods are destructive, and therefore not optimal for archaeological research. The same test material cannot be retested or used for other studies (Sproto 2003, Li et al 2003). Once tested, the artefact is gone forever.

6.4. *Atomic Absorption Spectrometry*

AAS is based on a similar principle to OES. This method can measure over 40 elements accurately, and can detect elements with low atomic numbers. However, it is a time-consuming method and is considered destructive, as the sample taken has to be dissolved in acid (Sproto 2003).

6.5. *X-Ray Fluorescence Techniques*

The foundation of XRF is the irradiation of electrons in the inner shell of atoms by X-rays. The electrons then move up to a higher shell but will immediately revert to their original position, giving off an energy that is unique to every chemical element. This process is known as characteristic X-rays. There are two approaches to measure the energy of characteristic X-rays.

A. The first mode involves the Wavelength Dispersive XRF (WDXRF) method. This measures the wavelength of the X-rays by diffracting them in a crystal of known parameters. As the samples used are usually in the shape of pressed powder or glass pellets, this method is unsuitable for many archaeological artefacts.

B. The second approach is the Energy Dispersive XRF (EDXRF) method, which relies on the direct measurement of the X-ray energy using a semi-conductor detector. This method allows for the analysis of a small area of 1mm diameter, and can be applied to objects of any shape and size. Quantitative and qualitative analysis of small samples taken from either the artefact's surface or interior can also be done. However, the detection and measurement of elements present in concentrations below 0.1% can be problematic. Ideal for distinguishing key components of ceramics, glass, glazes and the pigments used to colour them, this technique is relatively fast with only surface cleaning of the artefact needed. It is also non-destructive (Sproto 2003).

6.6. *Scanning Electron Microprobe Analysis*

SEM works on the same physical basis as the XRF method. To stimulate the electrons in the atoms, an energetic beam of electrons from an 'electron' gun is directed onto the surface of the sample in a vacuum. Samples for this method have to be specially prepared. Either they have to be a thin polished section or perfectly flat carbon- or gold-coated, mounted specimens. This method is destructive. Scanning electron microscopes are usually easily available in an archaeological laboratory and typically used to study metal and ceramic technology (Sproto 2003).

6.7. *Proton Induced X-Ray Emission*

PIXE is an extension of the emission of characteristic X-rays. It works by exciting the electrons using a beam of proton from a particular accelerator. The range of analytical possibilities is similar to that of SEM but this method is more suited to testing very small areas of light materials, for example layers of pigments or paper or the soldering of alloys in making jewellery (Sproto 2003).

6.8. *Neutron Activation Analysis*

NAA excites the nucleus of an atom by making it radioactive. The energy that the atom radiates through gamma radiation is characteristic for each element. This was a frequently used method to analyse trace elements in ceramics and metal. This is a destructive method as samples are taken from the interior of the artefact by drilling. Tests are then done on these samples by using a nuclear reactor. As this technique involves the use of a nuclear reactor, only certain laboratories have this capability. With the closing down of nuclear reactors, these research laboratories have become rare (Sproto 2003). None exists in Singapore.

For the purpose of this paper, I will concentrate on EDXRF testing on ceramics. Apart from its ready availability, it has the added advantage of being non-destructive to the artefacts and the process is relatively fast. I will also discuss my own EDXRF research where I tested Greenware ceramic materials, particularly from Asia.

7. How has EDXRF been used?

EDXRF was developed in the early 1970s. It uses a Lithium drifted or germanium detector so that it can detect almost the entire spectrum of chemical elements in an object simultaneously. A computer attached to the machine then registers the complex method of detection of the individual x-rays. As mentioned earlier, EDXRF can analyse a small area of 1mm diameter as well as objects of all shapes and sizes. A scientific method of testing, EDXRF is a non-destructive and reliable method of examining an object's chemical composition. EDXRF can, when used on a small area (e. g. 1mm²), give exact measurements but when one is measuring a larger areas (>1mm²) the measurements are more geared towards an average of the chemical array in that specific area, a relative concentration one could say. The testing of a larger area is preferred, as EDXRF will average out the data, which means less chance that anomalies will have a major impact on the collected data. In addition, the larger the area the better the data will represent the whole artefact, which will give a better dataset for future comparisons.

As a bulk characterization technique, EDXRF can rapidly and simultaneously detect chemical elements in an object that is heavier than fluorine (F9). By studying the trace or minor chemical elements in artefacts, EDXRF can contribute to the chemical fingerprinting of archaeological ceramics. With enough research, this chemical characterisation will identify the ceramics as being produced from a particular site. The definition of chemical fingerprinting according to Integral Consulting Incorporated;

Chemical fingerprinting describes the use of a unique chemical signature, isotopic ratio, mineral species, or pattern analysis to identify or distinguish different chemical sources, reconstruct historical or ongoing loading patterns, date a particular event, or address causation or toxicity concerns⁵.

However, source materials must be identified as well, so that the artefact's chemical fingerprints can be compared and recognised, which then can lead to provenance

⁵ <http://www.integral-corp.com/page.php?pname=capabilities/forensics/1>

recognition. It is not advisable to compare just one element per test, as the result will likely not be conclusive. It would probably be as effective as trying to compare one line from someone's fingerprint to identify him or her.

The application of EDXRF to Chinese porcelain was pioneered at the National University of Singapore by Prof. Yap Choon Teck in the 1980s. Yap and Tang (1984) researched the manganese (Mn) and cobalt (Co) ratios in Ming Dynasty Chinese porcelain. These underglaze decorated blue designs were tested for the cobalt-manganese ratios. The results showed that these ratios differed for pre- and post- Second World War artefacts. Yap and Tang further compared their research to similar research done by Young (1956) and Banks and Merrick (1967). They concluded that the Mn/Co ratios on pieces before 1425 were less than 0.5 ppm. However, these ratios were 2.0 ppm and above on objects from the Qing (1644-1911) dynasty. Yap and Tang (1982) analysed large number of pieces from the Qing dynasty and the modern era. The Qing pieces they analysed all had a Mn/Co ratio above 3.0 ppm while the modern pieces had a ratio of less than 0.7 ppm. In 1984, Yap and Tang expanded on this research, testing pieces from the early Qing dynasty to more recent pieces. Their findings were that the tested objects from before the Second World War all had a Mn/Co ratio of 2.5 ppm, while the ratio of the pieces after the war was less than 0.7 ppm. Yap and Tang concluded that by using EDXRF testing, "modern fake reproductions of good quality" artefacts could be identified in a non-destructive way. Comparing these results with modern post-Second World War pieces, which have a ratio of 0.7 ppm, it is likely that more testing needs to be conducted in order to substantiate their conclusion.

Miksic (et.al. 1996) tested several different colours of glass by identifying only five elements. This was based on the theory that the analysis of variance test could narrow the number of elements to the minimum number needed to account for all or most of the variation between samples, thus identifying those elements most responsible for differing characteristics of the material. Glass consists mostly of Silica (Si or Silicon) hence when the beads, bangles and glass shards were tested, $\text{SiO}_2 + \text{CaO}_2$ are AlO_2 are the most prevalent chemical components of the glass. The other chemicals they identified are Ga for light blue, Sr for black, Pb probably replaced Calcium Oxide (CaO_2). Nb was likely included in the glass to make the glass malleable for a longer period of time to enable it to be shaped more

precisely; U likely was included because of its property of producing a yellow colour. John N. Miksic, C. T. Yap, Hua Younan in their 1994 paper expanded on their glass research by comparing the Singapore beads with glass beads found elsewhere. The results of the tests seem to be fairly precise but this cannot be surprising as the chemical compounds in glass are far fewer than in glazed ceramic bodies.

Miksic and Yap (1990) tested ceramics from seven sites in Southeast Asia using EDXRF. The study was limited to stoneware and earthenware sherds, without porcelain. The samples used were excavated in different parts of Southeast Asia. After undergoing EDXRF testing, it was found that the fine paste earthenwares were likely to have been produced in “two widely separated centres in early historic Southeast Asia”. However, the authors did indicate that these findings are preliminary and more research needed to be done on this type of earthenware before any specific provenances can be recognised.

Miksic and Yap (1992) later carried out more research on 37 fine paste earthenware sherds. By comparing their test results with their 1990 outcome, they discovered that their initial theory of there being two production centres was most likely incorrect. It was suggested that there is a possibility of multiple production sites within the Southeast Asian region due to the wide dispersal of the test results. The result of this research shows that there is a need for more EDXRF testing of fine paste earthenware of known provenance.

Yap and Hua (1992) compared the chemical composition between Northern China (Ding, Xing and Gongxian) porcelains and Southern China (Jingdezhen and DeHua) porcelain. Their conclusion revealed that the Ding and Xing wares had an ‘appreciable overlap’ whereas all of the other wares had a clearly distinguishable chemical composition from the Ding and Xing wares. They also concluded that if one goes from the north to the south of China, the concentrations of Silica (SiO_2) and Potassium Oxide (K_2O) increase but the concentrations of Aluminum oxide (Al_2O_3) and Magnesium oxide (MgO) decrease. This research clearly proves that by using EDXRF it is possible to determine provenance of porcelain objects from these areas.

As described below (in Habu and Hall, 1999), a high level of accuracy can be obtained by employing EDXRF in order to find information relevant to the origins of ceramic objects. For the Singapore chapter of archaeology and the accuracy of EDXRF provenance research, it is too early to make definite statements. Large-scale chemical fingerprinting of artefacts found in Singapore and many other sites in Southeast Asia, including the testing of local clay sources, would be needed to achieve a high degree of confidence in such conclusions. Only after such large-scale testing would more definite conclusions regarding provenance of local earthenwares be possible. To do the same for stoneware and porcelain there is a need to test materials found at known kiln sites and clay sources, as Habu and Hall (1999) have done. Unlike Habu and Hall (1999), Yap and Hua (1992) compared Ding and Xing wares, which were likely produced from different source materials but likely, have similar additions to the clay (feldspar, quartz and dolomite). The graphs in the article by Yap and Hua (1996) show a close proximity between Ding and Xing wares (the kiln sites are 160 Km apart). It is unlikely that the chemical composition of the two wares is the same but also unlikely that the chemical fingerprint will be very different due to their relative proximity. The importance of this article is that it clearly illustrates that the chemical fingerprint, of ceramics produced in different parts of China, produce different chemical compositions.

By using the EDXRF technique, Habu and Hall (1999) compared the chemical composition of sherds from three ancient Jomon culture sites in the Kanto and Chubu regions, both located in central Japan. Jomon, a cultural era that lasted from 10,000 to 3,000 years ago, had a rich ceramic tradition. Habu and Hall's research indicated that the chemical composition of earthenware is best analysed by using trace elements. The result of the EDXRF test shows that the artefacts found are chemically distinguishable as the earthenware from the three sites cluster differently chemically. Their findings suggests that regional variability of the Jomon culture was more diverse than archaeologist have assumed. This proves that, EDXRF could be used to distinguish between earthenware production centres in places beyond Japan.

Hall (2001) examined trace elements obtained from early Jomon sherds of four sites in the Japanese Kanto region; the artefacts were either Moroiso or Ukishima style earthenwares. The analysis indicated that the four sites were not chemically distinguishable. This could

mean that the four sites retrieved their source material from the same Kanto region, which is a small area within northeast Central Japan.

Hall and Minyaev (2001) used EDXRF to test sherds from six sites taken from the Xiong-nu confederacy, located on the border of Russia and Mongolia. Situated in the inner Asian steppes, the Xiong-nu confederacy lasted from the 3rd century BC until the 2nd century CE. In their research, trace elements were analysed in order to cluster the tested objects. Using the classification maximum-likelihood approach, a mathematical probability method, Hall and Minyaev grouped the test results into three clusters, which could indicate three regional clay deposits. The mathematical analysis used here differs from Hall's 2001 paper where he utilised principle component analysis in a 3-dimensional scatter plot. By using this mathematical approach here, the results appear not to be promising as the researchers have, artificially, forced the elements into defined clusters as the elements did not fall into visibly clear groupings. Outlier identification is important in many applications of multivariate analysis, either because there is some specific interest in finding anomalous observations or as a pre-processing task before the application of some multivariate method, in order to preserve the results from possible harmful effects of those observations. It is also of great interest in discriminate analysis if, when predicting group membership, one wants to have the possibility of labelling an observation as one which "does or does not belong to any of the available groups."⁶ When observing the test results in a graph cohesive groups will be noticed in an instant. By 'grouping' the test results from each kiln site or predestined order it will become clear that these 'groupings' are forced as by circling the intended groups the circles become odd shaped. If we want to group test results it is best to work with an oval outline. This will enable the researcher to stay as objective as possible. The test results by Yap and Hua (1992) show the Xing and Ding results to be close but also clearly separate. It is up to the individual researcher to define their groupings as each research as a different objective.

⁶ A. M. Pires and C. M. Santos-Pereira; Using Clustering and Robust Estimators to Detect Outliers in Multivariate Data, <http://www.stat.jyu.fi/icors2005/icorsabstracts/pires.pdf> (last visited 6-3-2008)

Gigli et al < [http://www. geo. vu. nl/archaeometry/abstracts/ceramicsgeneral. pdf](http://www.geo.vu.nl/archaeometry/abstracts/ceramicsgeneral.pdf) > in their paper on Italian pottery, tested 67 sherds from the San Francesco Square site in Catania, Italy. EDXRF is employed to identify the chemical signature of earthenwares. The analysis of the results demonstrates that the most important trace elements are Rubidium (Rb), Strontium (Sr), Yttrium (Y), Zirconium (Zr), Molybdenum (Mo) and Niobium (Nb). A multilinear regression graph used to plot the elements indicated that all the sherds cluster closely together. Gigli (et.al.) also tested sherds from another site and compared them in a graph together with the sherds from the San Francesco Square site. The results from this comparison strongly suggest that the sherds from the San Francesco Square site come from one production workshop. The trace elements recognized in this article are similar to those identified by Miksic and Yap (1992). This supports the theory by Miksic and Yap (1992) that these trace elements could identify production centres and eventually lead to provenance recognition.

Based on the above research, it is clear that chemical elements within a ceramic object can be identified and quantified by using EDXRF. Through analysing these elements, there is a possibility of distinguishing between different ceramic production sites. Nonetheless, more such research on ceramics and its source materials must be conducted before provenance can be established. Other scientific methods for provenance research have been employed and are at least as successful as EDXRF. Earlier, in this paper, these methods have been described and dismissed for relevant reasons.

8. Methodology

8.1. *Why EDXRF?*

Energy Dispersive X-Ray Fluorescence or EDXRF has been employed in this research for several reasons. One of the most important reasons is usually availability which is also true for this research. The most important reason for using EDXRF is the fact that it is a non-destructive way of testing. This is crucial especially when testing objects belonging to collections. Another reason is that the test results from EDXRF are as precise as other methods. Moreover, EDXRF testing demands far less sample preparation than other methods demand, which in turn decreases the time that one needs to do the testing which could be used for other ventures regarding this research, such as extensive comparative analysis.

8.2. *The EDXRF equipment*

For this research using EDXRF the objects were tested using an annular cadmium-109 radioisotope source. The test subjects are placed on a circular space directly above the radiation source at a standard distance (See figure 5+6+7+8). The opening is 1.5cm in diameter and this is the area size that will be tested on each object on the EDXRF equipment. The ¹⁰⁹Cd source excites the atoms in the test subject with gamma rays causing energy to be released as fluorescent X-rays. The fluorescent X-rays (measured in keV) are detected with a Si(Li) X-ray detector with a 0.0125 mm beryllium window and a microprocessor-based multi-channel analyzer. The equipment is coupled to a computer for data storage and analysis (see figure 9+10). All chemical elements tested for have a unique energy signature, and will be visible as one or more peaks in a graph.

Since the surface of the test subjects is a glaze, which is a glass surface, the transparent nature of the glaze and the hardness of the glaze will allow the gamma rays to excite the atoms in the glaze and, to a minimal penetration, those of the clay. This will allow for both the glaze and the clay to be tested.

As there is a 1.5cm circular part which is tested, irregularity in the sample should average out. If a small area of one millimetre or less were to be tested, an inclusion, of for example a large sand grain or a shell fragment, may affect the results, suggesting a different

composition than what is present in the entire sample. In addition, variation in the clay could occur from one small spot to the next to due incomplete mixing of the original clay used for pottery manufacture. This problem can be resolved by sampling a relatively large area and examining the objects before testing in order to identify an area that has no or few inclusions.

During exposure to the source and subsequent detection each time an energy burst is detected the keV level is measured and added to the total count for that particular peak (see figure 9). A series of curves is produced corresponding to the various elemental signatures. Most elements will have multiple peaks with usually one dominant pinnacle (Figure 10).

The ^{109}Cd source also causes the largest set of curves (seen on the right of the Figure 10) which mask the detection of several elements. In addition, the lighter elements (below Ca) cannot be accurately determined. A total of 22 elements ranging from Ca to Pb are assessed (Ca, Sc, Ti, V, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rb, Sr, Y, Zr, Nb, Mo, Pb). Other elements can be assessed with the use of different irradiating source materials.

This research owes great thanks to the researchers at the NUS Physics Demonstration Lab and Mr. Ng Tong Ho in particular, who are largely responsible for helping me to understand the workings of this equipment. To Dr. D. K. Latinis I am indebted for the eloquent description and documentation of the equipment.

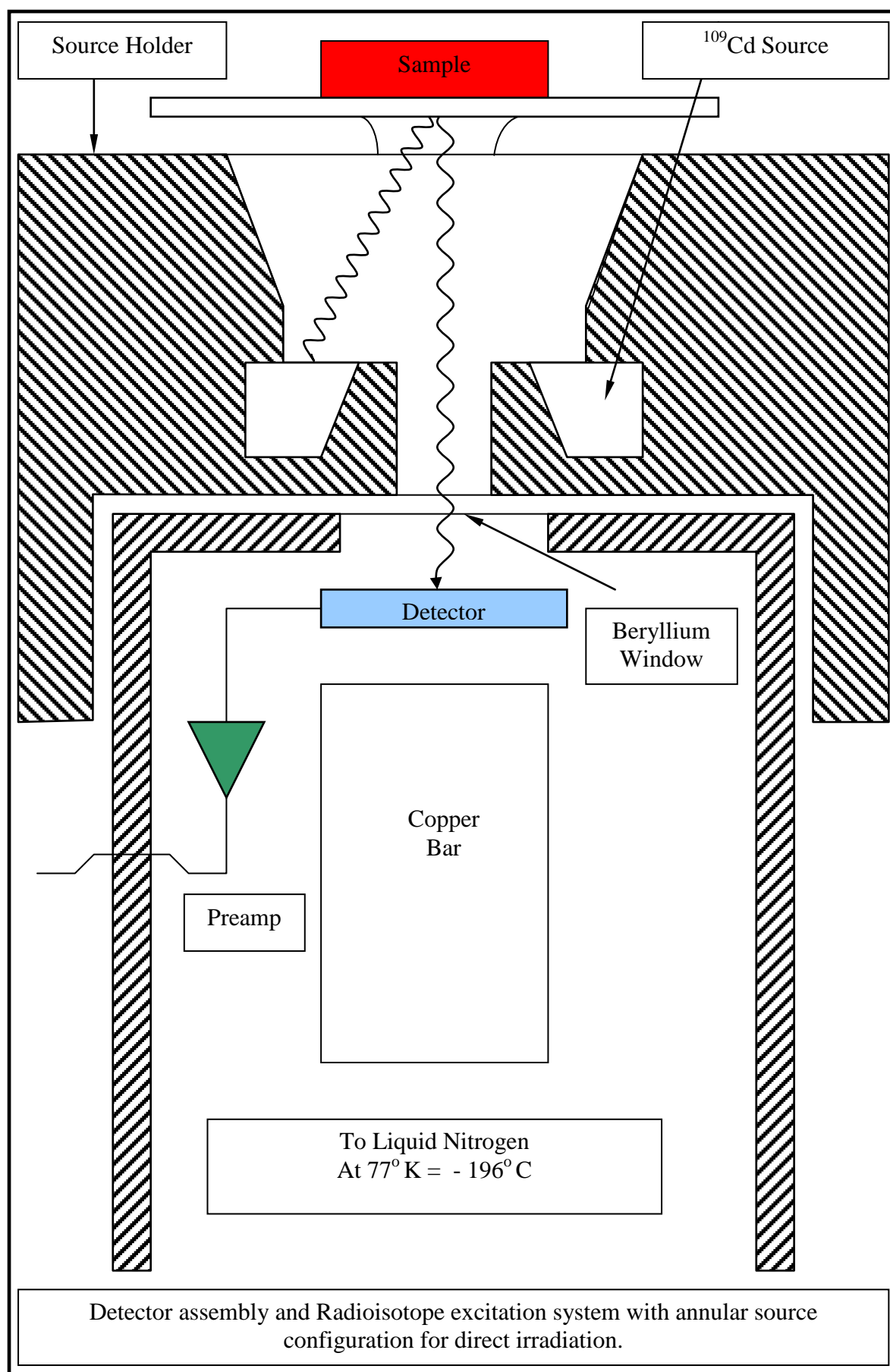


Figure 4: Schematic Drawing EDXRF Equipment



Figure 5: EDXRF Machine at the NUS Physics Demonstration Lab



Figure 6: Radiation Chamber 1



Figure 7: Radiation Chamber Close-up



Figure 8: Artefact Pedestal within Radiation Chamber

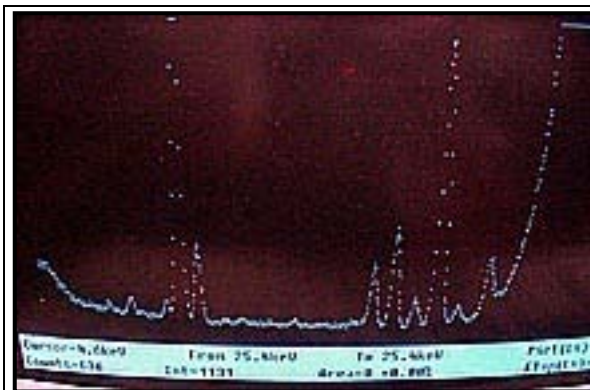


Figure 9: Computer Registering Elements

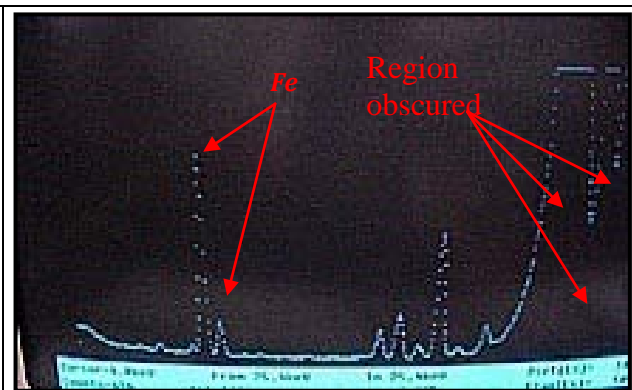


Figure 10: Peak Identifying

8.3. *The size of the chosen sherds*

The test area of the EDXRF machine is a circular area with a 1.5cm diameter. This test area is just above the beryllium window that allows the gamma rays to pass through and reflect back from the test subject. The chosen Greenware sherds for this research all have the common feature of being larger than the test area of the EDXRF machine, in order to not test the edges of the sherds. In order to test only the glaze it is imperative that the fragment size is larger than the 1.5cm diameter test window. The reasoning for the sherds being larger than the test area is as follows: as the sherds are larger than the test area it is a given certainty that the testing is only conducted on the glaze of the sherds. If the sherds were smaller than the test area, it is likely that the unglazed edges of the sherds would be tested as well. EDXRF test results are an over-all average of the chemicals tested on the subject's outer layer⁷.

If this outer layer includes an unglazed area on the sherds, the test would include this in its average. Hence, the results will not be comparable to analyses that only tested the glazed areas of other sherds

8.4. *Greenware*

Greenware is a mixture of porcelain clay and stoneware clays, which have a high iron content. Fired in a reducing kiln the body cools down to a rusty brown colour. According to

⁷ Ng Tong Ho personal communication

Nigel Wood (1999) Greenware glaze consists of Si (silicon), Al (Aluminium), Ti (Titanium), Fe (Iron), Ca (Calcium), Mg (Magnesium), K (Potassium), Na, (Sodium), Mn (Manganese), P (Phosphorus), S (Sulphur). This recipe is of interest to this research as it can corroborate or contradict the use of EDXRF for provenance research.

The colour of the glaze can be a wide variety of greens even to a bluish colour. The green with variations are the result of a lead-based glaze with additions of limestone and sometime small amounts of wood ash (Wood 1999) fired in a reducing kiln. This type of porcellaneous stoneware is the second most common category of material from the site. Within this ware, the most common colour is green in various shades, covering pastes of varying composition (Miksic 1992).

Miksic in his articles on the subject is referring only to the artefacts found in excavations he and his volunteers conducted on Fort Canning Hill between 1984 and 1992. Other sites within the larger site of the Civic District of Singapore have also yielded significant amounts of Greenwares. This suggests that Greenware was a commodity that was traded, considerably, in and around Singapore making it an interesting research topic. Though, there were no similar ports-of-trade like Singapura in the period of 1300-1394 there are other places where Greenware has been unearthed. Near Sinan in South Korea, a shipwreck was found with Longquan greenware on it (Ayers 1997). Greenware has also been found in Japan, the Philippines and Indonesia and further to the west in Egypt and East Africa. Most information on Greenwares in Southeast Asia from this same period seem to consist of marine archaeological finds in the Philippines and were all ships conducting trade there. Singapura on the other hand was a place of passage where possibly some trade was conducted. Southeast Asia did not have kilns or the technological knowledge to produce Greenwares themselves. This leaves us with the question; where did these Greenwares come from?

The short answer is China, where the production of Greenware, arguably, started from the early Shang period (1600-1028 B. C.) (Wang 1989). As to Singapore being a destination of these wares, Singapore was mentioned in the 14th century in Chinese and other texts from Java and Vietnam. These written records show that Singapore played a pivotal role in the

regional ceramic trade. Wang Dayuan of the 14th century states that amongst a number of organic and inorganic materials, Singapore also imported earthenware bowls, and porcelain bowls. The earthenware bowls are likely to be from regional sources whereas the porcelain bowls must have their provenance in China, as there were no regional porcelain production centres in this period. The discovery of Chinese ceramics on this island is then not surprising. The question of where the Greenware ceramics originate from within China is far more difficult to answer and this is the principal thought behind this research; are the archaeological Greenware finds from one or several production centres? To date it is difficult to determine, with a high degree of precision, the provenance(s) of the Greenwares found in Singapore (or any other site outside China). This research provides another layer to the better understanding of Chinese trade with Southeast Asia.

Four researchers have been asked to identify the 130 artefacts used in this research in any way they see fit. These four people, identified as A, B, C, and D, have done so and their answers may clarify the need for a scientific approach when classifying artefacts. Their combined answers are analysed in 'Subjective vs. Objective' (page 101). There is a fair chance that most of the researchers will identify known Greenware production sites as Guangdong, or Zhejiang, three provinces along the south eastern shore of China.

To identify each sherd separately, the use of EDXRF has been employed to retrieve the test objects character chemical fingerprint. The use of EDXRF is not only fast and cost effective; it is as described before, the scientific technique that needs the least preparation. EDXRF is a mobile technique; portable machines are now available in the market. EDXRF can be done in the field, at the archaeological site, needs little preparation, is non-destructive so future stylistic analysis is not jeopardised, and it is a low cost technique which provides the researcher with a clear result of the chemical fingerprint of each test subject. Further operational methods and techniques are described below.

Provenance research in archaeology is usually conducted by comparing the stylistical traits of an object. This is a subjective technique in which each researcher will judge the subject by his/her own standards and knowledge. By employing a scientific technique the chances of a piece being assigned to the wrong provenance group are minimized as will disputes

between scholars, researchers and other interested persons as the scientific tests are objective and leave no points for discussion, as of the provenance of the piece. This will give these interested groups more time to discuss stylistical issues with more substantial information. It would also make it possible for the first time to produce quantitative data on the proportions of wares traded from different production centres in China to different consuming areas in Southeast Asia. This knowledge would add a new and deeper layer to our understanding of early Southeast Asia-China relations, and the workings of the system of maritime trade, over a period of several centuries.

To have a system that can identify the provenance of ceramic objects there is a need for extensive testing of not only objects but also source material. Li (et.al. 2006) it may be expected that as major [chemical] elements such as Si and Al form the structural constituents of the dominant mineral phases present in clay sources, they may show only a relatively limited range in composition across different places. Trace elements, on the other hand, depend more on the compositions and geological histories of the original source rocks and are potentially more useful for fingerprinting ceramics made in different places.

Most ancient Chinese kilns used clays mined from the local areas and differences in the geochemistry and mineralogy of these raw materials both between and within production zones may be expected. The chemical composition of ceramics may also be influenced by production techniques, such as processing by washing and mixing of different sorts of raw materials. These may also vary from kiln to kiln and even change over time. Thus the raw materials and production techniques used by a kiln may give its finished products a characteristic chemical signature allowing their provenance and possibly even their age to be determined (Yin et.al. 2000, Leung et al 2000, Li et.al. 2003). By deduction of this statement, one can conclude that for each production centre that receives its source material from a different location, the clay will likely have a different chemical composition. It is unlikely that each clay source has the same history; the chance that settlements, forests, forest fires, flooding, burials and other natural and human influences affected different clay sources at the same time in the same way is negligible. The formation and weathering of each clay source will be different and thus the chemical fingerprint of the objects produced from that clay will test with different chemical trace elements. As Li (et.al.

2003) say, the major elements will be the same as clay has to consist of those major elements in order to become clay. For provenance research the trace elements with different comparisons will be of value to identify the unique chemical composition of each individual object.

8.5. *Sherd identification*

Two major excavations in Singapore were chosen for the fact that they had generated enough Greenware to fill the pre-set structure for this research. Empress Place excavation (EMP) and the Singapore Cricket Club (SCC) excavation had generated enough Greenware material to provide data with which to complete the analytical framework proposed here. During the identification process, minimally 200 sherds per excavation site were chosen. The selected sherds that came from the two sites that have an assemblage of Greenware that can fulfil two important criteria; one, the size of the Greenware sherds is larger than the test window on the scientific equipment and two, the number of sizable sherds is at least 200.

The sherds from EMP come from the following units;

- EMP 18 sherds - EMP indicates unidentified location, surface finds
- C4-1 10 sherds- Unit C4 layer 1
- C4-3 20 sherds- Unit C4 layer 3
- B4-1 8 sherds - Unit B4 layer 1
- B4-3 9 sherds - Unit B4 layer 3

The sherds selected from SCC come from the following units;

- SCC1 L7 28 sherds - Unit 1 layer 7
- SCC1 L8 37 sherds - Unit 1 layer 8

All sherds have been identified as being not modern reproductions of ancient Greenware ceramics by different experts.

8.6. *Structure*

In order to use a representative selection of sherds for analysis it was found necessary to develop a comprehensive structure that would be objective. Before any system could be developed it is important that the sherds be cleaned thoroughly; this was achieved by using what is known as a 'Steam-cleaner'. A steam-cleaner generates high pressure steam which, in our case, was expelled at 5 bar. This high pressure cleaning ensures that all the remains that are not original to the Greenware body are removed. The water used for this process was demineralised water, in order not to add any impurities onto the glaze or sherd.




Before the individual numbering process it was necessary to remove some of the surface at the breakage-side of the sherds. This was done by using a diamond drill bit attached to a Dremel MultiPro 395RD at a speed of 33,000RPM. This process revealed interesting information about the hardness of the sherds and was used for the first stage of the sorting structure: Hard Paste vs. Soft Paste. Thanks goes to the National Parks Board at Fort Canning for providing the Singapore Archaeological Laboratory and its volunteers a location and facilities for this work.

The glaze of a Greenware object [read: sherd] has many influences during its production that control the colour, firing temperature (nearer or further from the fire in the kiln), clearness of the glaze used, type of firing material and all other external influences. The colour of the clay most likely has less of these influences and it seemed an interesting project to sort the sherds in different groupings according to clay colour. To obtain these different colours in groupings I used what is commonly known as a colour fan. ICI produces a colour fan; "The Master Palette" used in this research, which was produced in 1996 and includes 1695 unique colours. The following colours from the colour fan represent the colours of the clay of all the sherds for this research.

Archaeologists have their own colour code, namely the Munsell colour chart. The difference between the two colour charts is that the Munsell chart focuses on the different colours of soil and the ICI colour fan focuses on the difference in colour of paint. The approach is different: where as Munsell developed a coding system to identify soil colours, the paint colour fan can be expanded if a new colour is produced. The Munsell colour chart is limited to soil colours; this does not include colours of clays and glazes produced around the world.

The paint colour chart has so many different colours that it is unlikely that there is any colour of fired clay or glaze that cannot be identified by comparing it to this colour fan.



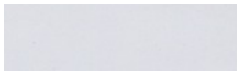
On fan number 181:

483	Sandringham	10YY 59/111	
489	Unicorn White	10YY 64/048	
567	Church Street	20YY 75/073	

On fan number 154:

929	Spiders Web	90YY 73/040	
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On fan number 133:

1316	Swiss White	30BG 72/017	
1415	White Fog	70BG 72/029	
1451	White Smoke	90BG 72/025	

The next step in the sorting structure was the difference in glaze thickness. The difference was rather clear, hence it was included in the structure. The division was between glaze that was thinner than half a millimetre and thicker than half a millimetre. The structure for the two sites is visually more easily represented in the charts below. The division between the two thicknesses of glaze could have an influence on the colour of the glaze as the transparency of the glaze will allow or disallow the colour of the clay to come through. This makes it unlikely that it is possible to further sort the sherds in groupings of glaze colour.

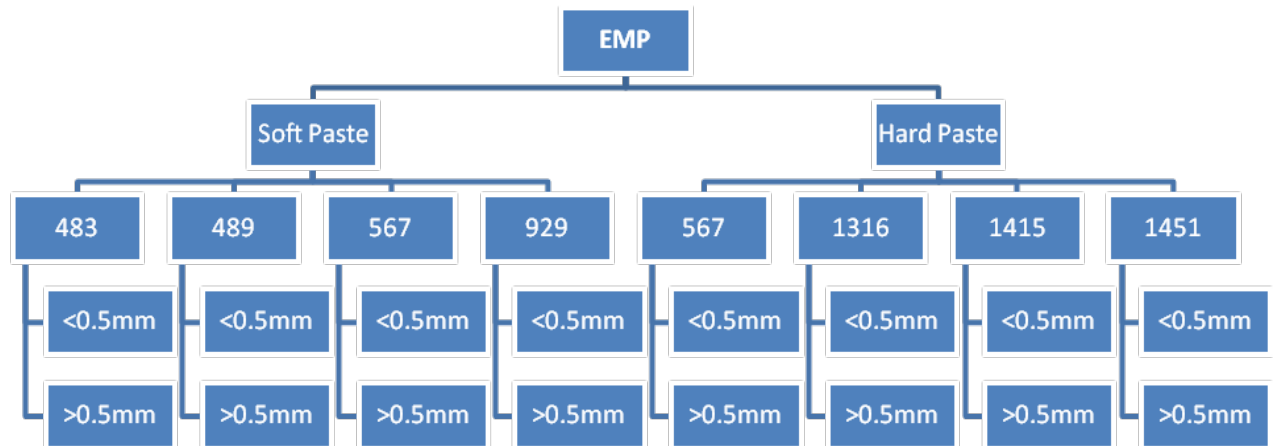


Figure 11: EMP Sorting Structure

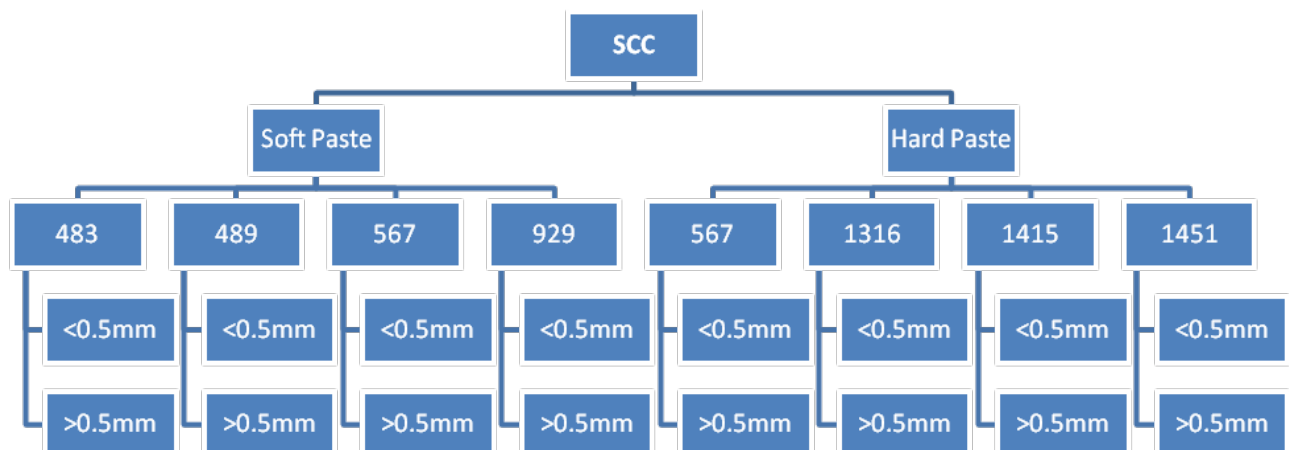


Figure 12: SCC Sorting Structure

The sorting for this structure started with 200 sherds for each site; in the end, 65 sherds were retained from each site. Four sherds per individual grouping were chosen, this will allow an equal chance for the sherds within one group to be either different or similar as the criteria are subjective and the scientific tests will have to prove their uniformity.

After literary research, it became clear that similar research projects include an average of 50 to 60 sherds; this research includes 130 research subjects. The conclusion drawn from this research could thus be more significant. However, the sherds are all from the overall large site of Singapore, which in turn could be the limiting factor and render the results merely representative for the Singapore site.

The sorting structure will allow for different angles to compare the data with each other. No publications found up to date have included a similar structure nor have they compared these aspects of sherds with each other. This could be an indication that this is a non-issue in the chemical fingerprinting of objects, or it could be a unique aspect of this kind of research. As all these sherds came from similarly shaped objects, likely to be bowls, it is possible that they could have been used to hold a substance, which could have settled under the glaze, as some of the glaze on the pieces showed crackling. The shape of the objects was not a criterion but coincidence lead to the use of similar shaped sherds. It must be noted that there are a number of sherds that are likely from large plates as well. To eliminate any possibility of organic materials that could not be removed by steam cleaning which might exert influence on the tests, only the outside of the sherds was tested.

Singapore's interesting ancient history has left much archaeological evidence for future research. This research incorporated sherds found at two sites within what is considered the old town of Singapura. The Empress Place and Singapore Cricket Club excavations have generated enough Greenware sherds to allow for a visually diversified sorting system using the most objective means possible.

9. How did I use EDXRF?

This research focuses on archaeological ceramics from two excavation sites in Singapore. The non-porous Greenware sherds used are from the Empress Place excavation (EMP) and

the Singapore Cricket Club excavation (SCC). The artefacts have been found in distinct 14th century stratigraphic layers. Visual examination by four scholars placed the sherds in several categories, mostly Chinese with the others being Vietnamese or Thai.

The objective of my research is not to determine which production centres the sherds come from but to see if it is possible to distinguish between different production centres. By using the non-destructive EDXRF method on the glaze, I have accumulated the chemical signatures of 130 Greenware sherds. During the initial phase, samples were tested for 300 sec, 900 sec, 1800 sec, 3600 sec, 7200 sec, and 10800 sec. It was found that when the glaze was exposed for 3600 sec (one hour), the results were as clear as when they were tested for a longer duration. Tests conducted for a longer period of time did not show a distinguishable difference in the chemical signature. As such, for high-fired glazed ceramics such as Greenwares, this amount of time is sufficient to produce reliable data for this kind of research.

The EDXRF information was then analysed with the software programme, Analysis of X-Ray spectra by Iterative Least-squares fitting (AXIL) software. The programme identifies the chemical elements registered during the tests, and generates numerical values for the chemical elements.

In this research, 25 elements were identified. Since only 11 of the 25 elements exhibited distinguishable peaks in the graph, the other elements were excluded. These peaks show how much of the chemical has been detected in the sample (see figure 9 and 10). The excluded peaks represent chemicals that are in the test results print out but are present in quantities which are too small include in the analysis. These data sets retrieved from the AXIL software were then entered into a database of the statistical programme, SPSS.

10. Statistical Programmes & Graphs

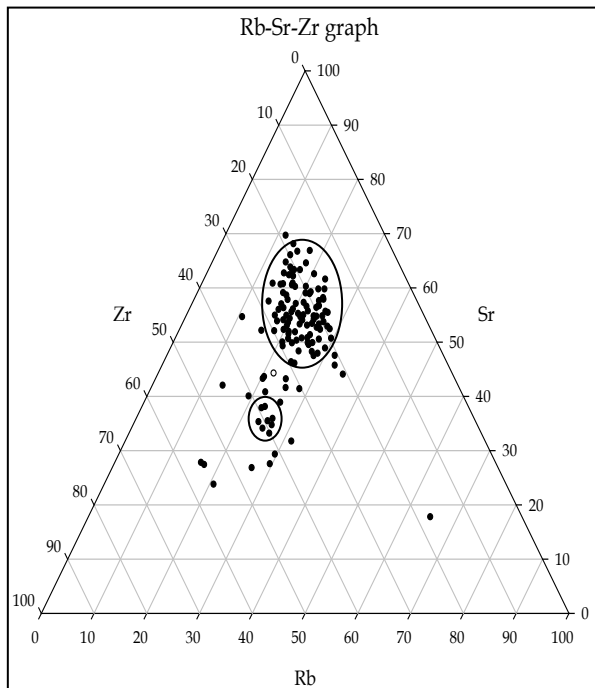


Figure 13: Example Ternary Graph

The choices of analytical software are numerous but two different programmes, namely SPSS and SigmaPlot, were considered. The choice of which software to use is one of convenience as both will produce identical plots with the data provided. For this research, initially, it was opted to use SPSS for no particular reason other than convenience.

Different statistical programmes produce different graphs. SPSS can produce 3-dimensional (3D) graphs, which on a computer are legible. However, once printed out on paper, the 3D graphs become 2-dimensional

(2D) and are no longer legible. SPSS has proven to be a more useful programme as it produces ternary graphs. These ternary graphs display, in a 2D manner, how the data correlate with each other. Yap and Vijiyakumar (1990) point out that the ternary graphs are not as accurate as scatter plot graphs. The main difference between the two types of graphs is that the scatter plot graphs deal with the raw data whereas the ternary graphs deal with a percentile concentration. This makes the assessment of the scatter plots not useful for analysis, unless an extra control element needed to be added that would render the graphs on the X- and Y-axes the same. Having compared all the scatter plots it has become clear that this adding of an element is not productive as it would compress the test results into a small corner of the graph with no chance to distinguish clusters.

Within the SPSS software, there is an array of choices when it comes to using graphs. Initially the option of three-dimensional (3D) graphs was chosen as the results have more angles to allow analysis to be conducted from more perspectives. Out of 25 elements tested (page 57), 11 elements had significant peaks making them eligible to be used for further

analysis. The 3D scatter plots generated 164 comparative analysis graphs to be analysed, as three elements may be compared in each graph. However, the results of the 3D graphs are not represented in this research, as the 3D graphs can only be accurately viewed on a computer monitor with the right software installed, not in print.

Next it was decided to use scatter plots, which were employed to generate accurate graphs using the AXIL data. These are 2D graphs and include only two elements to compare to generate a legible graph. This made it possible to reduce the number of graphs to be analysed from 164 to 56. After close analysis of the scatter plots it became clear that these scatter plots cannot be used, as the X- and Y-axes have different scales when compared, which would be comparing apples and oranges as it were. The 2D graph axes have a numerical value, which is correlated with the number of times the EDXRF test registered the element. When comparing an element with a high occurrence with an element that has a low occurrence in the object tested, the graph would have one axis with higher numbers, into the thousands, and one short axis with low numbers. A brief venture into making both axes the same yielded no positive results as the results would be compressed and would not represent the actual data. This leaves the use of ternary graphs, which are a 2D representation of 3D information. The use of ternary graphs is only possible in the SigmaPlot software. In order to obtain the ternary graphs it is necessary to normalise the data. This process of normalisation is an extra mathematical step and consists of averaging the data, which then becomes a percentile representation of the actual data.

10.1. Data entry

The EDXRF tests were run through the AXIL programme to generate numerical data for each test. The EDXRF data which the machine generates is represented in peaks on a graph (see figure 9 and 10) which are transformed to numbers once the data is run through the AXIL programme, attached to the EDXRF machine used. These 25 elements plus one row of sample names had to be entered into an Excel file for future use. As no conversion programme is available which would allow the data to be cut and pasted into Excel or any other programme, the numbers had to be entered manually, which comes up to 3380

entries. After the data was entered, it was imperative to check the data for any data entry typos as this could have a significant impact on the graphs that were to be generated by using this data. It turned out that more than 98.5% had been entered correctly and after checking the entered data twice there is the small possibility that less than 0.5% of the data could still be mistyped. This is negligible for the remainder of the analysis, because out of the 25 elements tested, only 11 will remain which reduces the chances that any data have been mistakenly entered even closer to zero. The datasheets produced by the AXIL programme are available in appendix 1 for further perusal.

In this research 130 Greenware samples were tested for their chemical fingerprint using Energy Dispersive X-Ray Fluorescence (EDXRF) with a restriction of 25 elements per sample.

Initially eleven elements were selected for further research, namely K, Ca, Mn, Fe, Zn, Rb, Sr, Y, Zr, Nb, Pb. Library research made it clear that only trace elements are possible markers for provenance research. All eleven elements selected here may be considered trace elements since they are not basic component of clay or glaze.

When producing Greenware objects potters have to use a standard recipe, otherwise the object produced would be something else than Greenware. The clay body and the glaze on its surface are very sensitive to slight variations in chemical composition, as well as temperature, oxygen content, and firing period during the firing process. The clay purification process is likely to have differed slightly according to time, place, and possibly the individual craftsman. The chemical elements within the clay however had to be very similar in all regions where Greenware was produced. The production recipe of the glaze must have been similar as well, otherwise different colours would result. The purification of the clay, and glaze preparation may have had slight variations, but the major elements in both recipes must have been very similar.

The clay purification process was done by submerging and stirring the excavated clay in a large basin with fresh water. This process would allow the organic material to either sink to the bottom or float to the surface. The impurities on the surface of the water would be removed after which the clay is allowed to sit in the water for several weeks in order for the

heavier organic and inorganic material to sink to the bottom of the basin and the clay mineral to settle. The water at the surface would then slowly be removed and the clay is carefully scooped out and pressed, in a cloth in a basket, to remove excess water. This process would remove heavier materials like stone, sand, volcanic material, and unattached elements like Fe and Pb from the clay mixture and with them some other elements. The purified clay will have a slightly different trace elemental composition compared to the raw source material the kiln would receive. The notion of testing source material in order to assess provenance is thus a thesis that is unlikely to be a useful one.

10.2. Clustering

Statistically, the important pattern sought by analysts observing the results of chemical analysis such as EDXRF is the clustering of samples, and the degree of differentiation or distance between clusters. If samples do not cluster, this indicates purely random variation, which tells us nothing about the classification of ancient ceramics. The analyst has a choice of techniques, which may enable him or her to discover clusters, which are assumed to represent non-random variation of chemical composition. The act of detecting the clustering of samples in the scatter plots is probably best done by simple observation of groupings; this is the standard practice found in published studies. These groupings can assume a multitude of shapes depending on the necessity. In general, the shapes of the groupings are determined by the unused space between the clusters⁸. The meanings of these clusters could be multiple; they could indicate different production centres, different kiln sites, different period in time, or even differences in exploitation of the clay from a clay source. The different possibilities provide interesting opportunities for future comparative research.

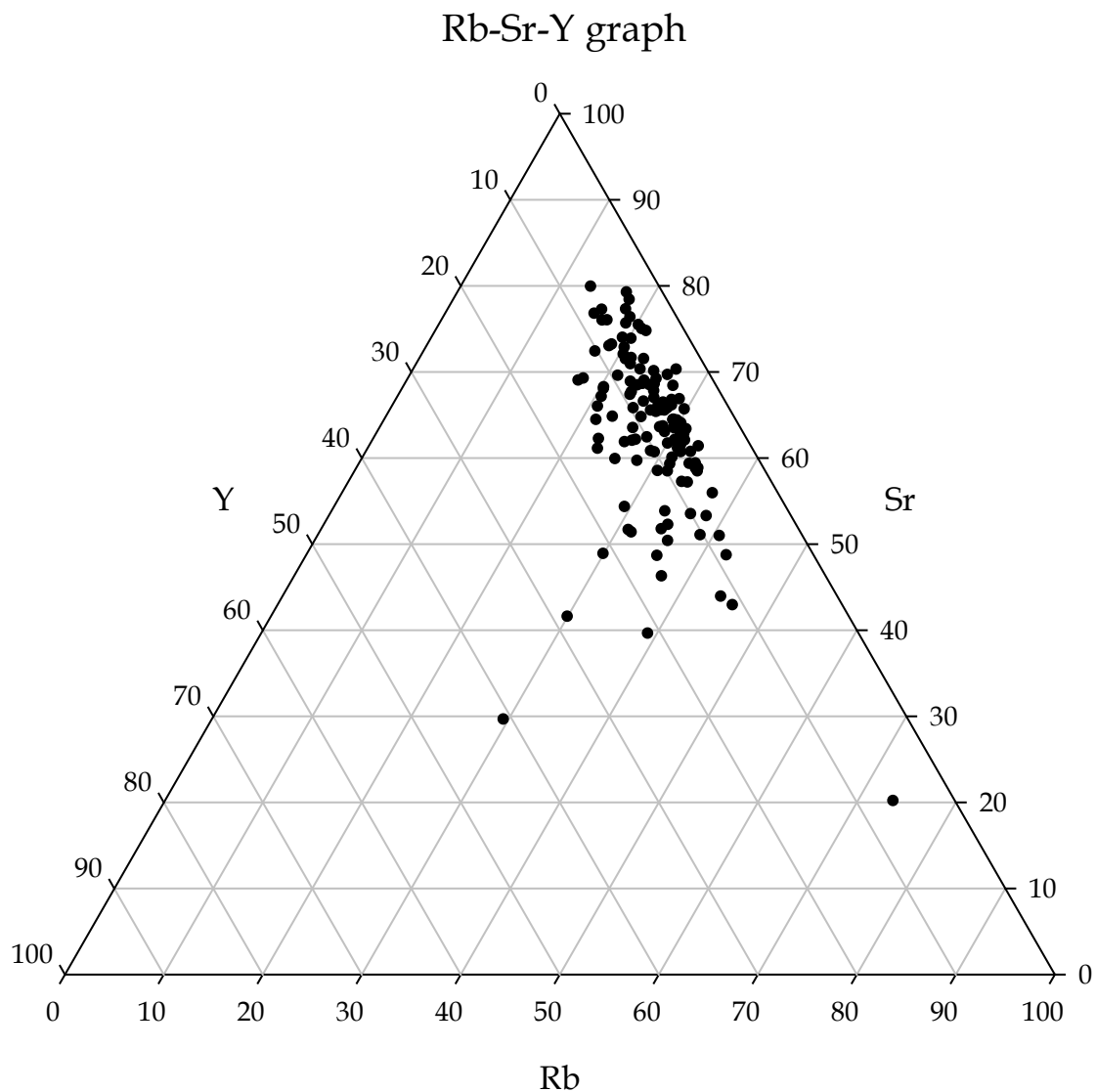
In order to determine the best combination of elements to compare it is first necessary to compare all the elements with each other and then to assess the most significant grouping. All authors seem to agree that the trace elements in the chemical fingerprint are most useful. In order to be as complete as possible I have analysed all the possible comparisons

⁸ Dr. D.K. Latinis, personal communication.

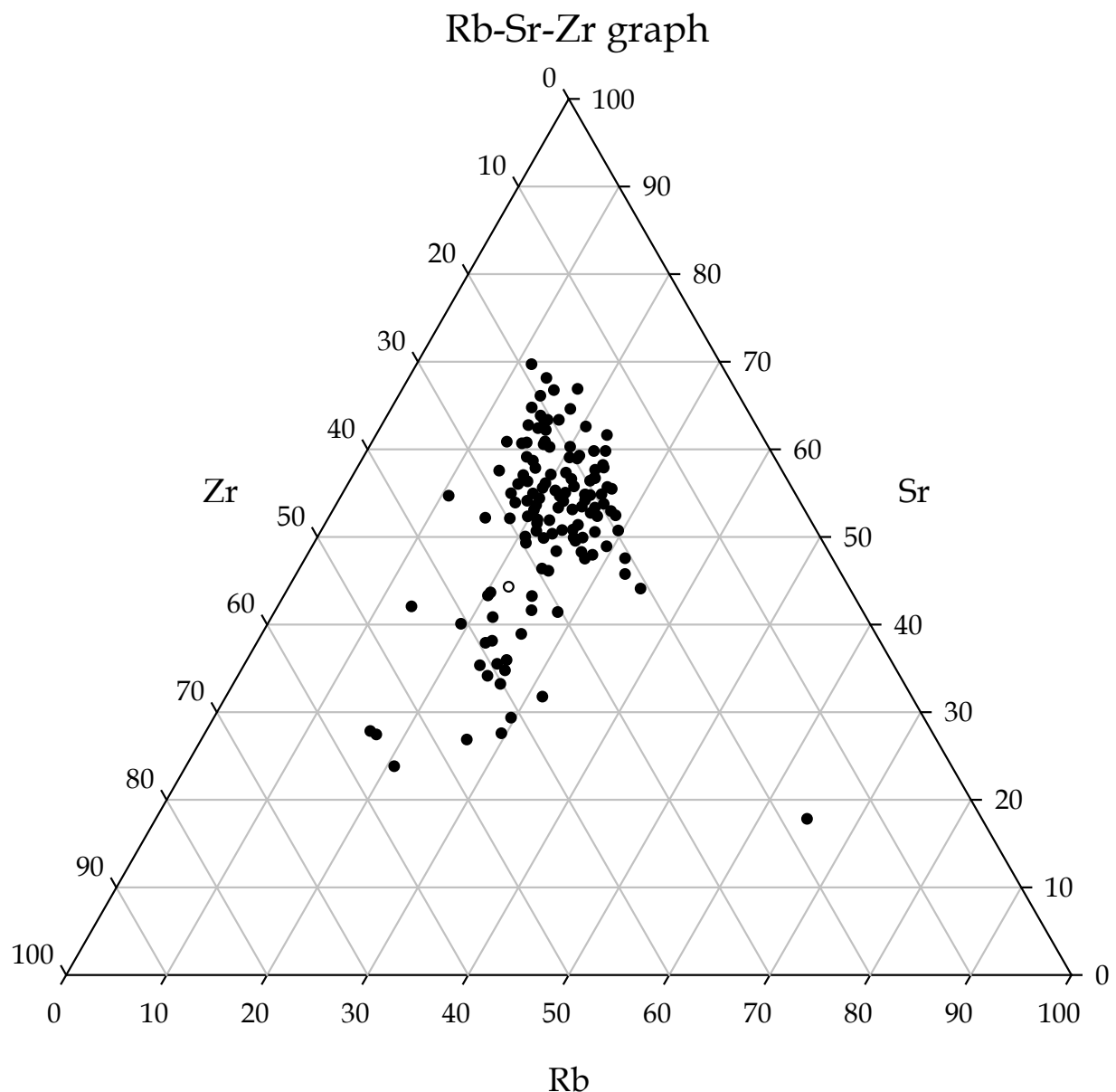
in the graphs but a complete treatment of the data would produce 164 graphs. In order not to reproduce data merely for the sake of inclusion, only the trace element comparison graphs are included in this thesis.

11. Graph Analysis

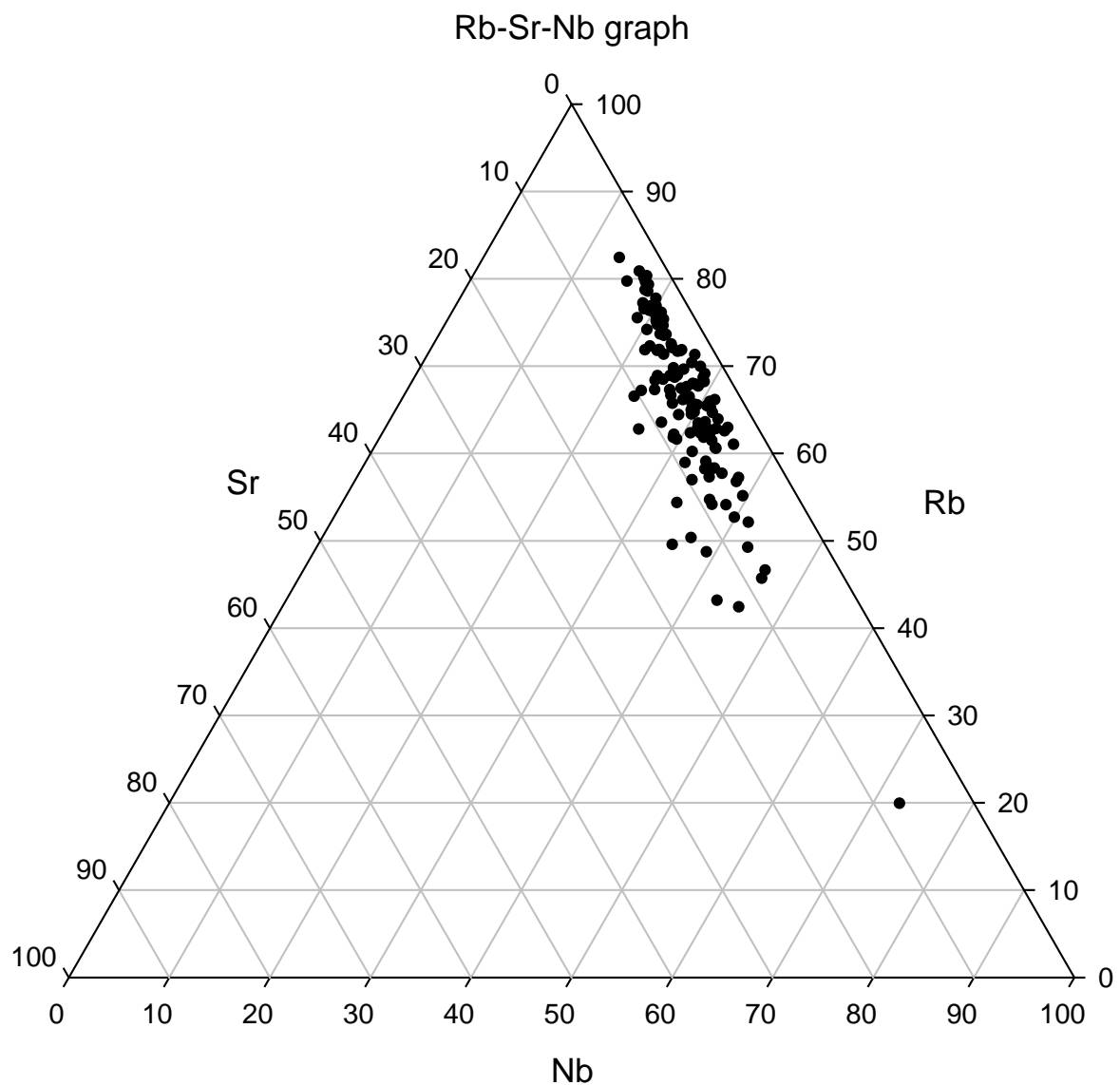
Rb = Rubidium, Sr = Strontium, Y = Yttrium, Zr = Zirconium, Nb = Niobium, are the only trace elements left after excluding all the elements that did not produce a readable chemical peak after testing and excluding all major elements. This leaves 10 graphs to be analysed.



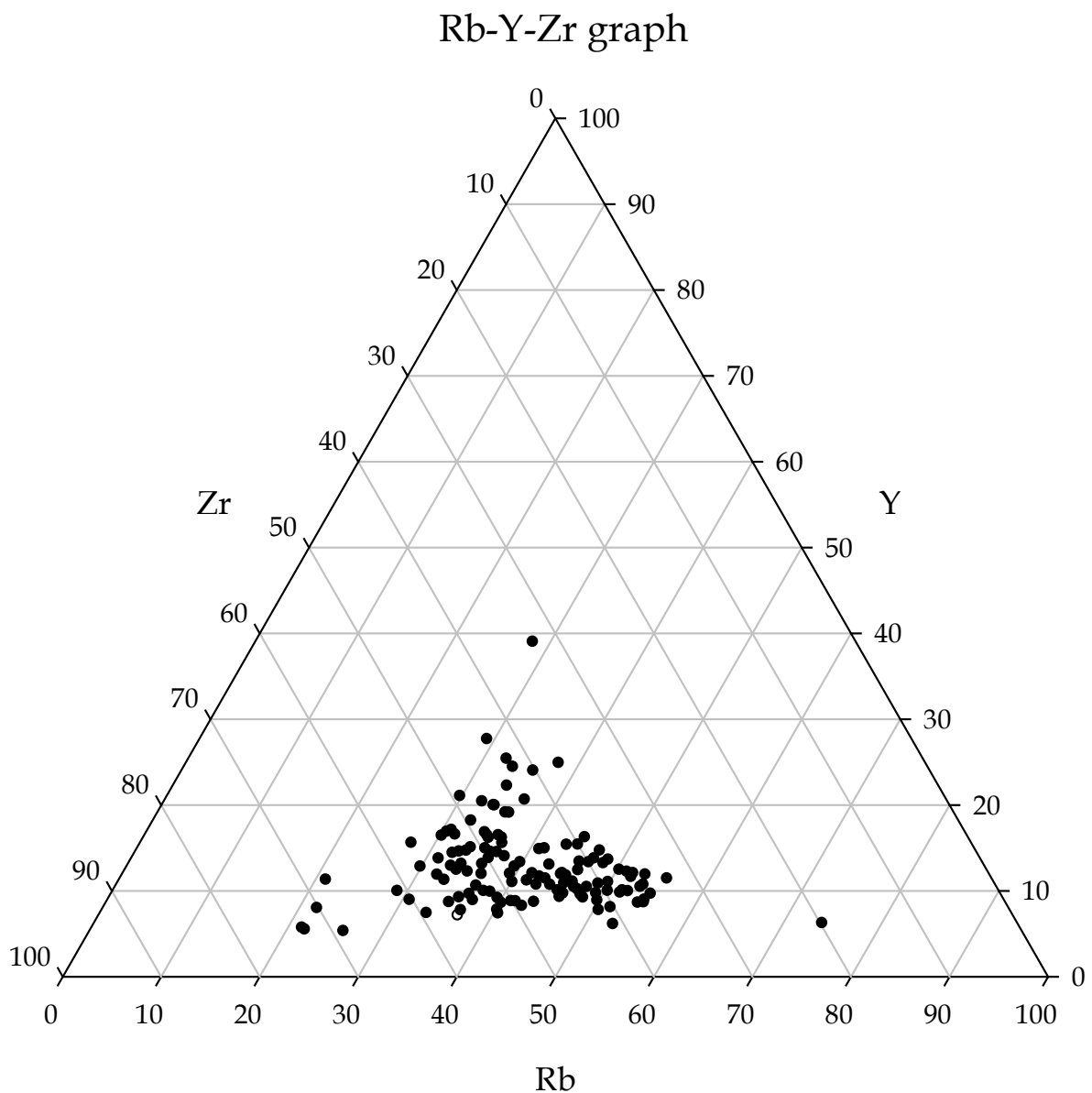
The Rb-Sr-Y produce what seems like a clustering even though, the cluster is compressed toward the Sr-axes. There seem to be some smaller clusters in the large grouping, which suggests that this graph could reveal more information. If the other graphs do not produce a better spread or clustering, this graph could be used for cluster analysis, though with obvious difficulty resulting from the compression.



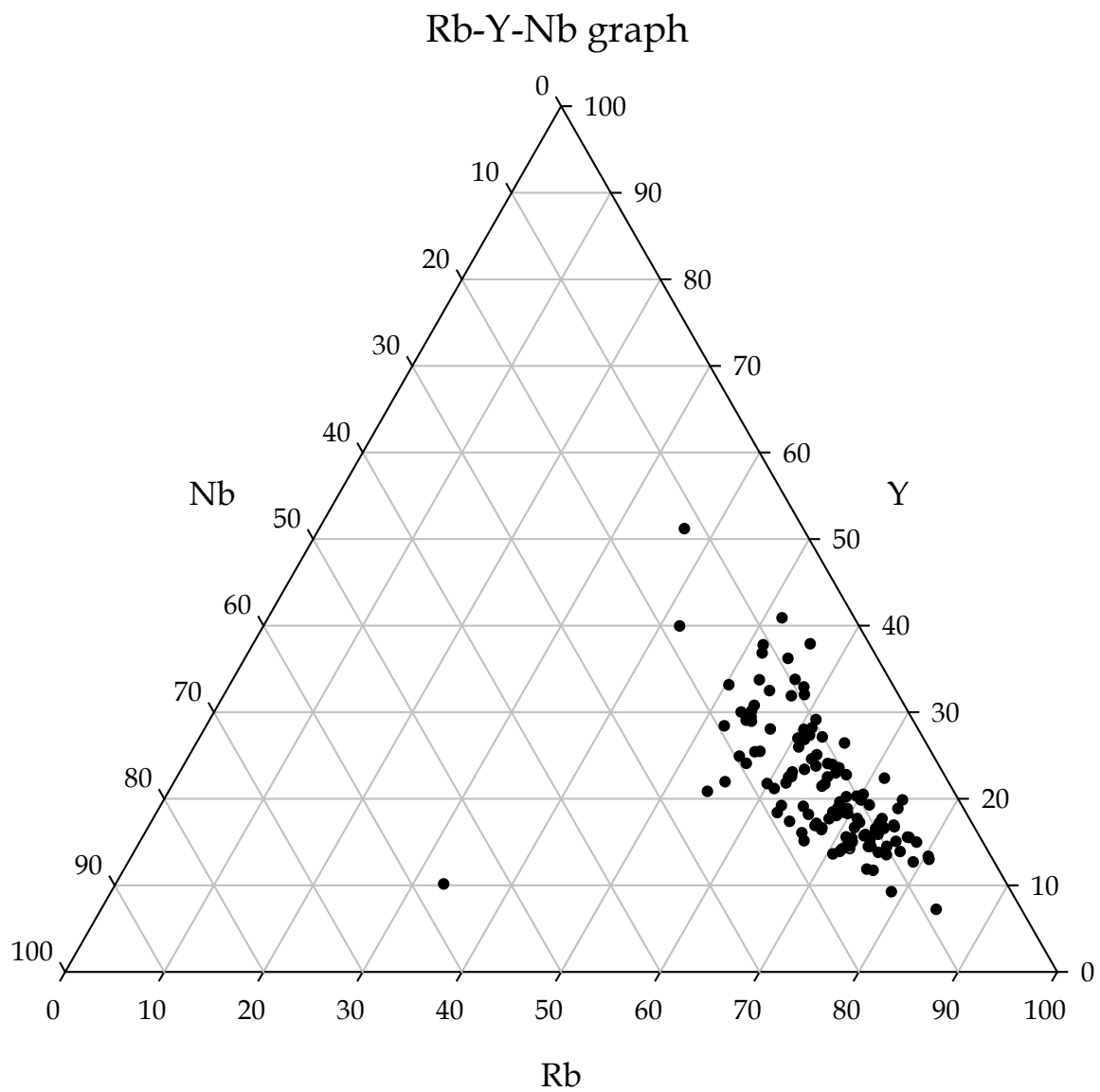
The Rb-Sr-Zr graph produces a clear clustering, which could produce the results hoped for at the beginning of this research. In the large cluster, there are smaller groupings visible. This graph is much better than the previous one because the results are more centred. This graph will be used for further comparative research, rendering the previous graph superfluous.



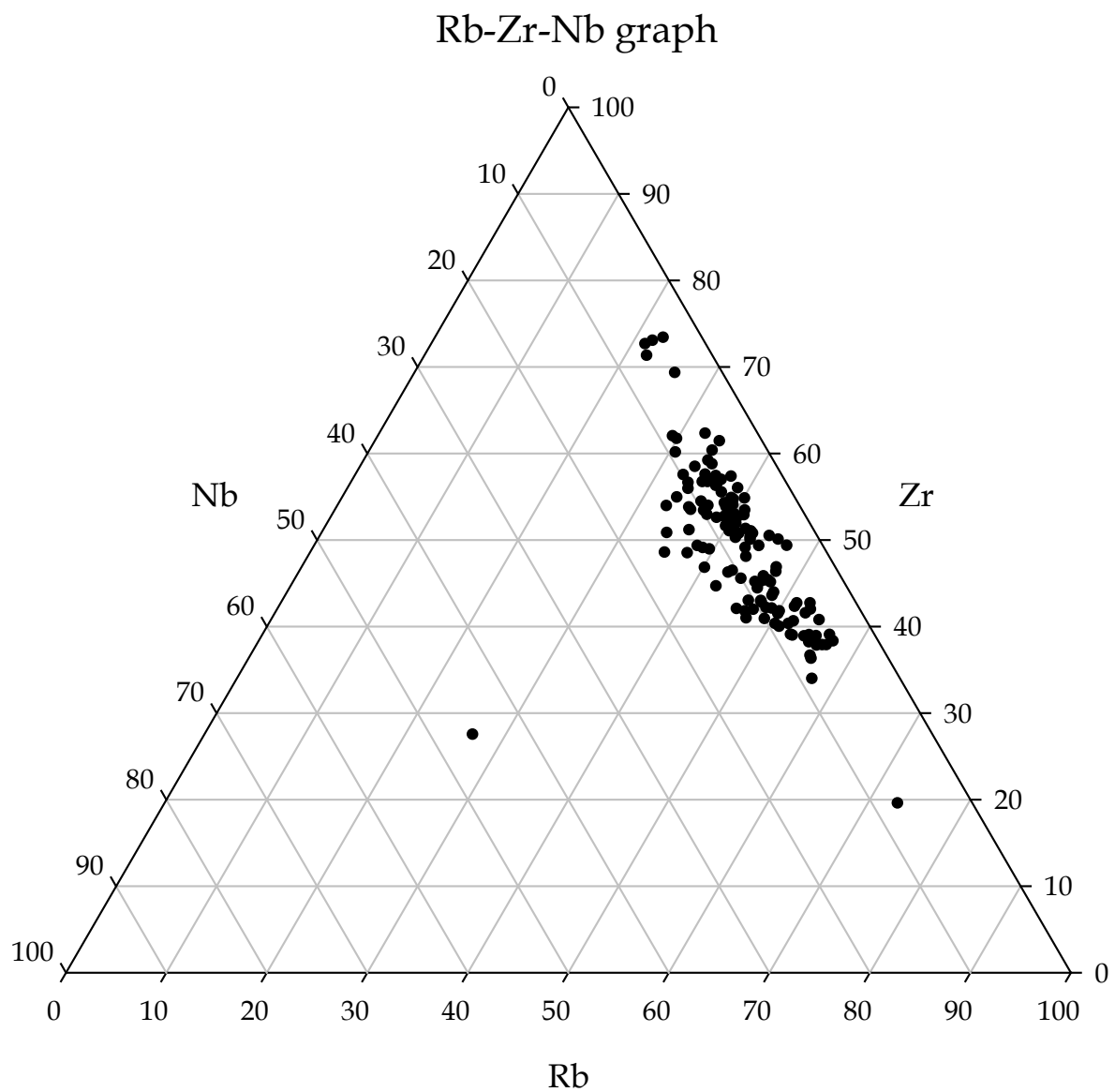
The Nb-Rb-Sr graph produces a compressed cluster near the Rb axes. This graph offers minimal research opportunities and thus will not be used for further analysis.



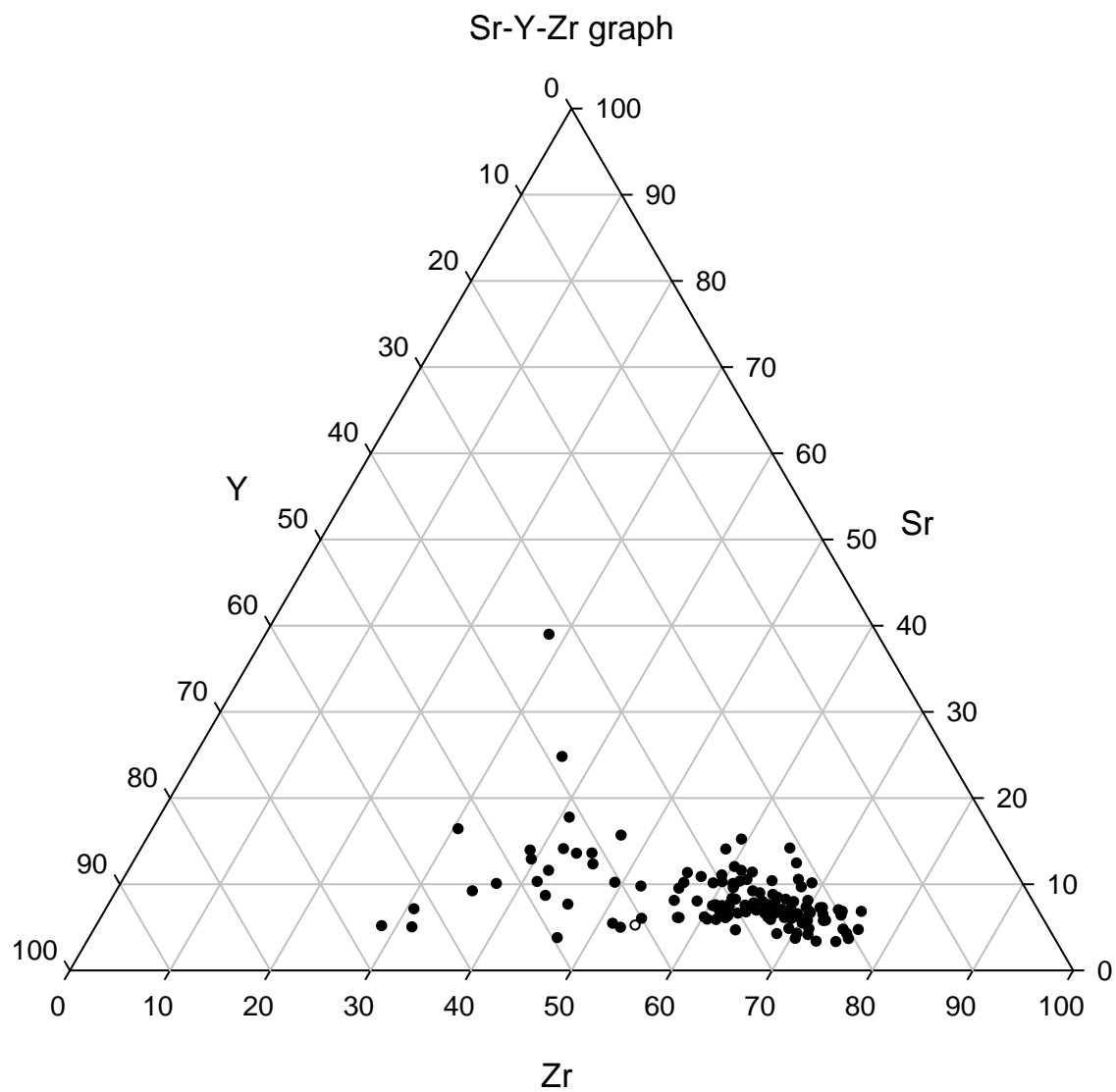
The Rb-Y-Zr graph produces an interesting spread of what look like clusters. This graph does seem to produce a clustering but it is clearly not as legible as the Rb-Sr-Zr graph. As the results are compressed to the Rb axes, this graph will be excluded from further analysis in this research.



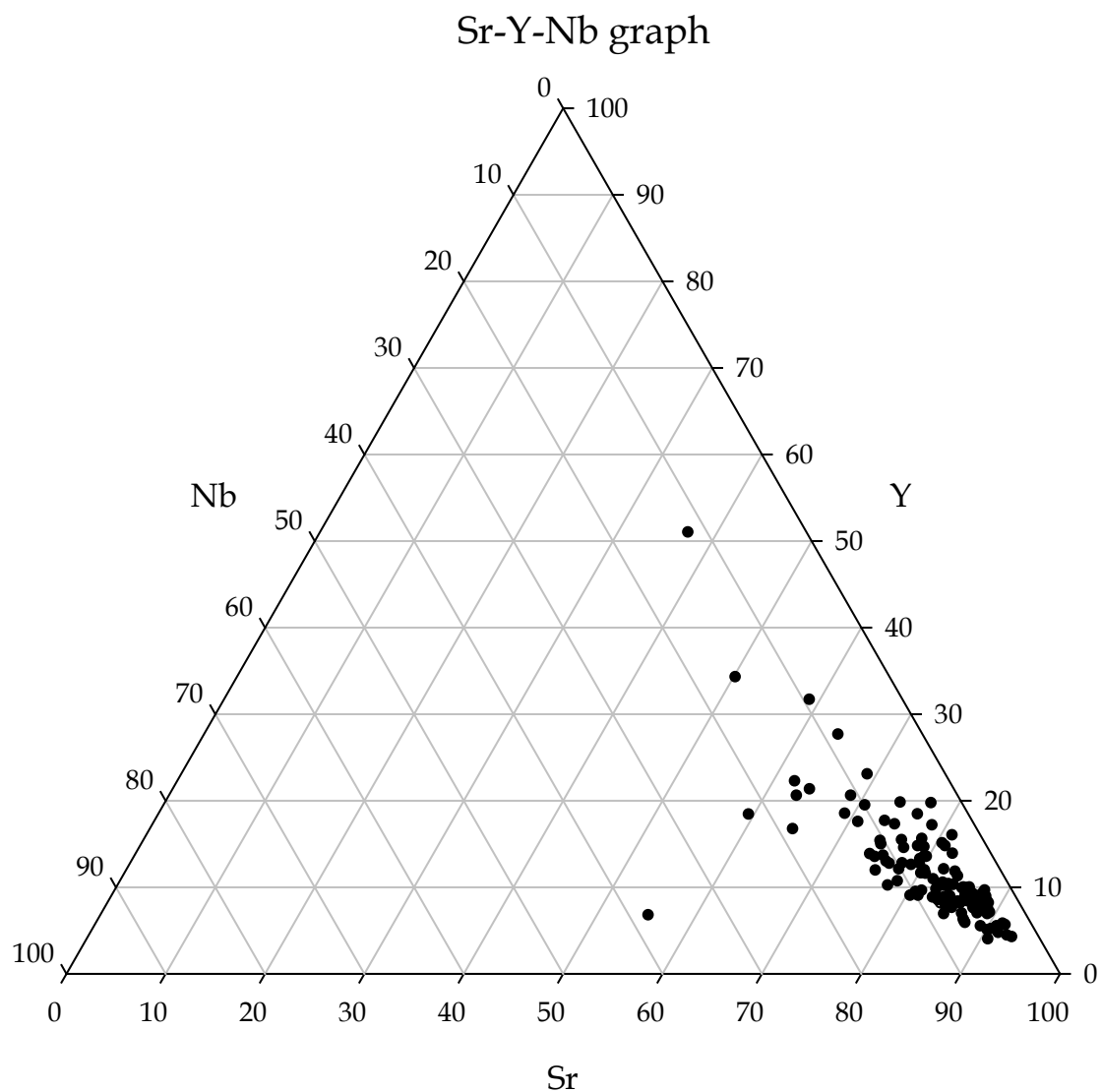
The Rb-Y-Nb graph produces a clustering that is compressed in the corner of Rb-Y. The compression of the results in a corner excludes this graph from further analysis in this research.



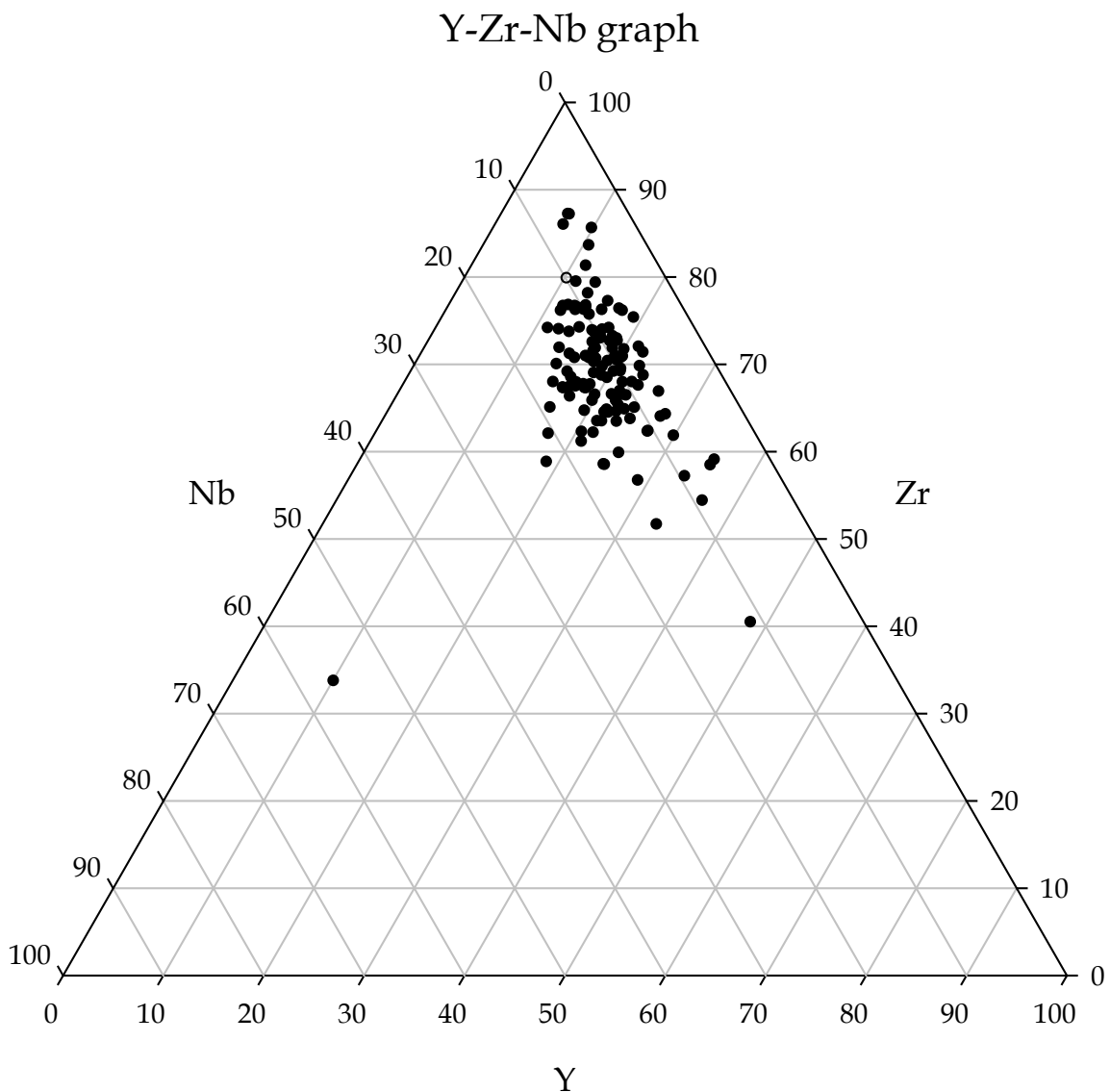
THE Rb-Zr-Nb graph produces a compressed cluster near the Zr axis. This graph offers minimal research opportunities and thus will not be included in further analysis.



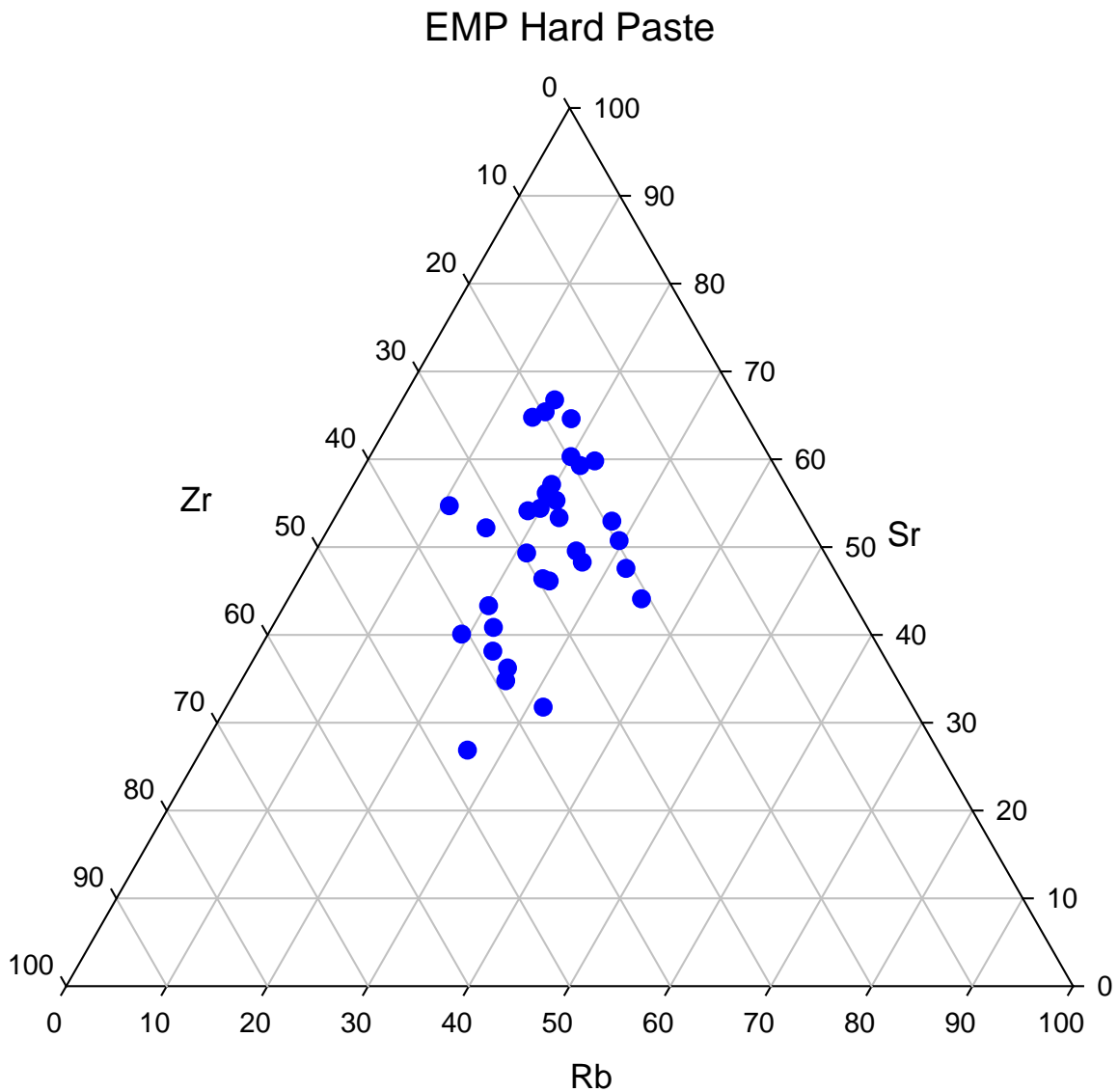
The Zr-Sr-Y graph produces a compressed cluster near the Zr axis. This graph offers minimal research opportunities and thus will not be used for further analysis.



The Sr-Y-Nb graph produces a compressed clusters at the Sr-Y axis. As the results are compressed to the corner, this graph will be excluded from further analysis in this research.

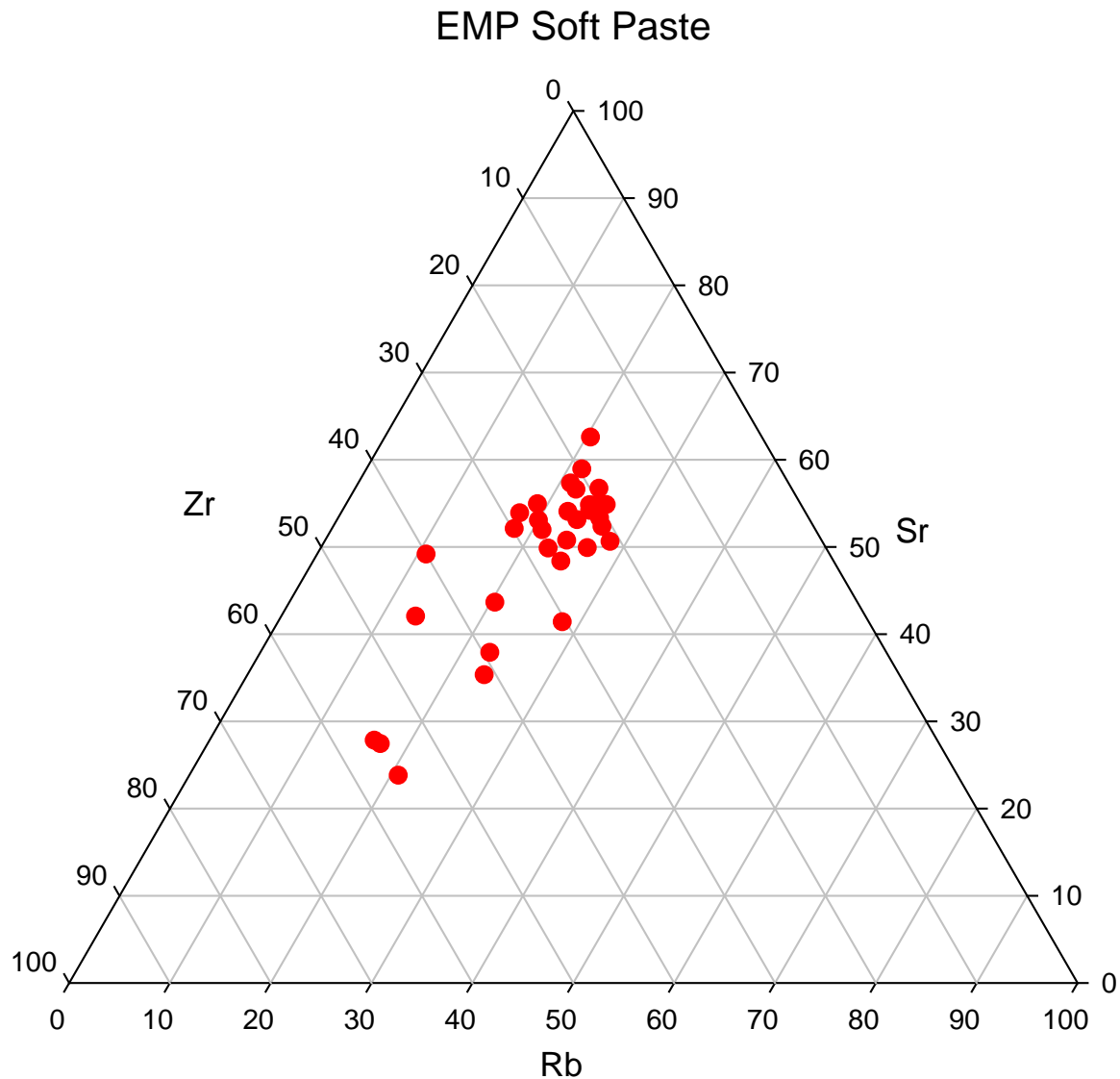


The Y-Zr-Nb graph produces a distribution that seems legible though compressed towards the Zr axis and the Zr-Nb corner. There are several smaller groupings visible within the major cluster, which makes this graph interesting for further analysis. However, the Rb-Sr-Zr graph shows more promise. In addition, the clustering is slightly compressed towards the Zr-Nb corner, which excludes this graph from further analysis.



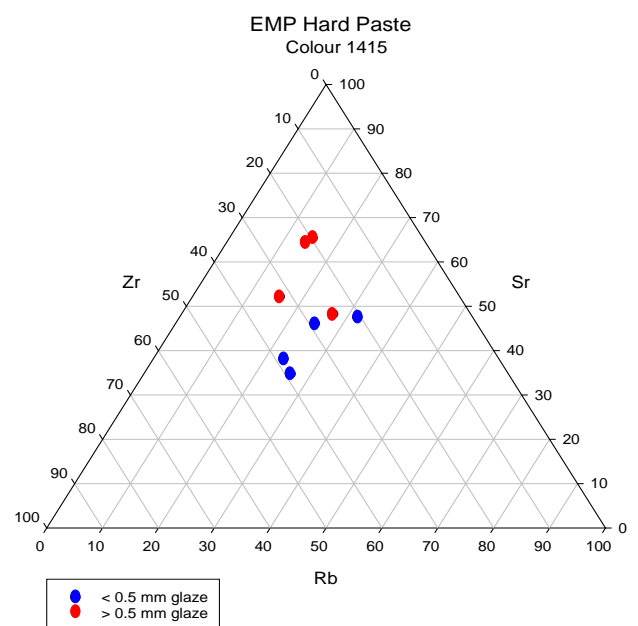
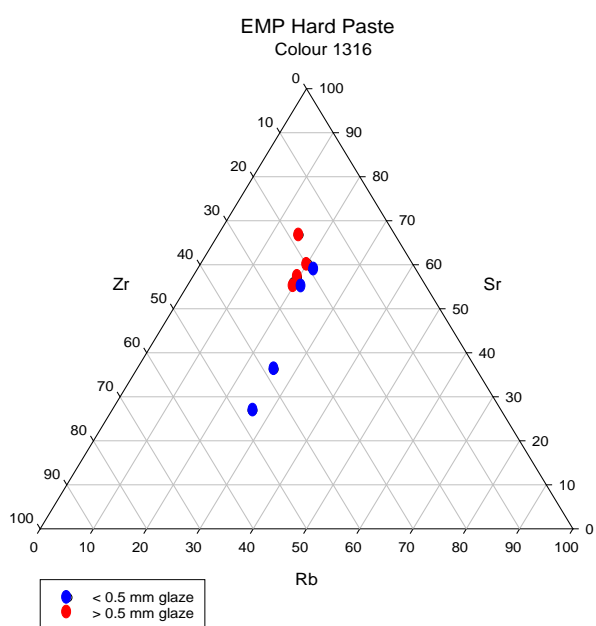
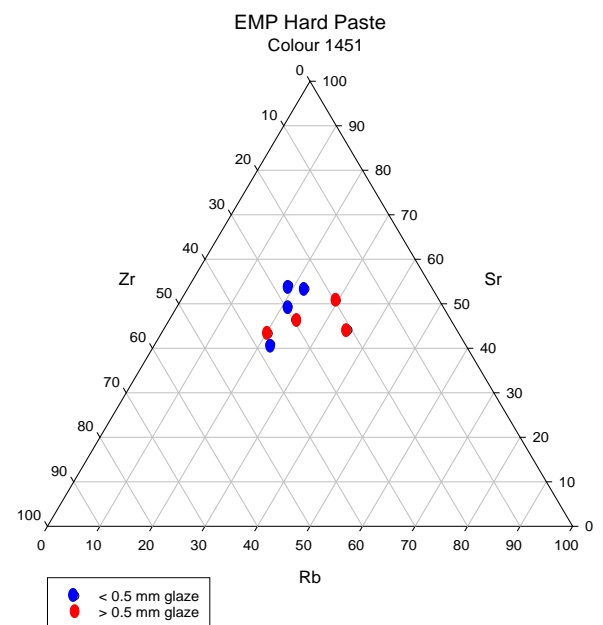
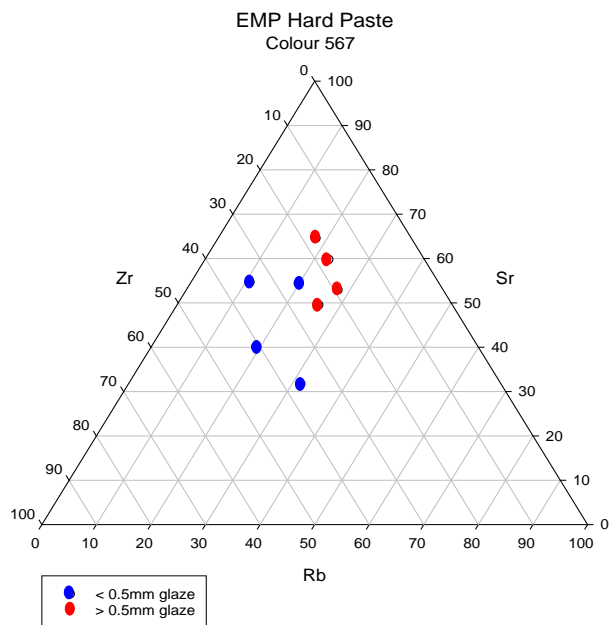
EMP hard paste, ceramic paste that cannot be scratched, even by a steel blade, produces what seem to be a number of clusters. This suggests that the hard paste was transported to Singapore from several different production centres. The outliers could suggest deviations in the standard production procedures. The possibility that the outliers belong to yet another few production centres is slim, considering that they are not located far outside existing clusters. Yet, this possibility cannot be entirely discounted, as the clusters do not spread far apart.

The fact that all the artefacts are retrieved from a fourteenth century soil layer could suggest that the outliers are time specific. The possibility that some of the objects are of older age may explain why they fall outside the clusters. Source material keeps degrading and changing glaze production, likely improved over time, is an important aspect of the ceramic production process.

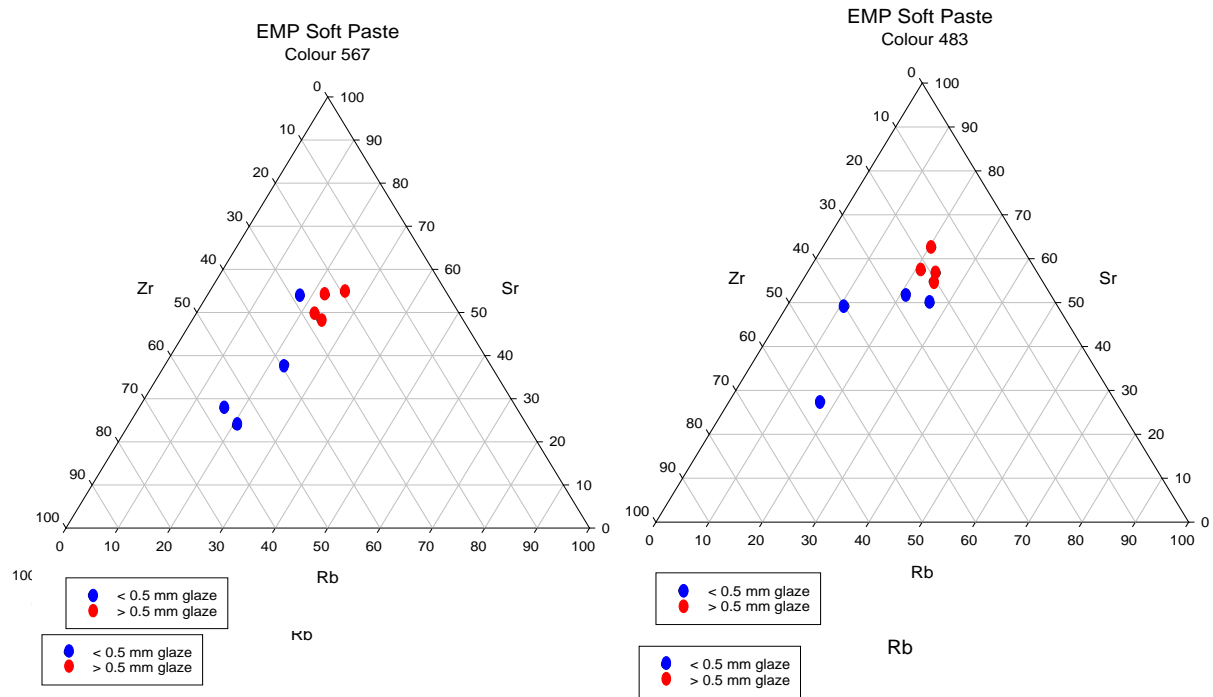


EMP soft paste, ceramic paste that can be scratched easily, at first glance produces a large cluster with six outliers and a small cluster of three further away. Though the outliers could be seen as clusters, I either would suggest they are temporal anomalies, production deviations, or produced elsewhere with different source material. The small group of three could be a different production centre in a different region; more research is needed to test this hypothesis and to identify conclusive provenance.

Upon closer analysis of the large cluster, four smaller groupings can be identified. This suggests that all these objects are likely produced using the same source material but possibly at different kiln sites within the large production region. The clay used was likely to be from the same source material but the differentiation in the results is possibly due to the slight difference in glaze production. This could be the identifying factor for the small clusters. This train of thought then suggests that during the EDXRF testing the glaze and clay are both being tested. However, that would render the outliers as miss-productions or miss-tests. There is the possibility that the paste colour then could become the identifying factor for the small groupings.

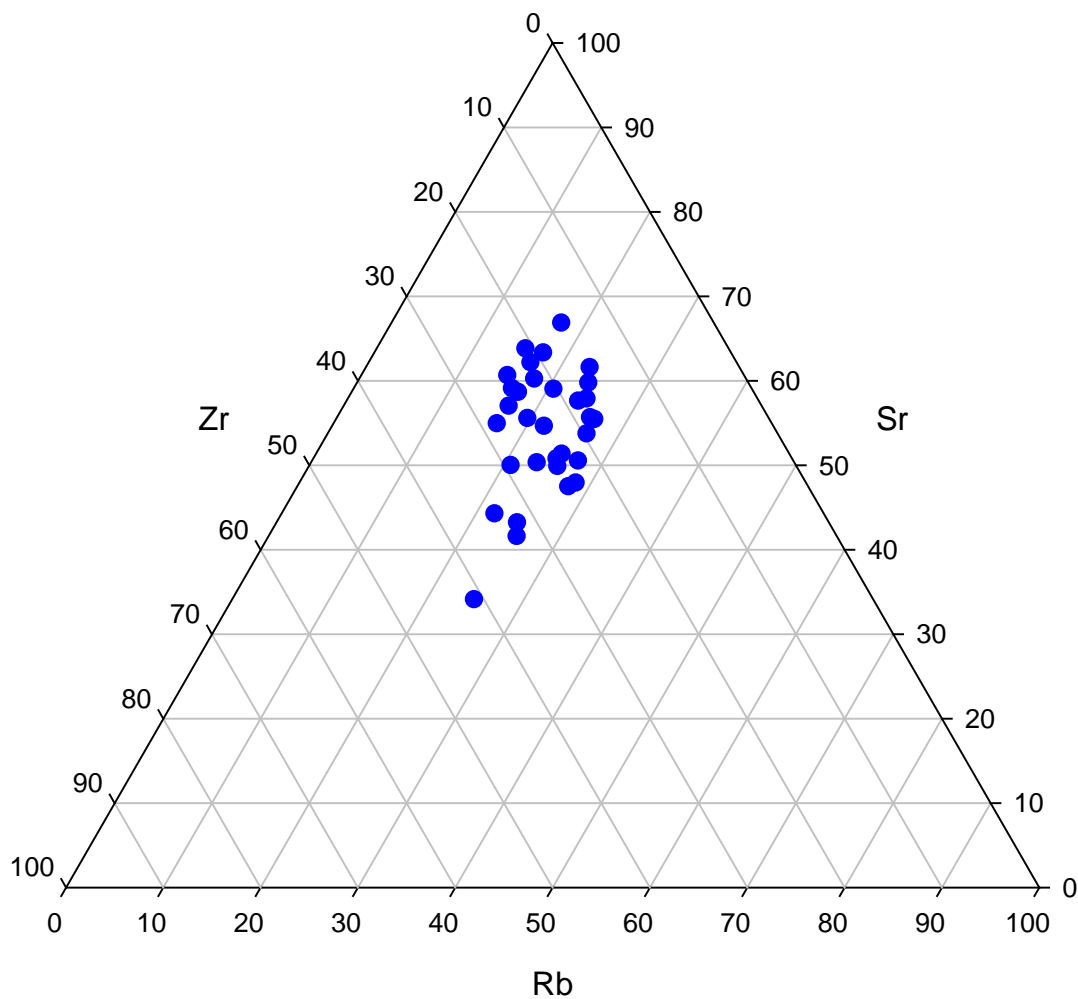


Both the paste colour and glaze thickness of the EMP hard paste are compared in the above graphs. Neither seems to render any more information on the specifics of the EMP hard paste clustering. The paste colour and the glaze thickness of the EMP hard paste can henceforward be ignored.



As with the EMP hard paste, the EMP soft paste colour and glaze thickness do not provide any further clues regarding the clustering of the results.

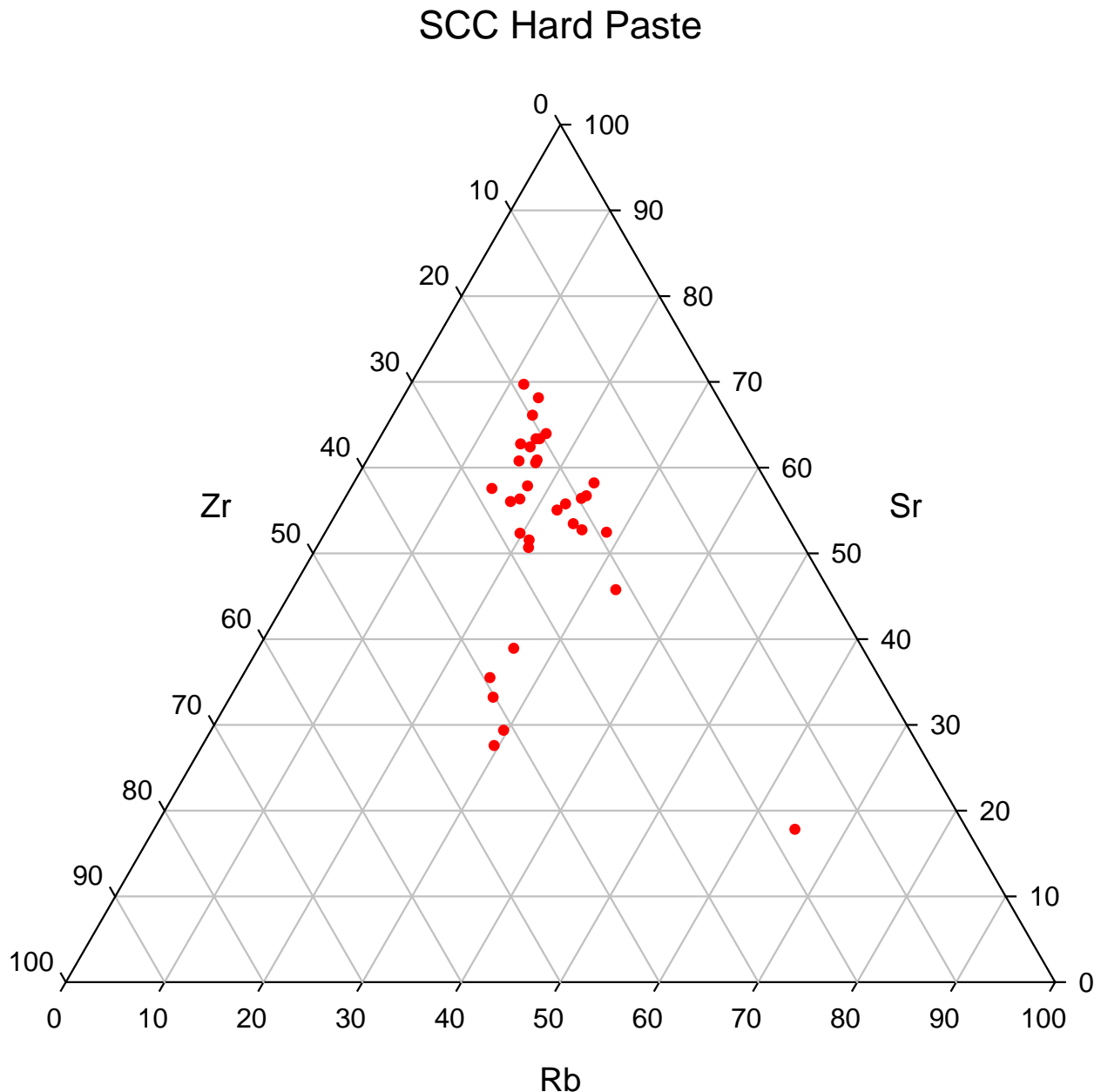
SCC Soft Paste



The SCC soft paste, for which 32 test results were compared, here shows a large oval cluster with minor clusters within it. There are five small clusters within the large oval grouping and one outlier just below it. This, again, can suggest that the same source material was used but that there are minor differences in the purification of the clay and the production of the glaze. This suggests that each kiln site had a slightly different production procedure for the clay and glaze, which is possible if we factor in the possibility that relevant technology has always been improving. Another hypothesis can be that, instead of clay purification, different additions could have been made to the clay. Yet another train of thought could be that the different smaller clusters are actually different source materials. This research does not include the testing of source material; hence it is not possible to exclude this hypothesis. The last thought could well be a viable one for the simple reason that the recipe for Greenware cannot vary much between production centres in China. Since the above comparison is done on trace elements, this could suggest that the results are dependent on the regional setting.

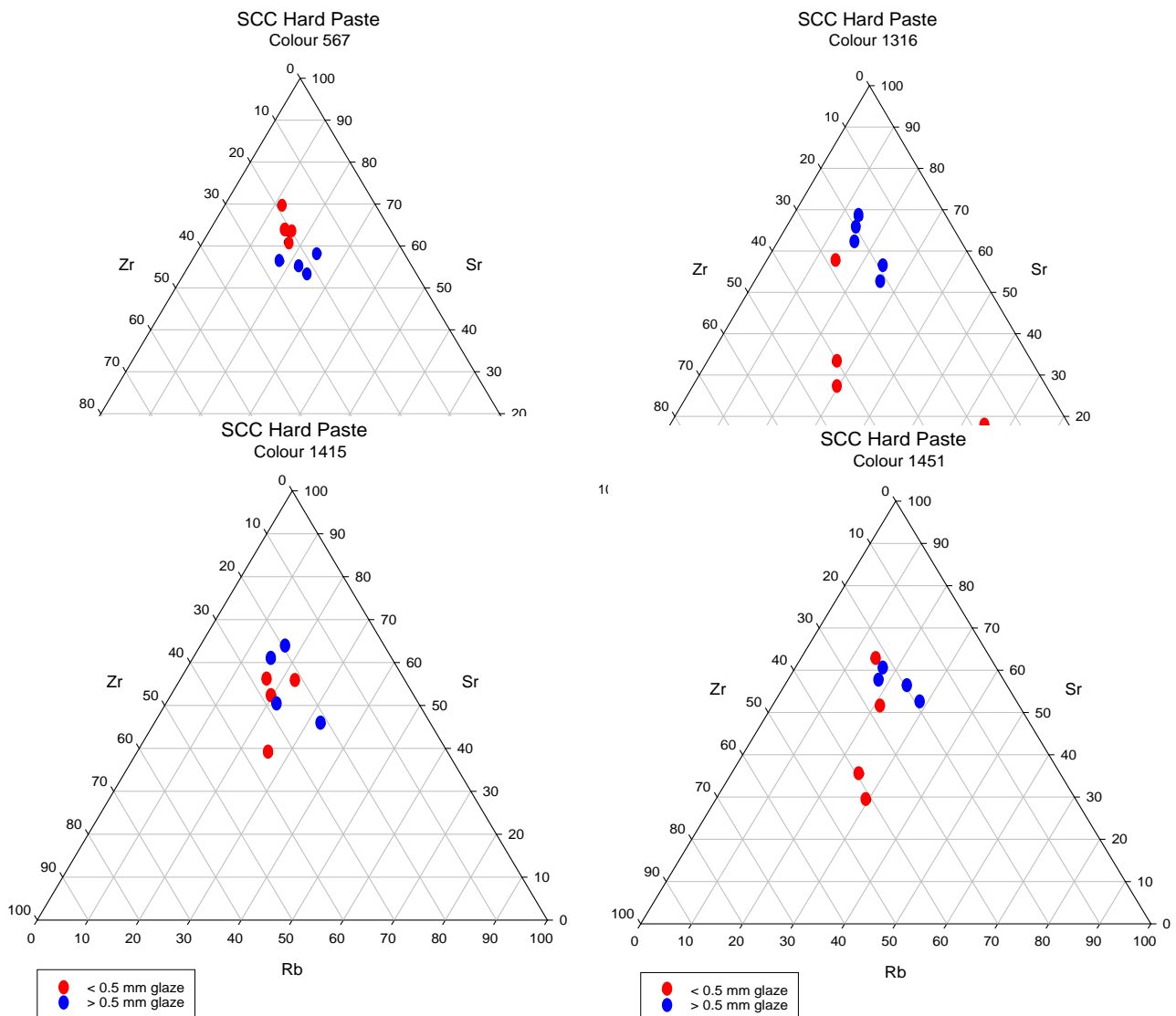
In order to be complete one cannot exclude a last hypothesis, which suggests that the clusters could indicate a different time of production. This is not in the sense of time of year, as here we are looking at trace elements, which are not likely to be influenced by seasonality. We could possibly look at the production of similar material from different years. It is possible that this type of comparison would be better identified by using 2D scatter plots. The likely development of degradation of the clay, and its trace elements, would show clustering in either an upward or a downward succession.

The elemental isotope half-life then comes into consideration. What is the life expectancy of trace elements used in the above graphs? This will be further discussed on page 75.

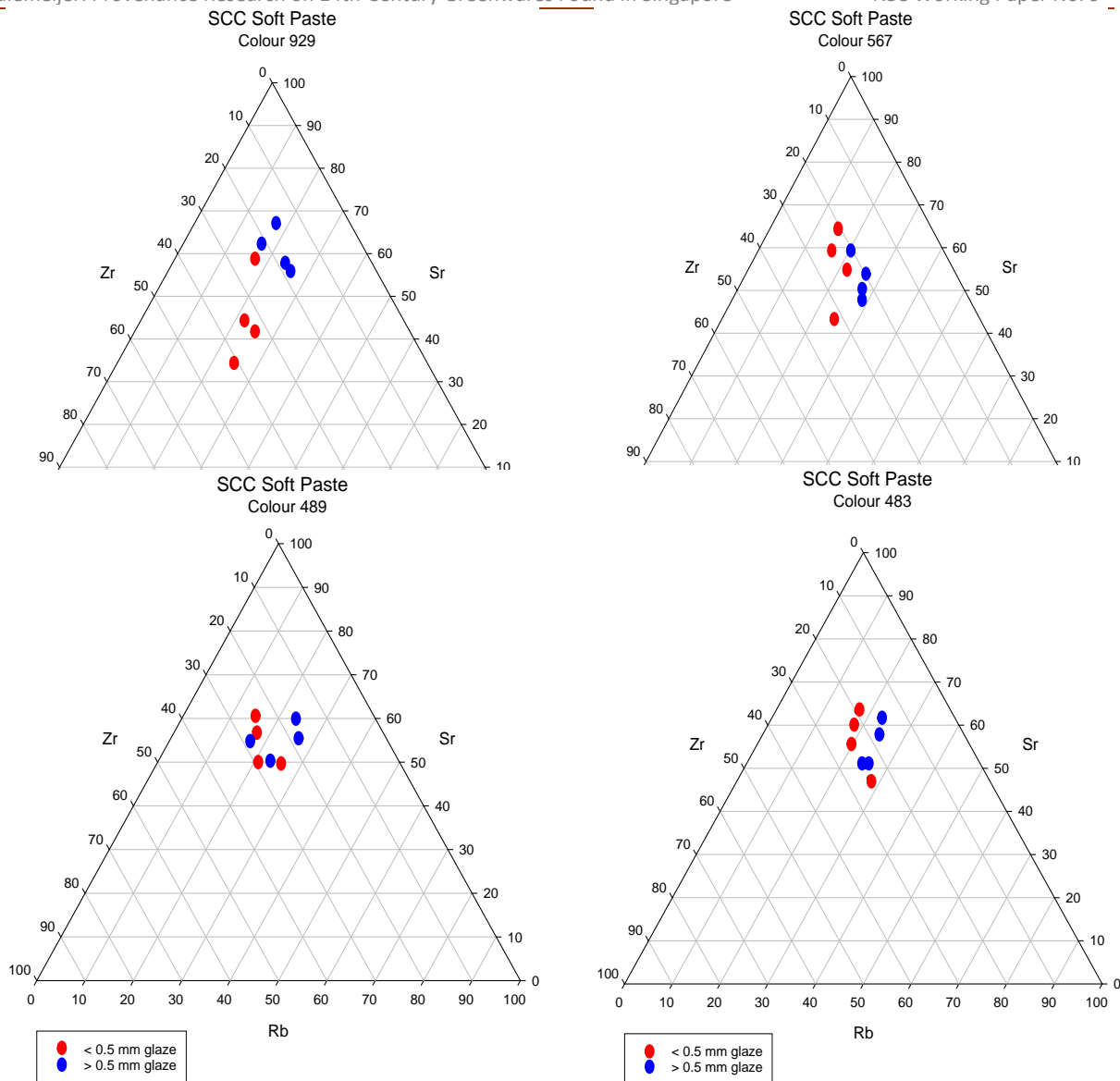


SCC hard paste again shows clustering of the results: five distinct groups with an elongated group below and two outliers. This could indicate that since there are so few outliers, only specific quality pieces were transported to the area where at the present time the Singapore Cricket Club resides. As there are no specific records of what was at the SCC site in the fourteenth century, further analysis of the entire forty thousand artefacts retrieved from the SCC site could possibly clarify this point. This combined with an excavation spanning a large area with total analysis of the assemblage and chemical fingerprinting of all ceramics would be needed to come to a better understanding of this site in ancient history.

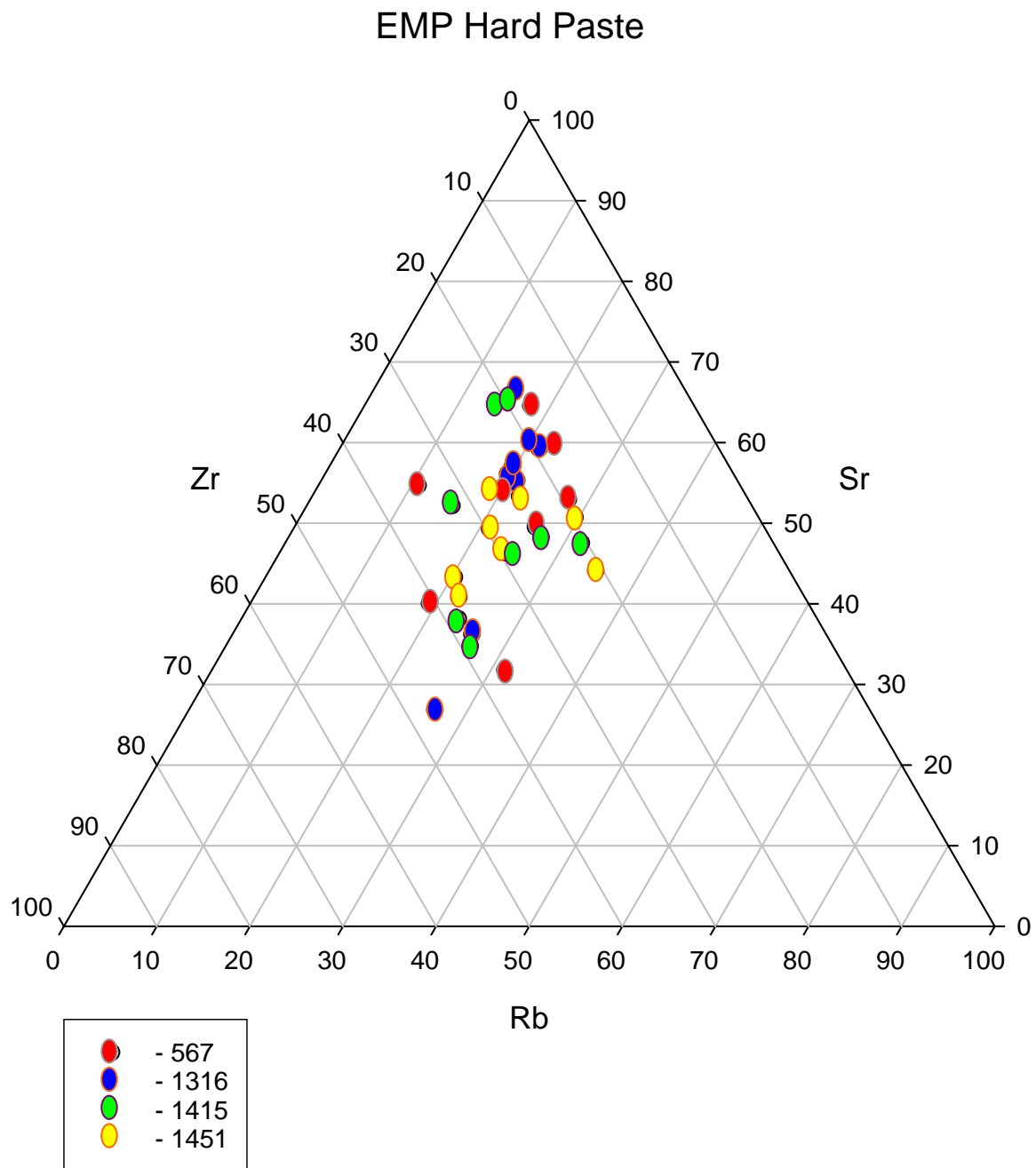
The suggestion of high quality hard paste Greenwares at the SCC site implies certain affluence at the site. Whether at material comfort was enjoyed at the site or some other location nearby cannot be answered at this time. Here again the same questions can be asked about the clustering, hence these questions need to be addressed and the scatter plots need to be produced.



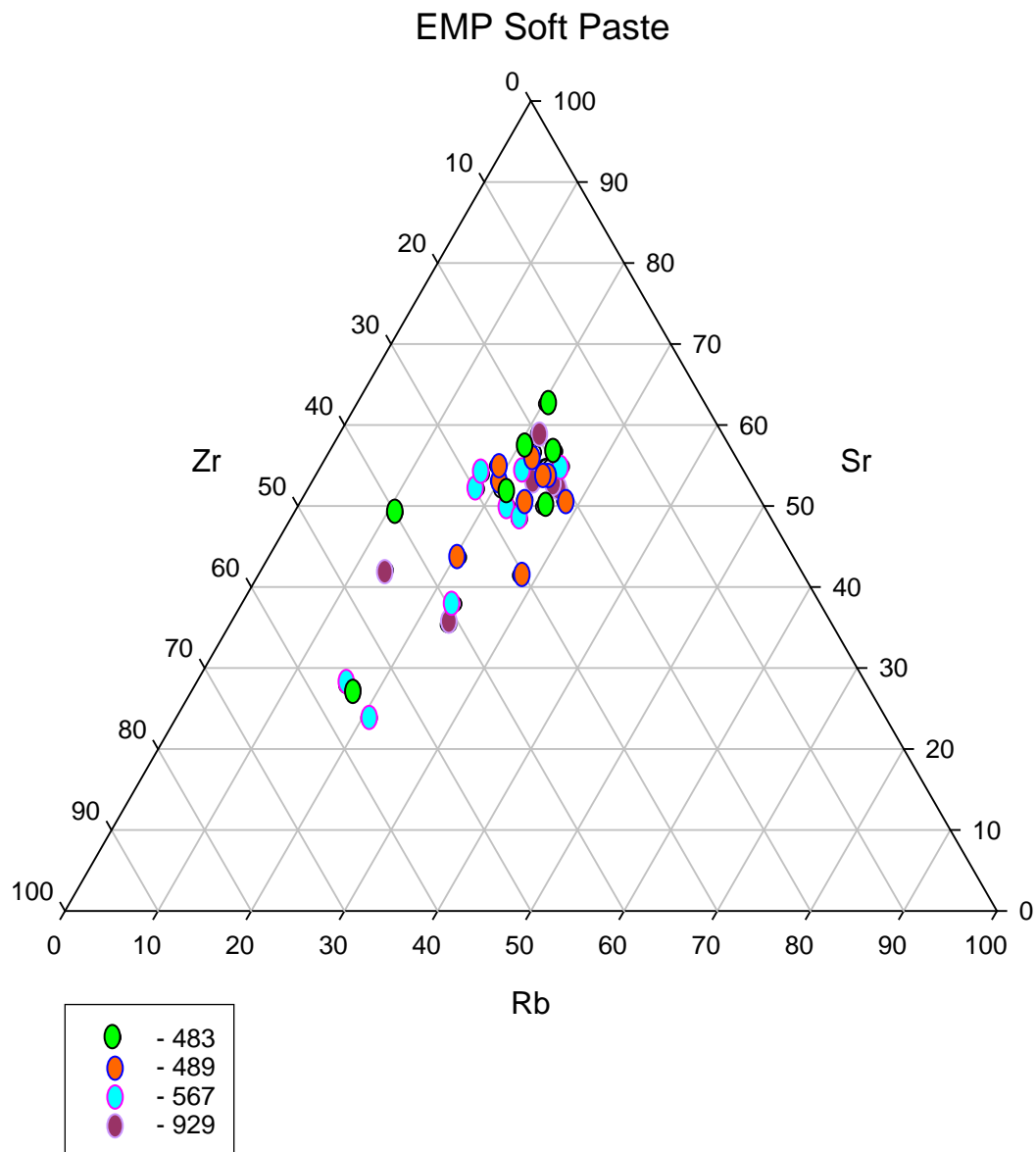
The SCC hard paste and glaze thickness do not offer any more clues regarding hypotheses offered earlier.



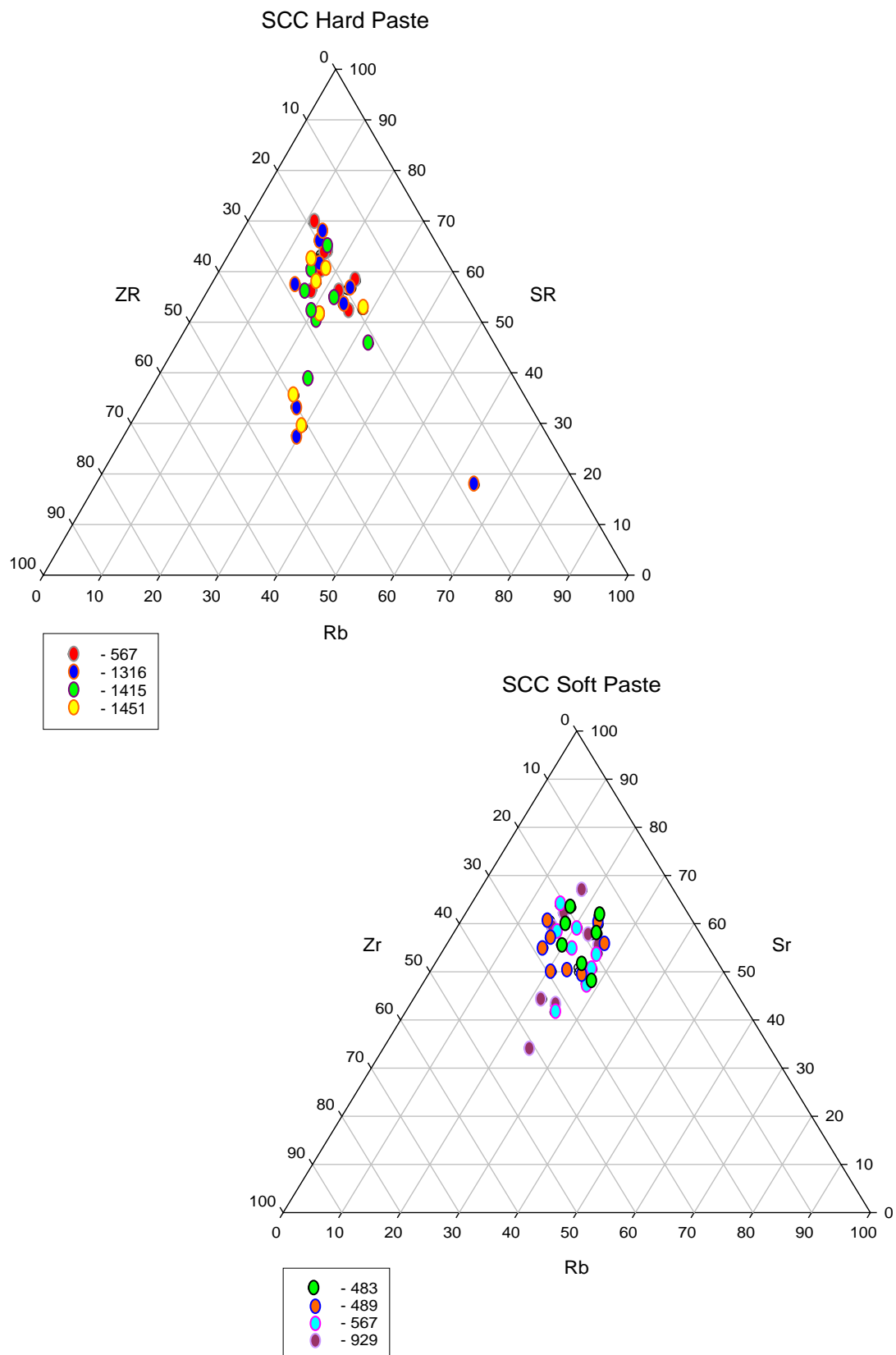
The SCC soft paste colour and glaze thickness also do not represent any possible explanation of clustering.



The sorting system established before testing of the subjects consisted of a first tier, which is site differentiation; the second tier division is between hard paste and soft paste. Comparing the third stage of paste colours within the EMP and SCC hard paste tier, it becomes clear this is not a meaningful differentiation within the hard paste collection.



The comparison of the soft paste paste colours within the EMP and SCC soft paste sub-division, of the sorting system, shows that the different paste colours are not useful to identify differentiation within the soft paste collection of either excavation.

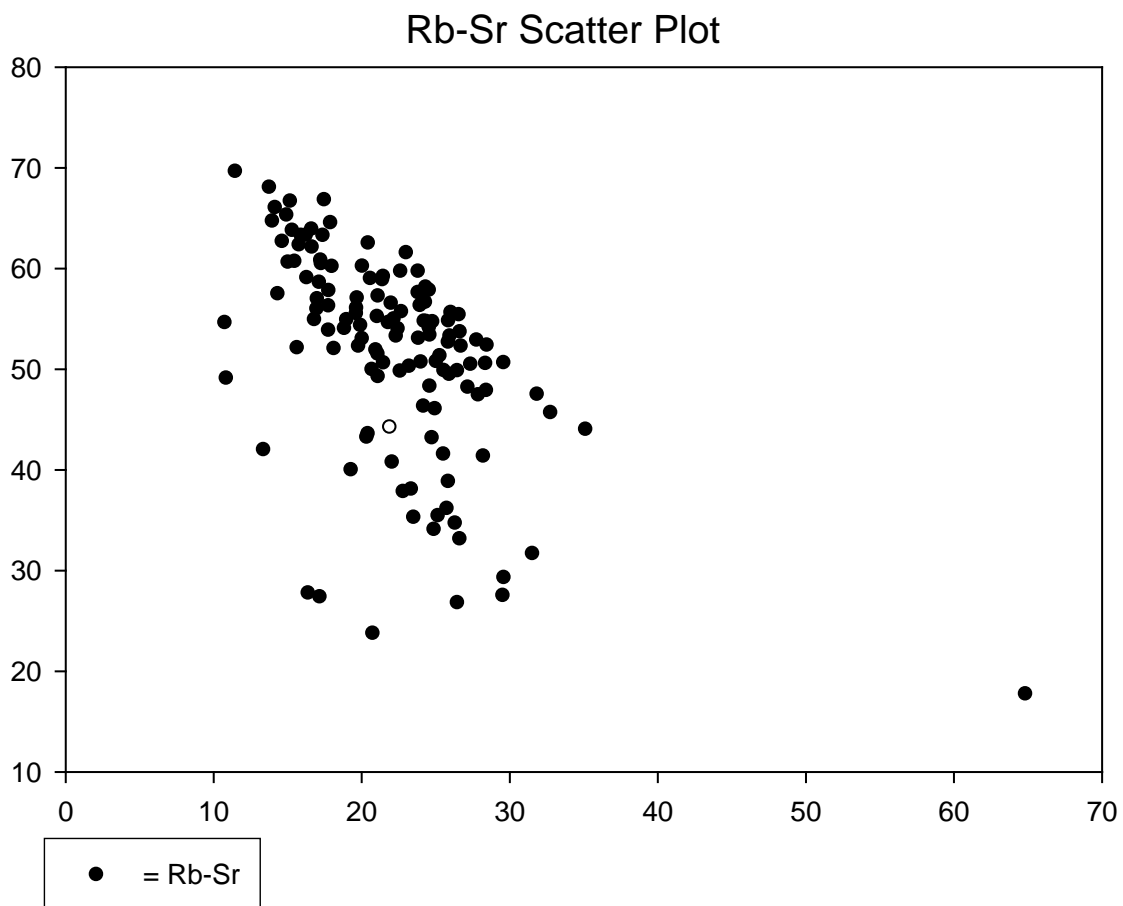


It is clear that the paste colour tier does not generate any significant clustering. In addition, the tier that compares glaze thickness also does not generate any additional information for further analysis. The above graphs do show that there seems to be more correlation between the Greenwares found at SCC than the objects recovered at EMP.

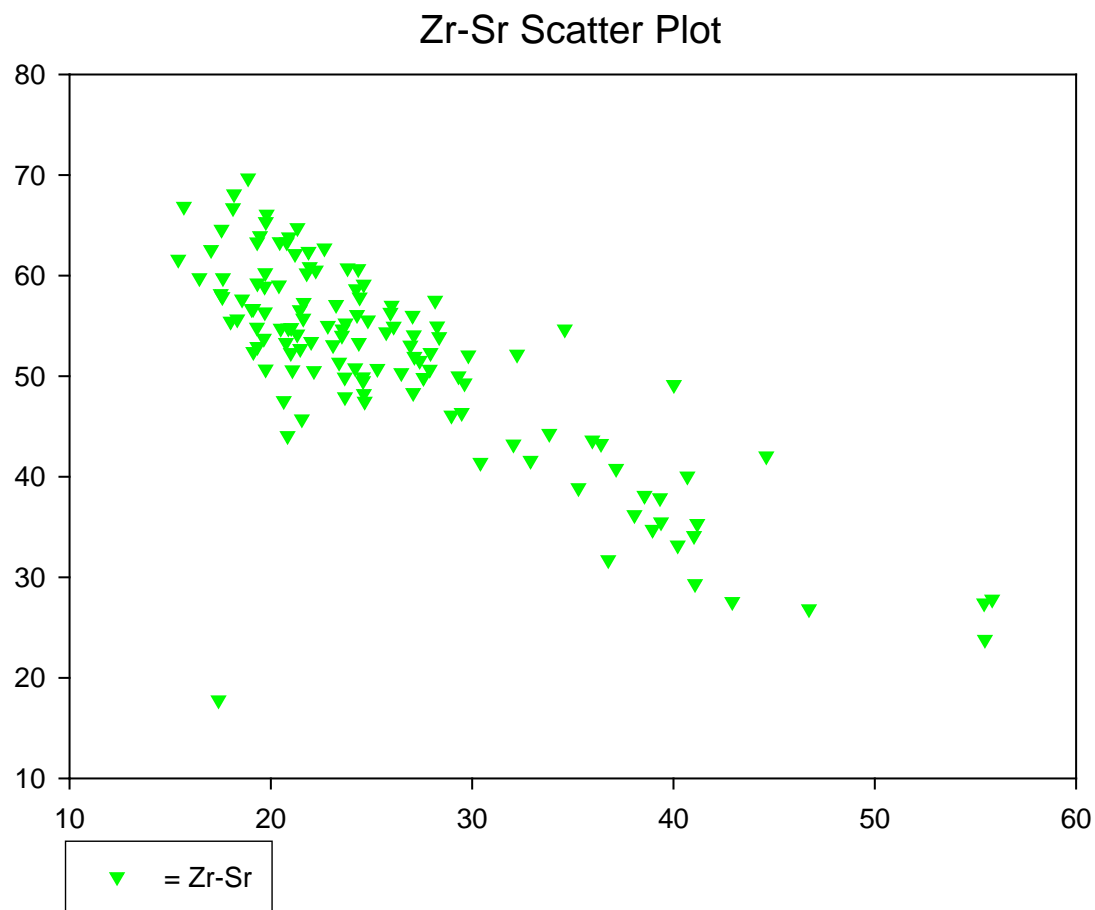
The sorting system on page 68 can thus be reduced to site, soft paste, and hard paste. The inclusions of paste colour and glaze thickness have thus been reduced to novel ideas but which are not useful in the scientific chemical fingerprinting analysis. The only significant clustering difference is found between the EMP and the SCC subjects.

As the ternary graphs show, the combination of elements that produces the most readable combination should also be exploited for the scatter plot comparison. Rb, Sr and Zr compared with each other add 3 graphs for closer analysis.

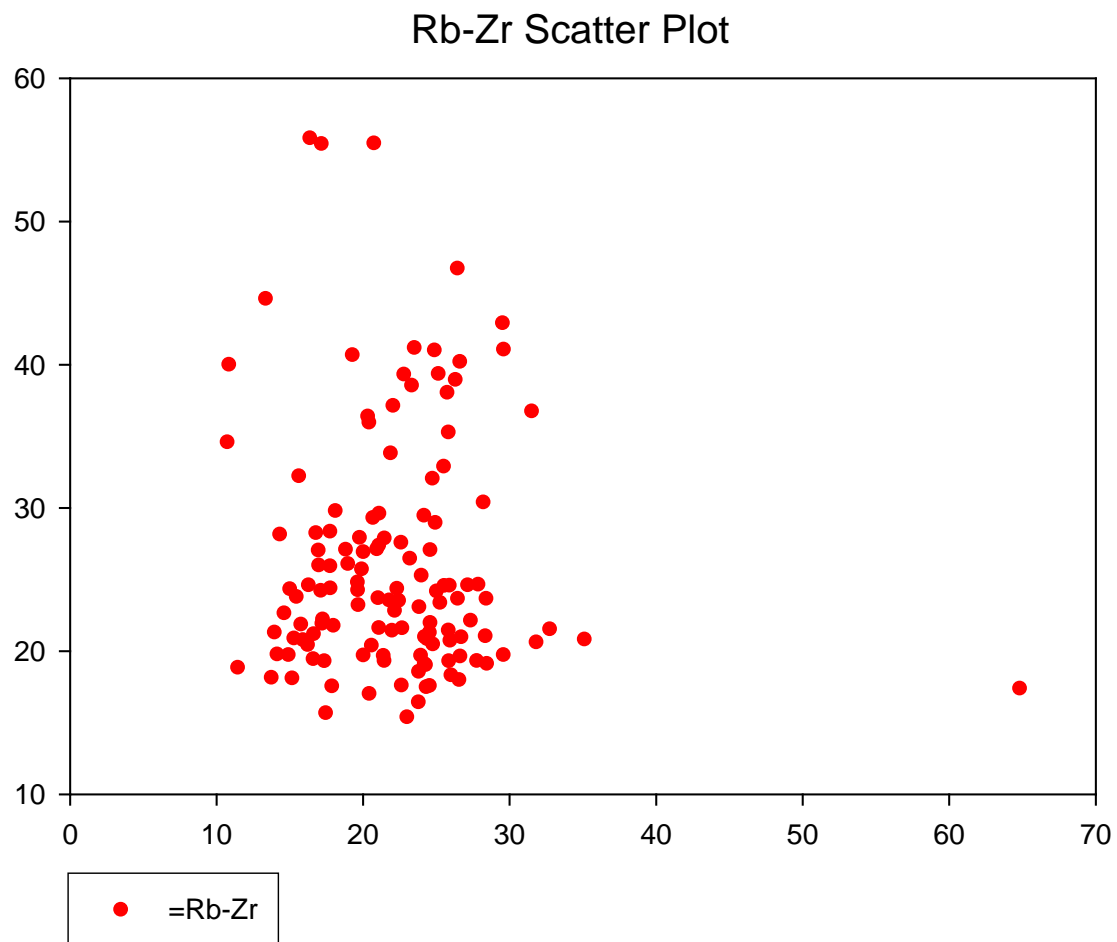
12. Scatter Plot Analysis



The elements Rb-Sr produce a graph that physically looks similar to the ternary graph of three elements on page 50. This combination of elements will be examined in more detail later.

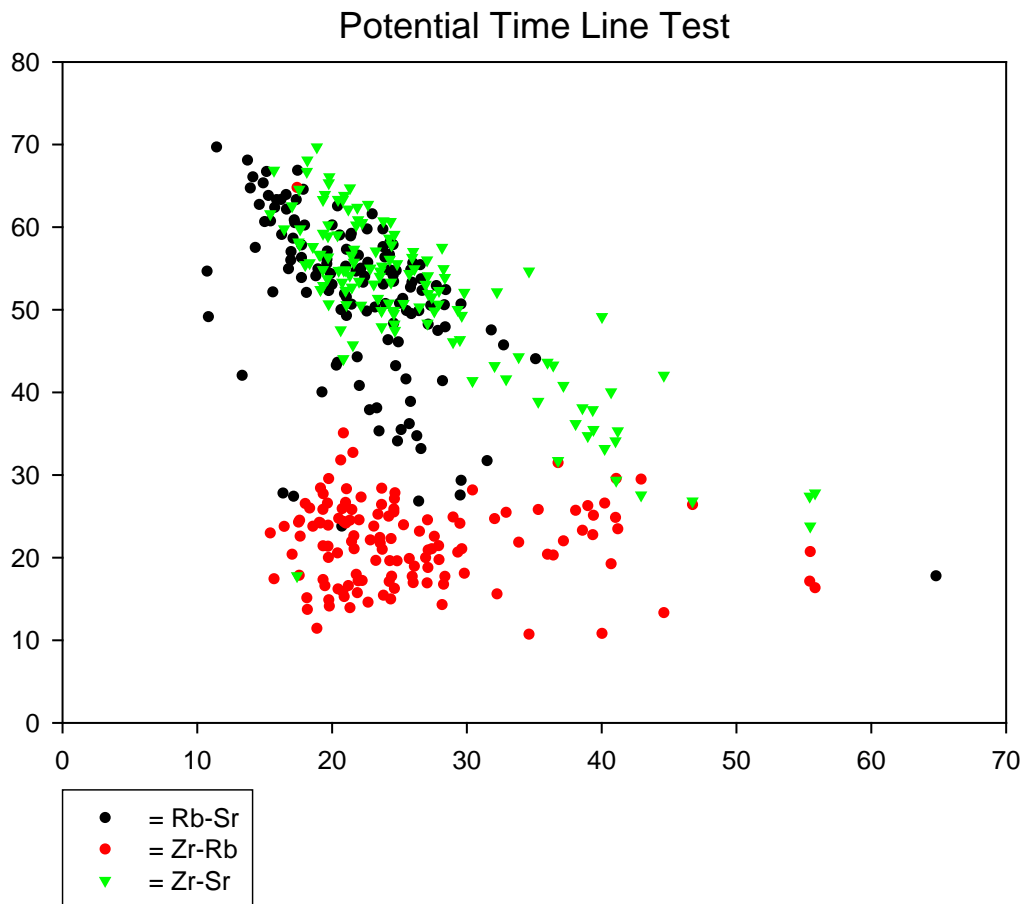


The Zr-Sr comparison produces a slightly elongated readout. This could be an indication of time but before any statements along those lines are made more comparisons need to be completed.



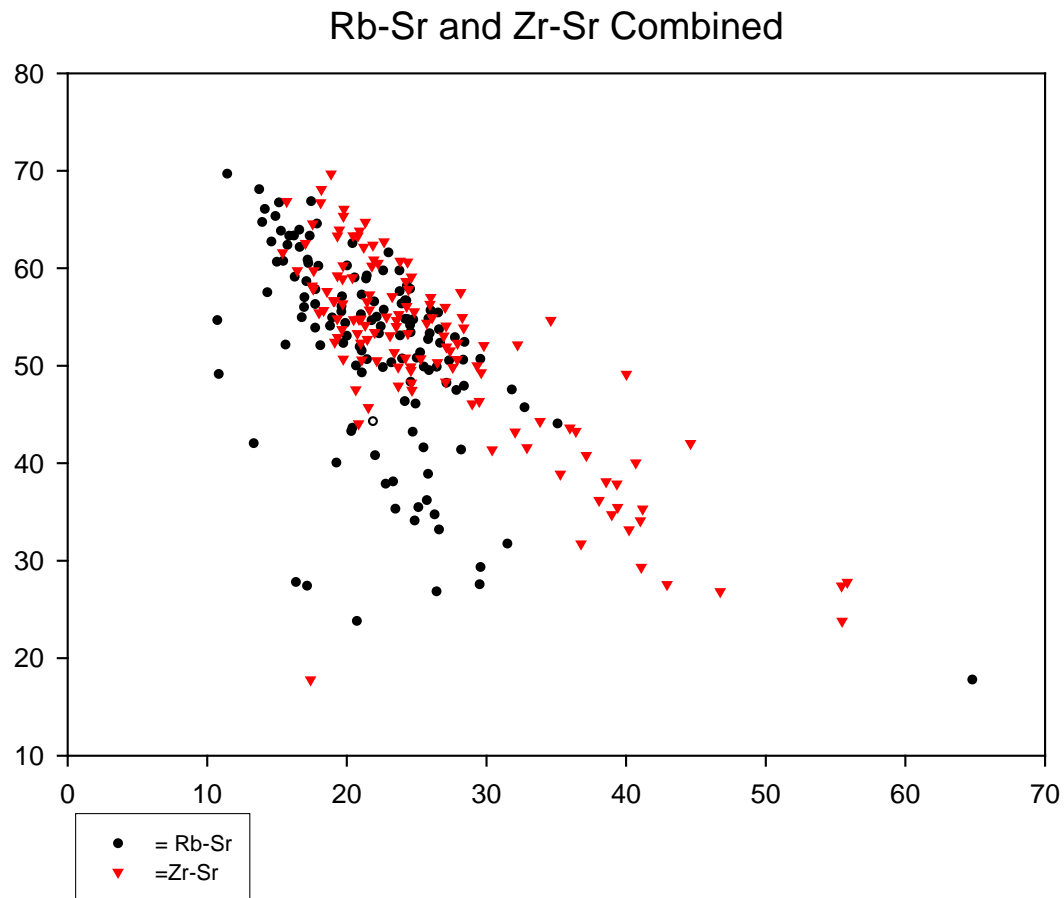
The Rb-Zr comparison creates a different cluster than the previous two. Though physically similar to the ternary graph on page 51, but in reverse, there seems to be no real increase or decrease of test results.

The two previous graphs of Rb-Sr and Zr-Sr will be further explored for possible temporal indications, excluding the Rb-Zr graph for reasons given.



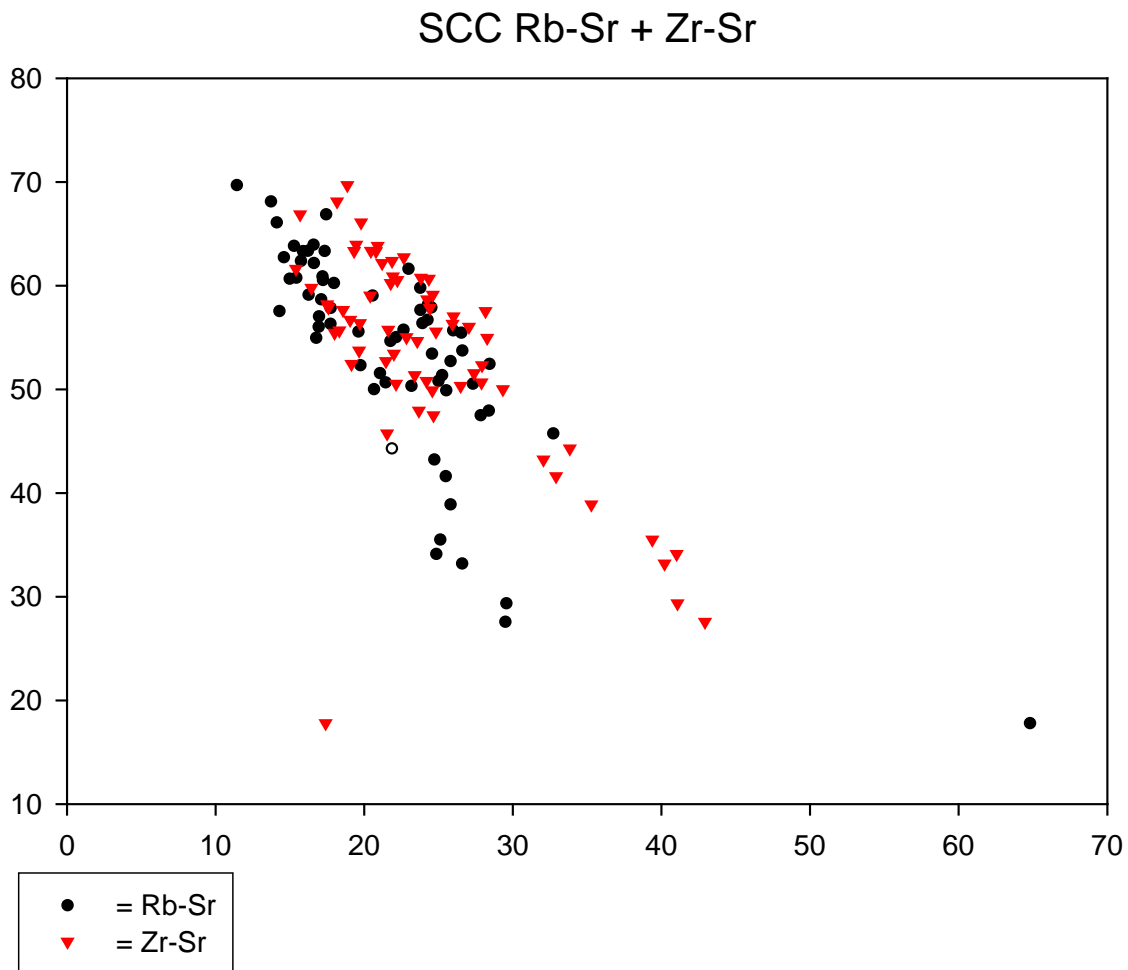
When comparing the three combinations together in one graph it becomes clear that there is no significant statement that can be made about the Zr-Rb combination. When one is comparing elemental development or degradation in a time context it is to be expected that elements will increase in order to fit their natural development or visa versa. Zr-Rb does seem to have only one element that increases; this could mean that Rb, on the Y axis, is not affected by time. The consideration of elemental half-lives is not necessary here as the half-life, or life expectancy of the elements focussed on in this research, are all well over thousands of years. Compared to the other two graphs this is likely not true. Could this mean that the Zr-Rb combination may possibly be more indicative of place of production, or source material? There is a need for more specific research to draw any conclusions on that subject. The Zr-Rb combination will henceforth be excluded from this research.

Rb-Sr and Zr-Sr will be combined in future graphs for greater comparison.

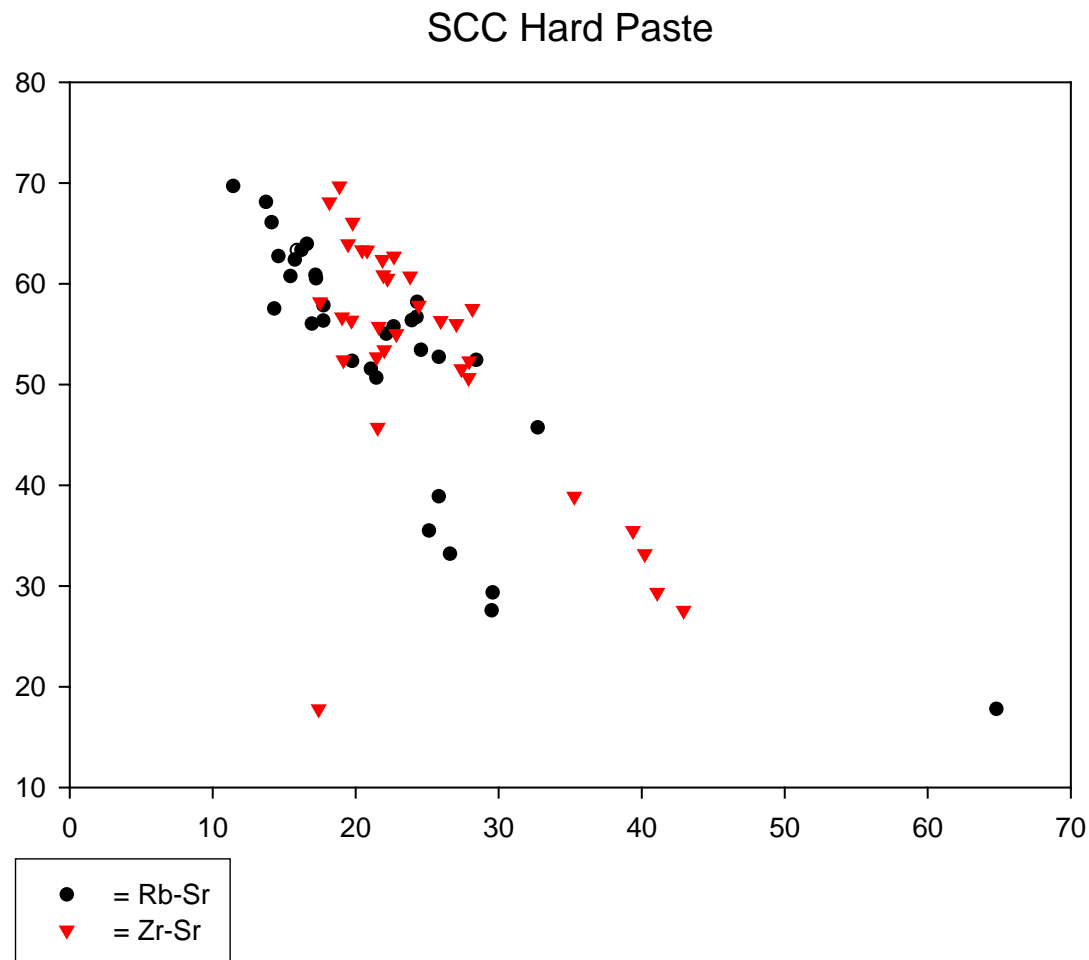


The Rb-Sr combination has a slightly less diagonal development compared to the Zr-Sr combination, yet both have over thirty test results positioned outside of their main cluster. As the ternary graph on page 51 suggested, the larger number of sherds will fall within the main cluster. In both these comparisons, there are more test results outside the main clusters than in the ternary graph comparison.

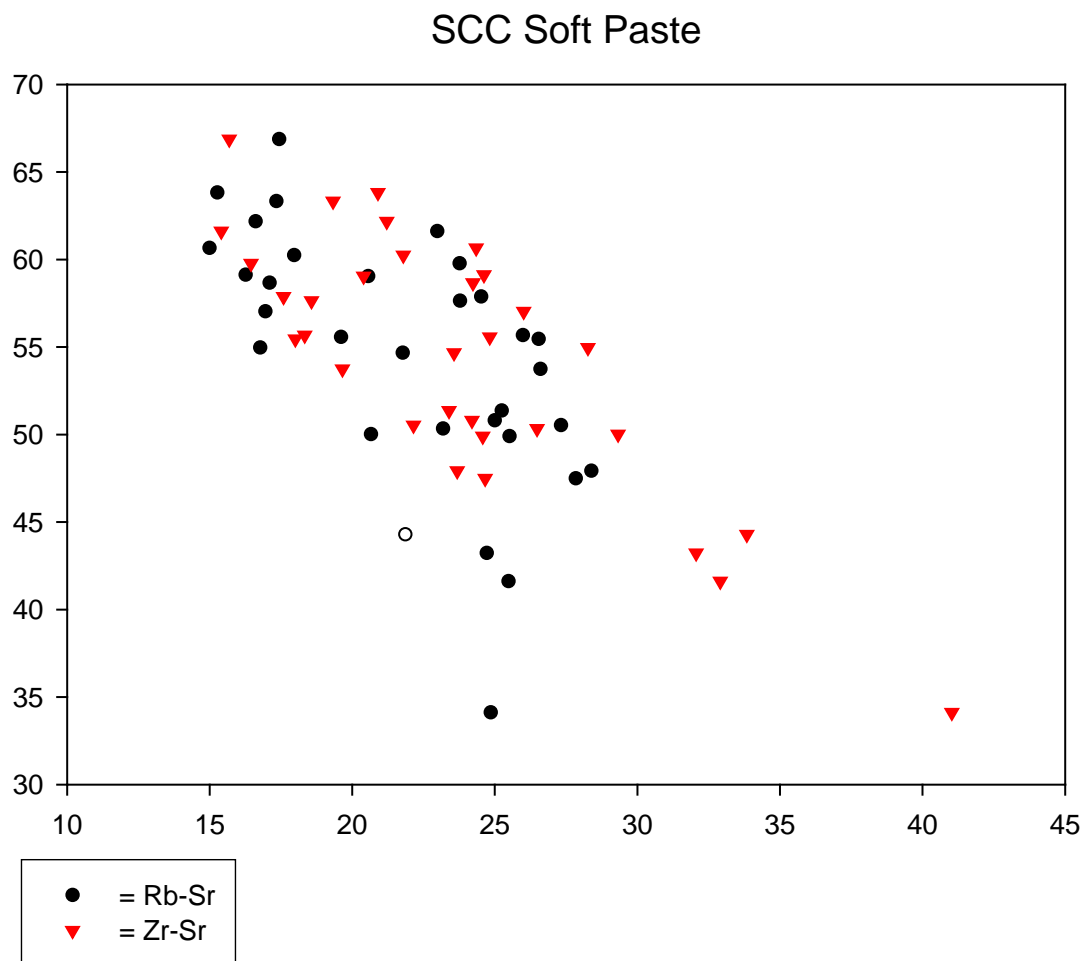
Could this be an indication of time? More artefacts need to be tested in order for any conclusions to be drawn from these results here.



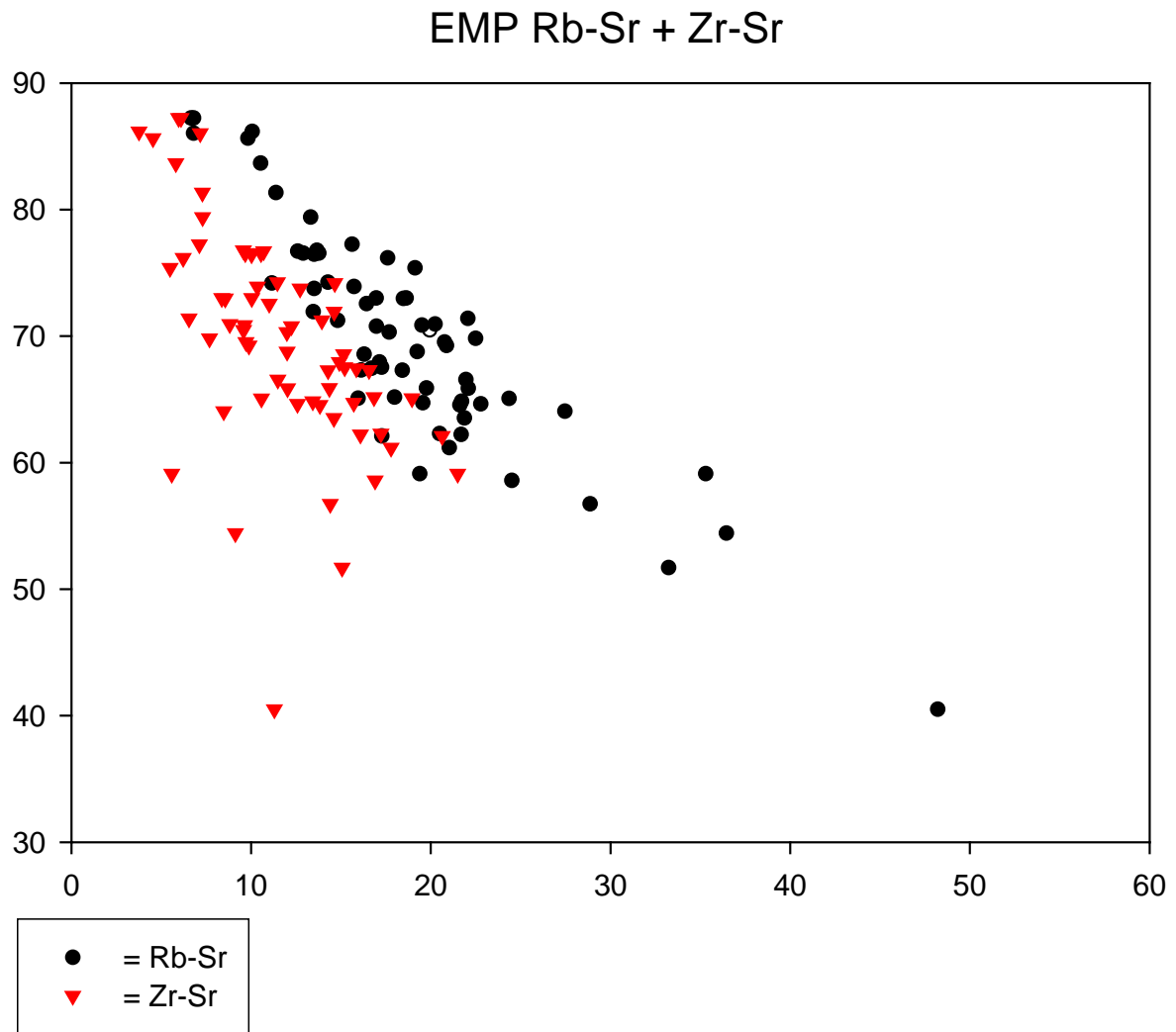
At first glance, this graph seems difficult to read. Upon closer inspection, it becomes clear that in both combinations the main cluster comprises between the 10 and 30-percentile point on the X axis and the 50 and 70-percentile point on the Y axis. In addition, this suggests that Sr (in both comparisons on the Y axis) decreases over time but in tandem it also suggests that Rb and Zr increase over time. From a chronological point of view, this could suggest that most of the artefacts were produced in a similar period. From a provenance perspective, this indicates that most objects share a similar production centre, but not necessarily a similar kiln site.



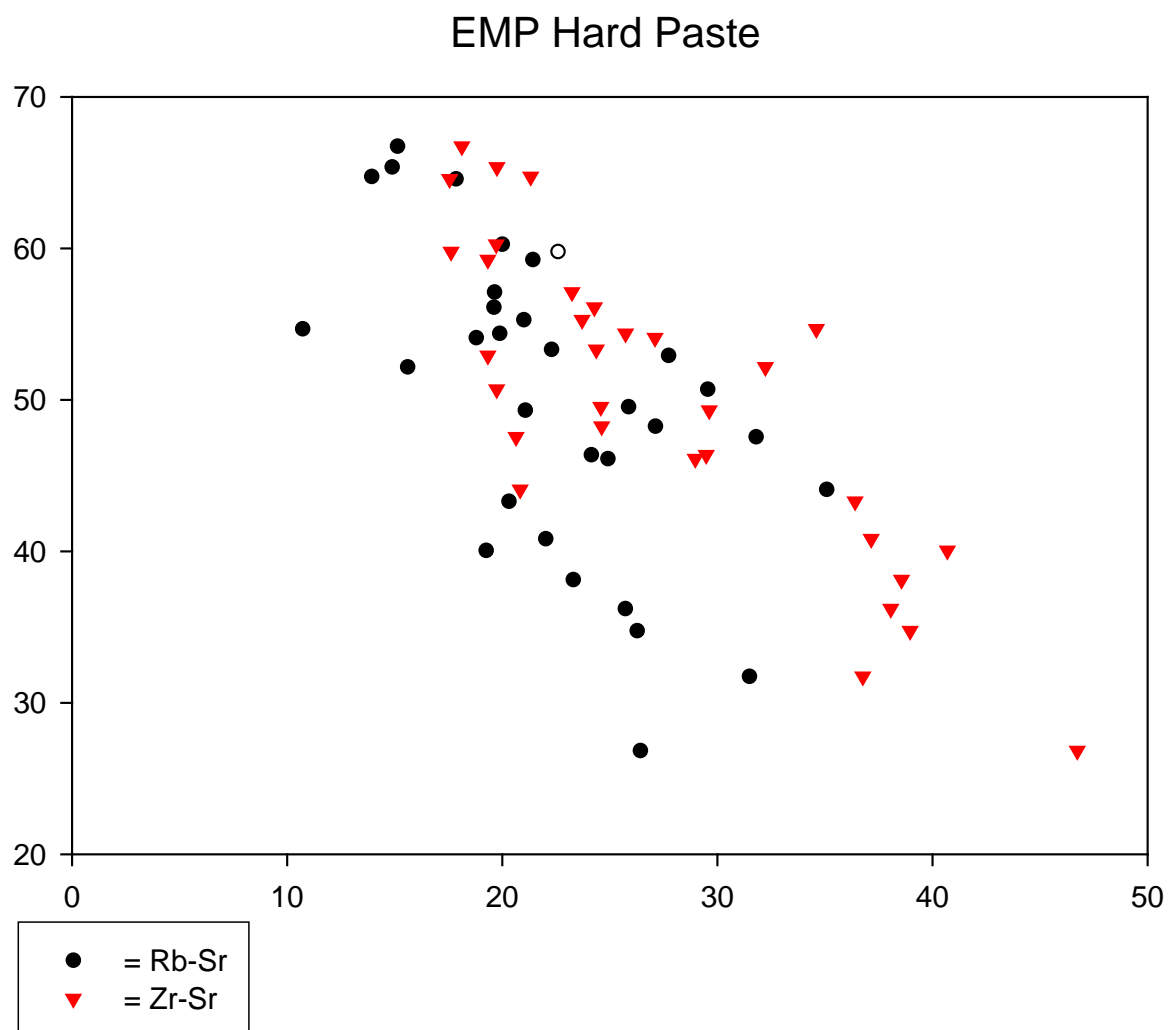
Comparing the SCC hard paste it seems that there is a significant improvement in the readability of the graph. Again, as in the relevant ternary graph page 76, both comparisons produce what seem like either a time line or an indication of several production centres. Both elemental combinations show a diagonal line of small clusters. The bulk of these small clusters are situated in the same area indicated previously, which from a chronological point of view could indicate that these objects were produced at the same production centre but at different times, hence the decrease in Sr counts. If this were right, it is difficult to explain why Rb then is increasing.



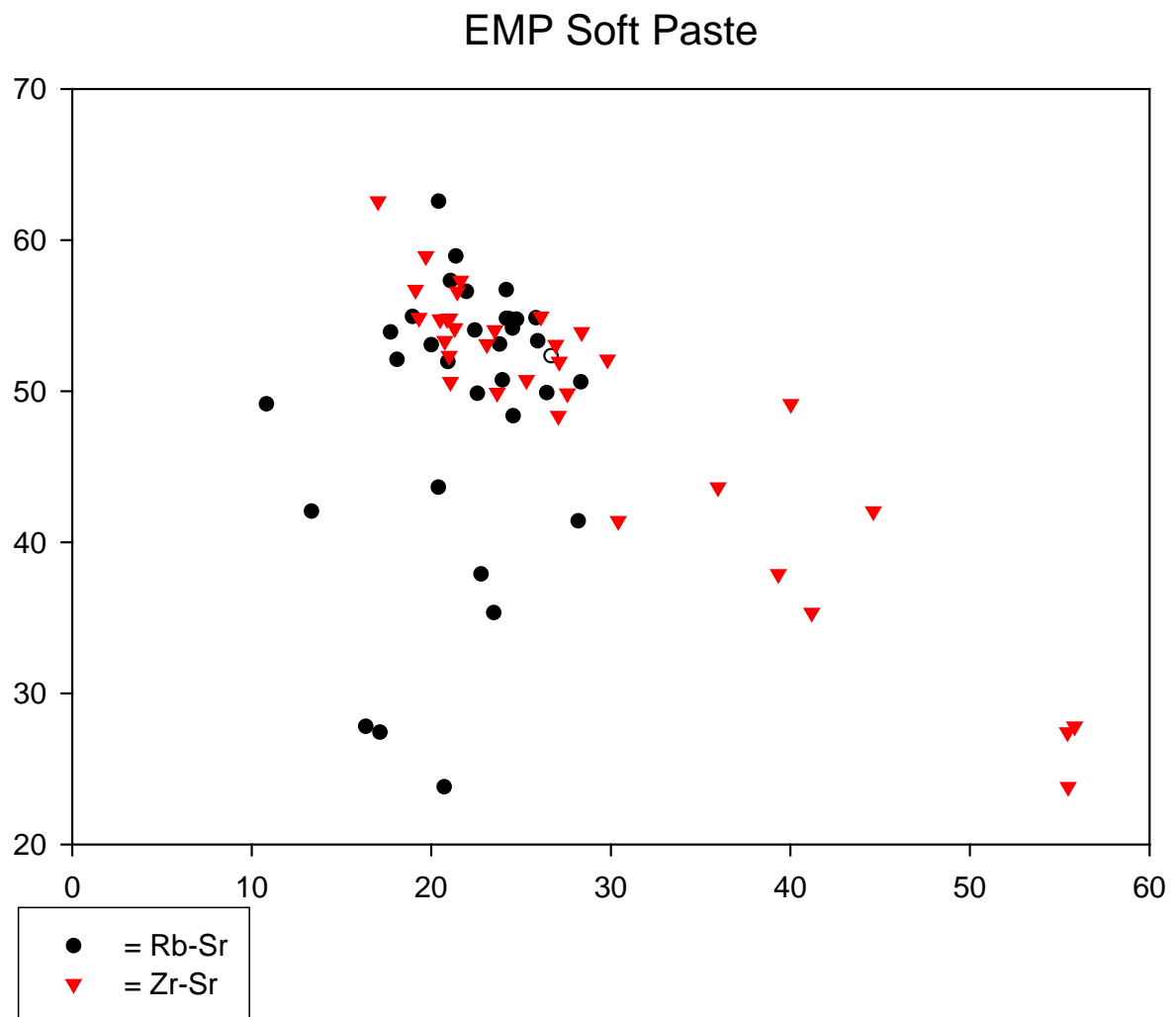
The SCC soft paste seems to produce a very different picture than the SCC hard paste, where the relevant ternary graph (page 78) shows a fairly tight cluster. Here due to the shorter X- and Y- axes the results are more spread out. Though this spread seems confusing, it does suggest that the soft paste sherds from SCC are of a similar time and/or a similar production centre, as most of the results cluster within a relatively small percentile point range.



The comparison of the EMP sherds is confusing. The EMP results have produced a higher Y axis value than in previous comparisons. As none of the data has been altered in any way I cannot explain this anomaly. In addition, where the other graphs produce higher values for the Zr-Sr elemental comparison on the X axis, here for some reason the values are reverse. All methods involved are executed in the same standard way as all the other scatter plots yet the results are abnormal. I have no explanation for the oddness of these results.

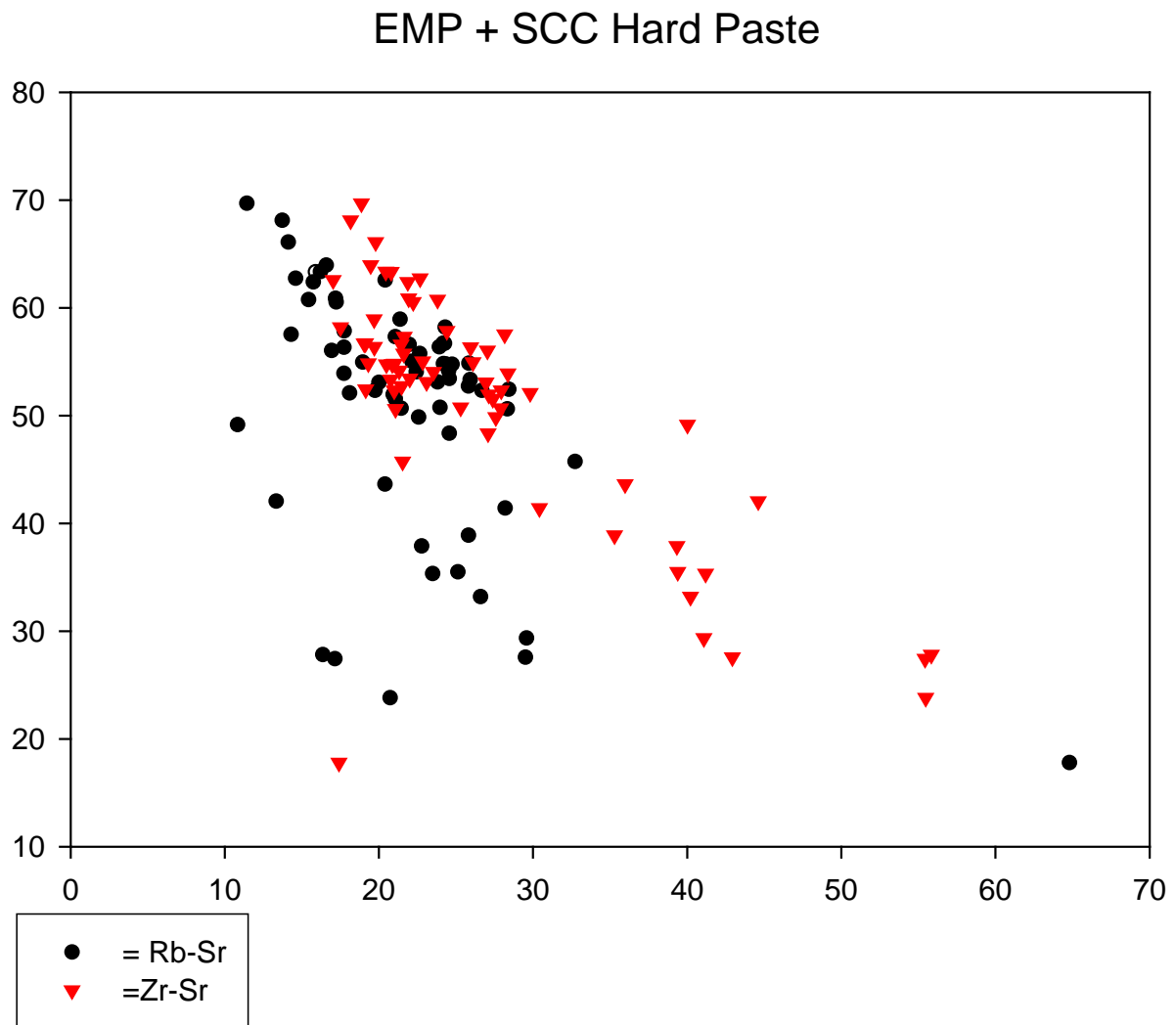


Although, this graph seems confusing, most of the results fit within the 20-40 percentile point on the X axis and between the 45-70 ranges on the Y axis. As the results are so closely related, it is likely that the artefacts share either a similar time of production or a similar production centre. The minor clusters could indicate the same batches of artefacts produced at the same time at the same, or nearby kiln sites.

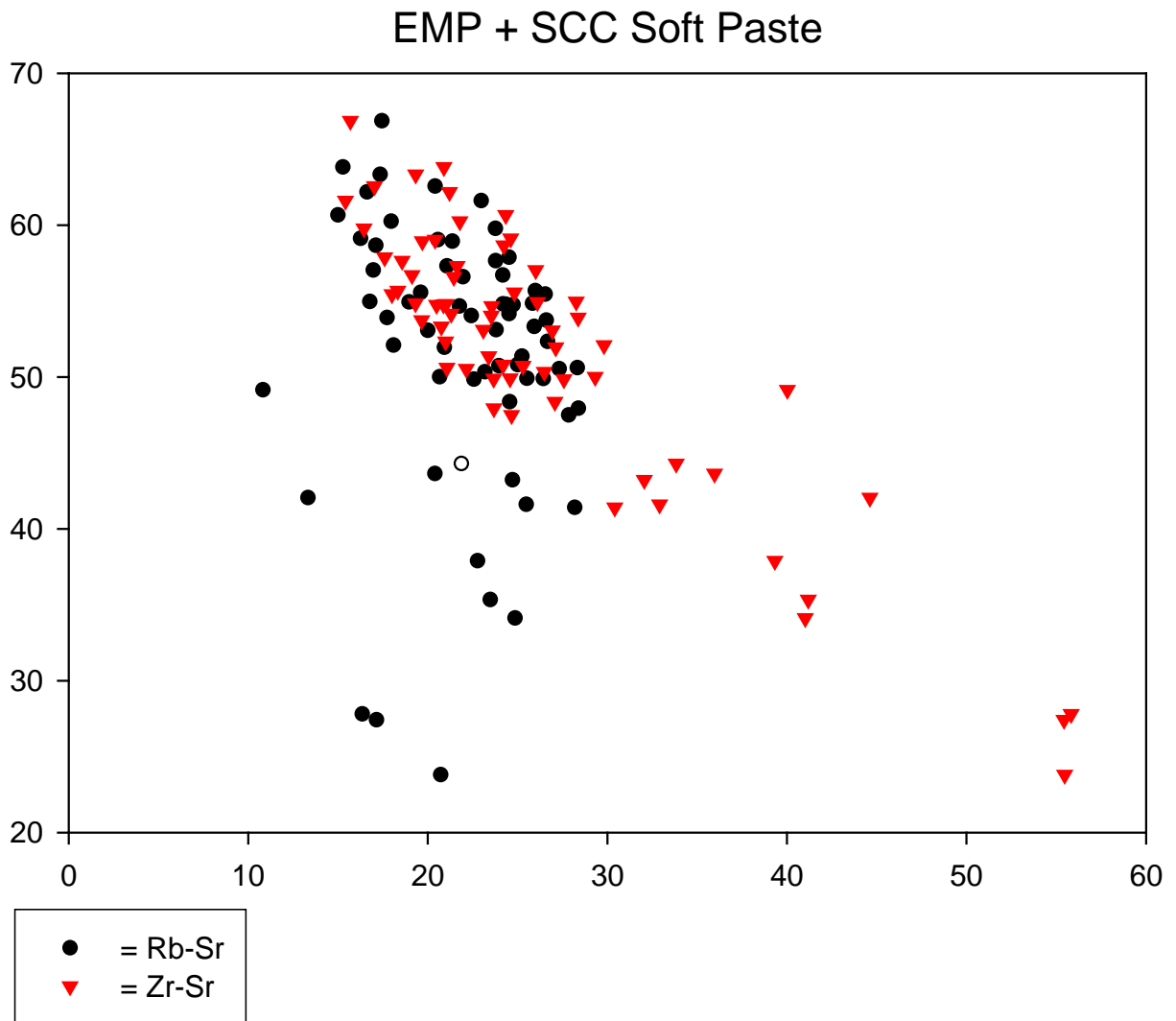


A tight cluster of EMP soft paste for both comparisons again suggests a similar time and/or production centre. The spread of the outliers is nearly identical but mirrored. As previously mentioned the small clusters could indicate the same batch of artefacts produced at the same time at the same kiln site.

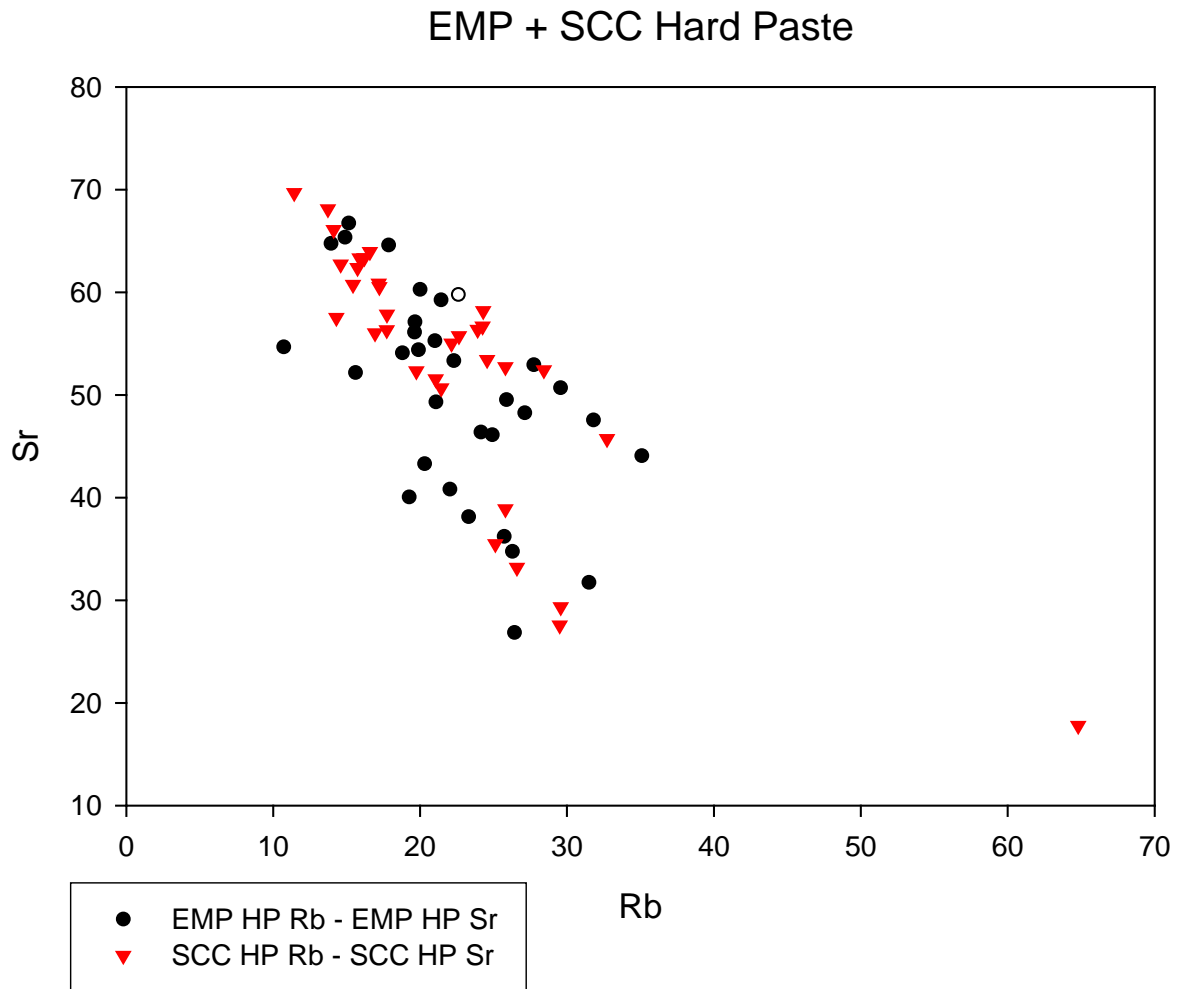
The two last graphs seem to have produced normal data again; neither is over the 80-percentile point.



The EMP hard paste showed a wide spread of results but that was due to the short axes. Here in combination with the SCC hard paste the EMP results are more compact. Further in this research the two excavation sites will be combined within one graph but then comparing only one dataset, this might increase the readability of this data. Both element combinations produce an elongated string of results, though the bulk of results cluster closely again. This could suggest, yet again, that the objects were produced in a similar time and/or production centre. The separation of elemental comparison where the sites are coloured differently will likely lead to a better understanding of this graph.



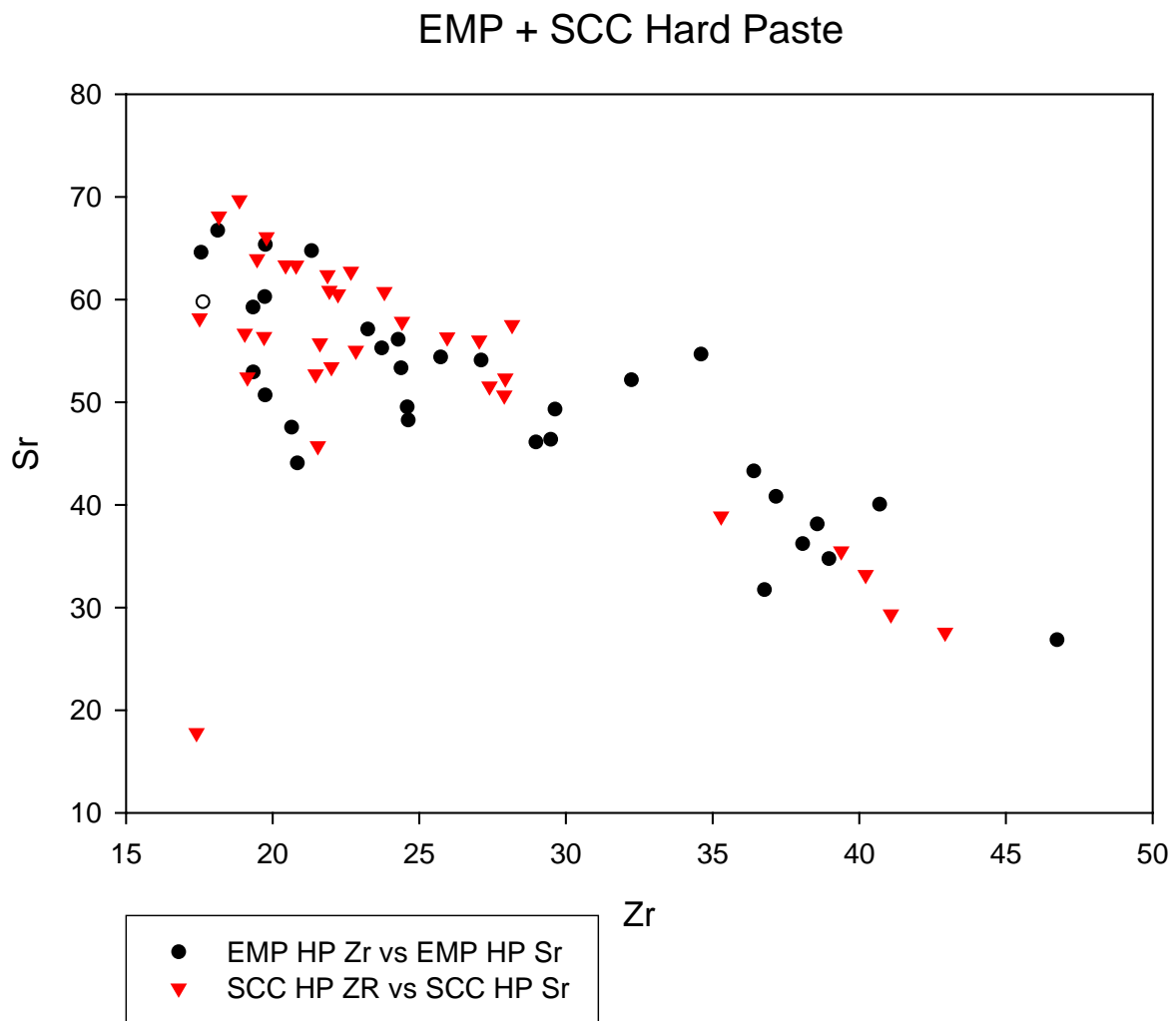
The EMP and SCC soft paste comparison forms a tighter cluster than the hard paste combinations. Both have thirteen outliers and, in addition, one has to take into account the fact that the Y axis only starts at the 20-percentile point. The Y axis is the reason for the spread of the main cluster. Regarding time and production centre, it seems almost certain that the soft paste Greenwares are all very similar in either time and/or provenance.



EMP hard paste in black and SCC hard paste in red both show clustering but, interestingly, the clusters are interwoven with each other instead of, as one would expect, overlapping each other.

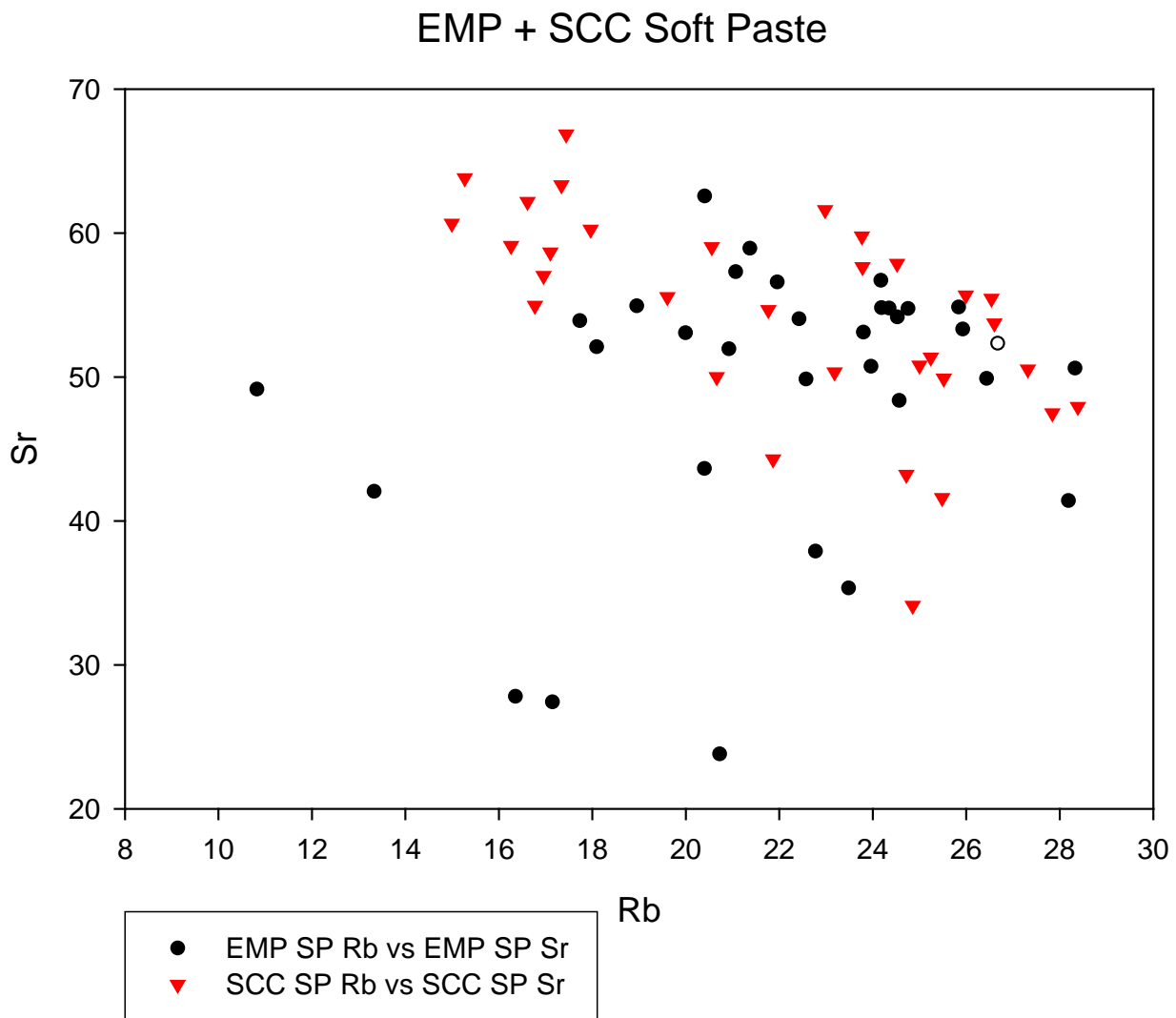
Would the suggestion offered earlier page 76 about the SCC objects having a different provenance and/or time be valid?

It is clear more research needs to be conducted in order to understand the significance of the fact that the EMP hard paste and SCC hard paste test result are parallel instead of overlapping.



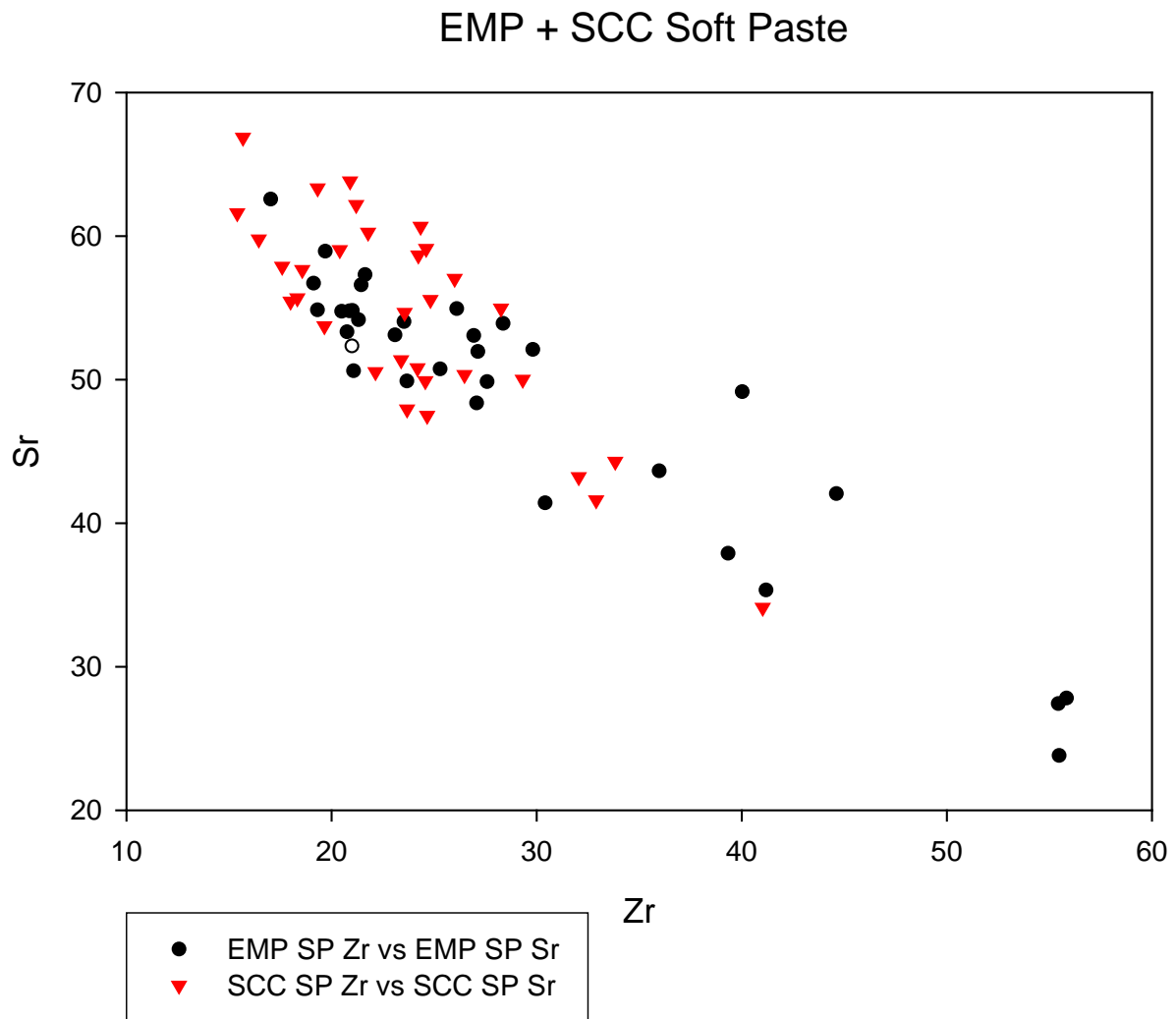
The same hard paste from both excavation sites with Zr-Sr combination does not seem to produce a similar closely related graph. The slight spreading, horizontally, takes little away from the clustering, even though the X axis starts only at 15 and finishes at 50. Again, SCC, in red, seems to cluster more tightly, suggesting quality and maybe even temporal and provenance similarities.

The fact that EMP was a likely area for loading and off-loading cargo from ships may explain the wider chemical fingerprint spread. It is likely that objects for different areas in Singapura arrived here before dissemination either to other parts of the city, or were re-exported to other towns in the region.



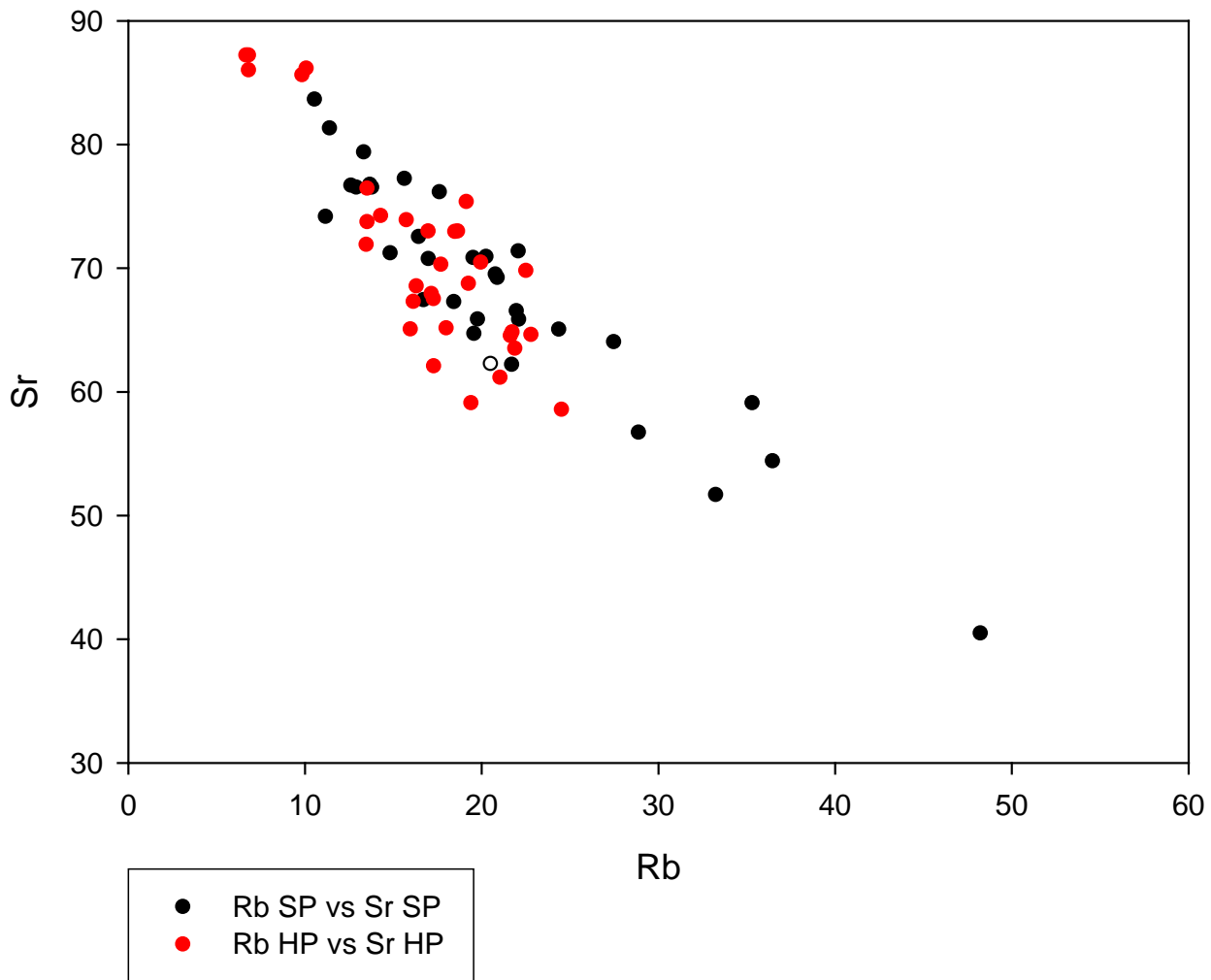
Taking into account that fact that the X- and Y-axes are shorter than in other graphs, EMP shows a very close cluster, which suggests that some of the pieces brought to Singapura from the same time, production and quality were discarded upon unloading. Rb seems to be a constant in the analysis of this graph; most of the 33 results included are within a 10-percentile point area of the Rb-axes.

Both combinations of elements Rb-Sr and Zr-Sr show, as expected, chemical fingerprints which fall within the 20% area of 45-65 on the Sr axis. This is likely to suggest a similar time and a similar production centre, but not necessarily the same kiln site, as there are slight clusters within the major clusters.



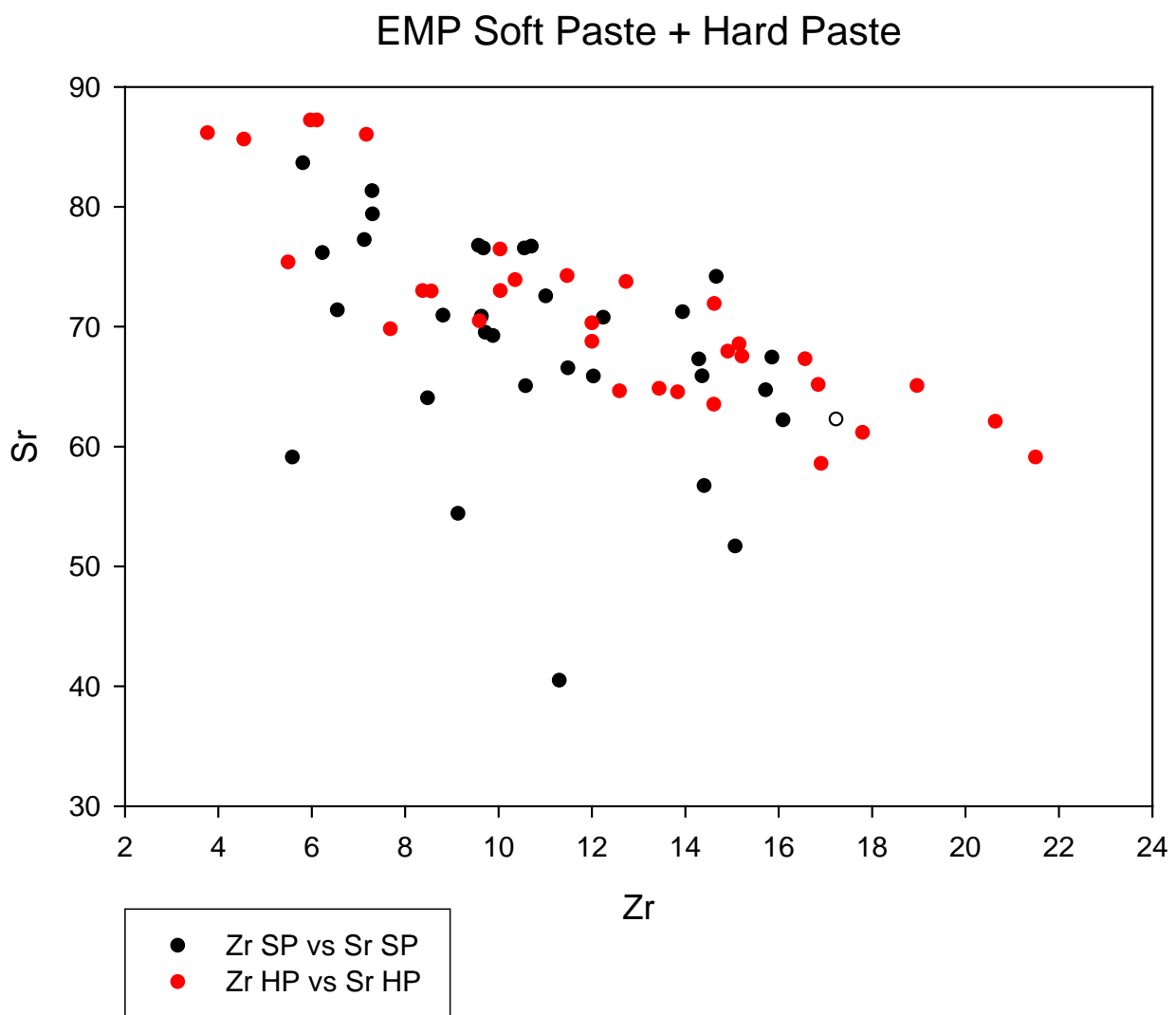
Again, the X- and Y axes are shorter, yet the main cluster is between the 15-30 percentile point on the Zr axis and 45-65 percentile point on the Sr axis. However, the cluster is slightly spread wider horizontally; again, with the EMP results there is, as expected, a close-knit cluster. The SCC results are more spread out, suggesting a pattern of purchase of availability, not one that suggests ordering in bulk. Although, the number of sherds tested from this site are too few to determine if this site was a wholesale site for greenwares in fourteenth century Singapura or if it was a place where someone 'collected' a certain quality of ware.

EMP Soft Paste + Hard Paste



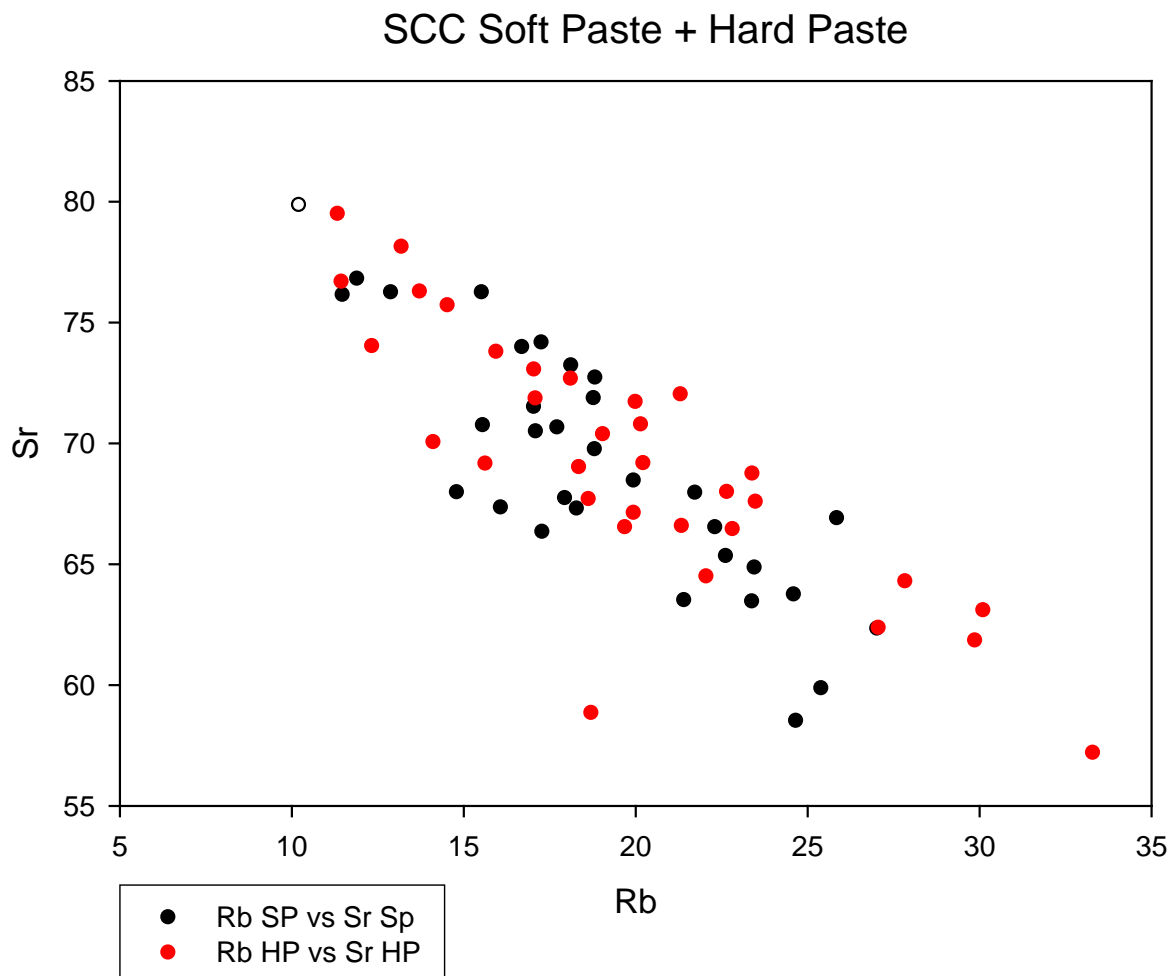
EMP hard paste, again, shows a graph with an anomaly as the Sr axis presents results up to the 88th percentile. In combination with the SCC, hard paste results, the graph shows results just beyond the 70th percentile point. The EMP and SCC hard paste graph (page 94) starts on the Sr axis at the 10th percentile point; here it starts at the 30th percentile point.

Ignoring these anomalies, this graph suggests that it is likely that soft paste and hard pastes were produced at similar times and in similar production centres.

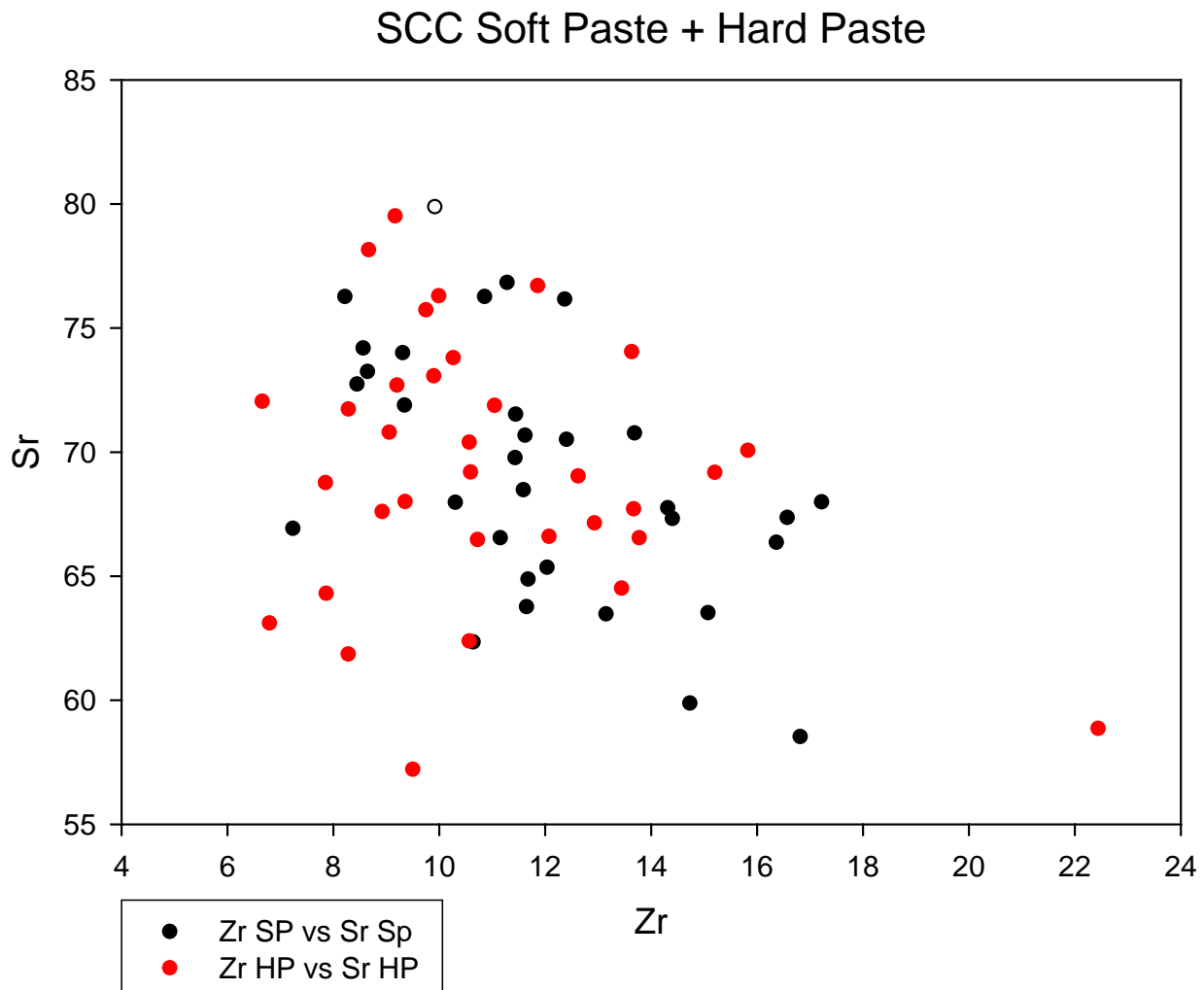


Again, I ignore the anomalous Sr axis, as there is no explanation I can offer for it.

There are six soft paste outliers and a similar number of hard paste outliers. The close relation between the soft paste and the hard paste results suggest, again, a similar time and place of production.



SCC soft paste and hard paste comparison, for the first time, also produce an anomalous graph ranging up to 85 percentile points on the Sr axis. Here it seems that soft paste and hard paste are closely related in possible time and provenance, as they cluster within a 20-percentile point range on the Rb- and Sr- axes.



Again, this graph displays an anomalous Sr axis, which for the sake of this research I will ignore.

The Zr axis is short which clusters the majority of chemical fingerprints within 14% of the Zr axis. This suggests that the SCC soft paste and hard paste are temporal and provenance equals.

An interpretation of the last four graphs could possibly be that the soft paste and hard paste objects are produced and maybe even, fired in and around the same time, which was expected as the objects have all been excavated from fourteenth century layers. It is possible that the soft paste objects were placed further away from the fire in the kiln than the hard paste objects. It is a well-known fact that the type of kiln used to fire the Chinese

ceramics is called 'Dragon kilns'. In addition, it is common knowledge that these kilns fired porcelain, stoneware, and earthenware in this order from closest to the fire to furthest away from the fire. The celadon, or Greenware, objects would be fired in between the porcelain and the stoneware objects. The objects closest to the stoneware are likely not to get enough heat from the fire for the Greenware clay to vitrify completely unlike the same objects closer to the fire, placed just behind the porcelain objects in the kiln. This is a possible explanation for the similarities in soft paste and hard paste chemical fingerprints but not the difference in their clay hardness.

More research on the degradation of trace elements in clay source is needed in order to understand the significance of these graphs from a time line point of view.

13. Subjective vs. Objective

After analysis and comparison of the Greenware objects by chemical fingerprinting it would be interesting to compare these objective scientific results with the subjective stylistic analysis of scholars and avid ceramic connoisseurs. However, it is clear that the stylistic analysis is by far the fastest and financially least onerous method, though it can produce contradictory results from one person to the next. The experts spoken to will be indicated as A, B, C and D. The aim of this section is to try to determine if the stylistic data is corroborated by the chemical data.

13.1. Stylistic analysis

Stylistic analysis is conducted by comparing the attributes of each individual artefact with the knowledge set of the observer. Each researcher of ceramic objects has developed a personal system of recognising artefacts to determine the provenance of the object. Herein lies the expected diversion among them. All will agree that the objects shown to them regarding this research are known as Greenware or Celadon. The determination of provenance is for all researchers different as their knowledge set developed separately.

13.1.1. *Researcher A*

Researcher A was asked to identify 131 artefacts without setting any parameters or guidelines;

Chinese, Zhejiang, Longquan:	47 sherds
Chinese, Fujian:	30 sherds
Chinese, Guangdong:	10 sherds
Unknown:	44 sherds

There are likely to be unknowns in any data set as some of the objects have little stylistic evidence to identify them by. This researcher has possibly identified the artefacts not only to provinces but even to production sites. One of the artefacts had been broken without knowledge of this researcher or that of researcher A. Only after analysis of the data, it became known that this break had occurred but as more interesting detail the two halves of the broken sherd were sorted in different groupings, which confirms that stylistic analysis is subjective. The broken piece was likely too small to be identified and was placed in the grouping of unknowns. I do want to continue with this comparison as I believe that if at least three out of four of the researchers identify sherds to be from a specific province or better yet from the same kiln site, there must be some objective criteria for their classificatory method.

13.1.2. *Researcher B*

Researcher B was asked to identify 129 artefacts without setting any parameters or guidelines;

Chinese, Zhejiang, Longquan:	86 sherds
Chinese, Fujian:	28 sherds
Chinese, Fujian, Putian:	2 sherds
Vietnamese:	1 sherd
Unknown:	12 sherds

One sherd was not available for this researcher as it was being tested at that point in time. Researcher B seems to have categorised the sherds in provinces and a country. It is

becoming clear that there are distinct differences between the knowledge set of researcher A and B.

13.1.3. Researcher C

Researcher C was asked to identify 130 artefacts without setting any parameters or guidelines;

Chinese, Zejiang, Longquan Kiln: 121 sherds

Fujian: 9 sherds

This researcher also added that 99% of the sherds were from the Yuan Dynasty (1280-1368). He seemed to judge the sherds by their quality and ignored most of the other attributes the sherds showed, if any. The Longquan sherds were labelled as good quality by this researcher, ignoring the obvious difference in soft paste and hard paste.

13.1.4. Researcher D

Researcher D was asked to identify 100 artefacts without setting any parameters or guidelines;

Chinese, Guangdong: 26 sherds

Chinese, Fujian: 22 sherds

Chinese, Zhejiang, Longquan: 10 sherds

Unknown: 42 sherds

30 sherds were not available as they were being tested at the moment researcher D identified the sherds. Similarities between A, B and D are becoming clear.

14. Analysis

The Zhejiang, Longquan comparison provides us with an opportunity to conduct further research on the Singapore excavated artefacts.

Even though all the researchers identified more than 90% of the sherds as Chinese, most prefer not to go beyond identifying the province of suspected production. It is now interesting to look at the named regions within China to see how the four researchers collectively compare.

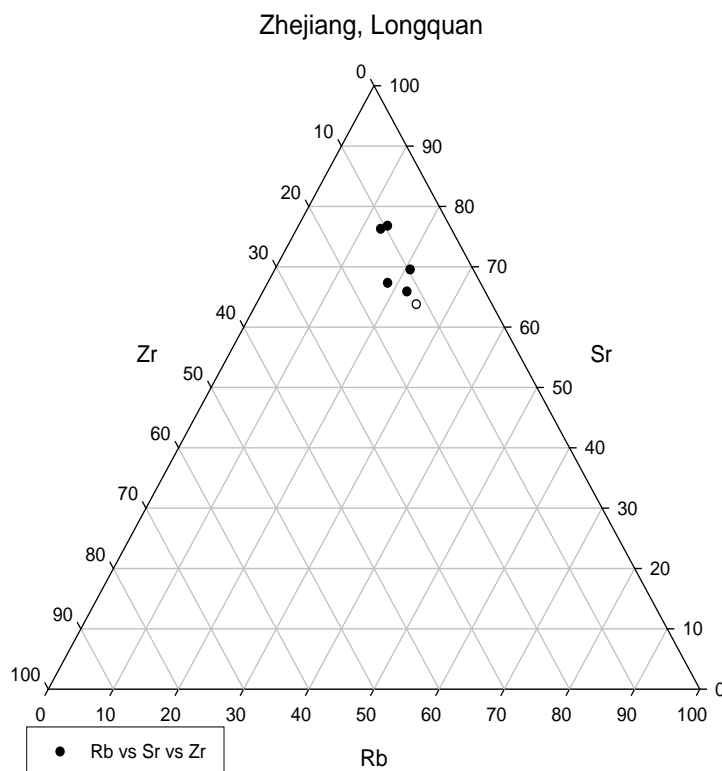


Figure 15: South-eastern Provinces of China

A		C	
Zhejiang, Longquan:	47 sherds	Zejiang, Longquan Kiln:	121 sherds
Fujian:	30 sherds	Fujian:	9 sherds
Guangdong:	10 sherds		
Unknown:	44 sherds		
B		D	
Zhejiang, Longquan:	86 sherds	Zhejiang, Longquan:	10 sherds
Fujian:	28 sherds	Fujian:	22 sherds
Fujian, Putian:	2 sherds	Guangdong:	26 sherds
Vietnamese:	1 sherd	Unknown:	42sherds
Unknown:	12 sherds		

Three of the four researchers identified three provinces along the south-eastern coast of China for the origins of the Greenware objects, namely Zhejiang, Fujian and Guangdong. All four were specific to identify a kiln site within Zhejiang province, namely Longquan.

Beside researcher C, researchers A, B and D all identified over 20 sherds as being from Fujian province. A and D identified around 25 sherds from Guangdong province. Besides D the other researchers saw the majority of the sherds coming from Zhejiang province.



The four researchers have all identified the kiln site in Zhejiang province, Longquan as a source of provenance for the Greenwares found in Singapore. When comparing the Longquan grouping from each researcher with each other, there are large differences; C identifies only 10 sherds from this kiln site where D finds that 121 sherds come from this location. Comparing the individual object research numbers from each researcher's

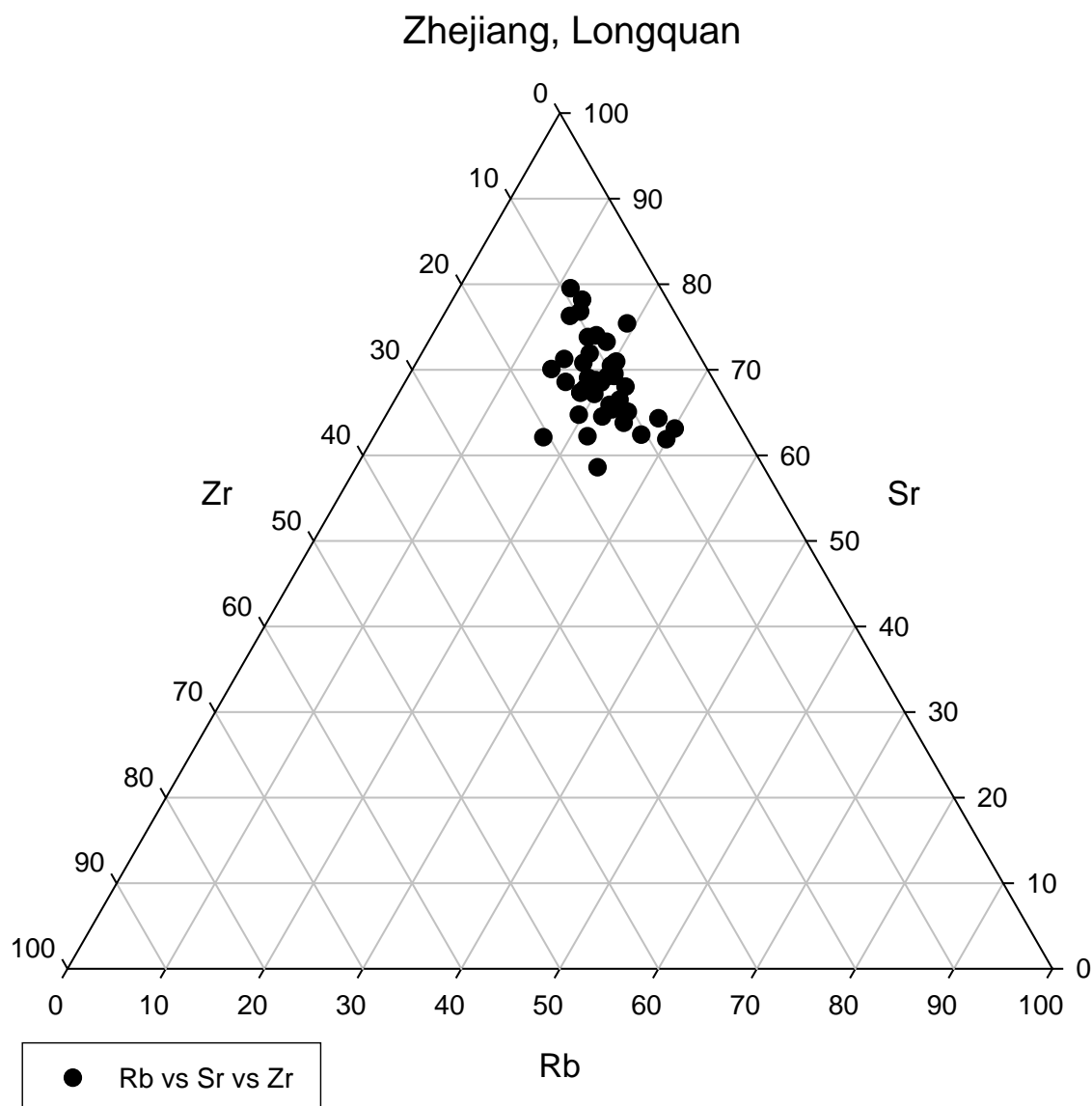
Longquan group, there are only six sherds that they all confirmed to be from there. Comparing the six chemical fingerprints with each other provides a good insight on the possibilities of scientific study of chemical fingerprinting provenance research of Greenwares.

The six chemical fingerprints of the six sherds shown in the graph above show a close proximity with each other. There are two clusters within these six artefacts compared. Two sherds cluster closely together, and are therefore likely to be from the same kiln site. The four other sherds could be from different kiln site or production centre. This result should be expected as four researchers agreed on their provenance.

The clear separation of the two clusters is an indication that the subjective analysis is likely to omit aspects not visible to the naked eye. The likelihood is that these two clusters indicates two production centres. This contradicts the four researchers' analysis to a certain extent. One of the clusters is likely the Longquan production centre whereas the other is likely to have produced similar artefacts of a similar quality. Considering that researcher D

did not have the full collection available it was deemed important to see if A, B and C would identify more sherds in the same chemical grouping.

This comparison created a group of forty-one sherds jointly grouped within the Longquan kiln site according to them. Comparing these forty-one sherds with their chemical fingerprint they cluster close together in the ternary graph below, with a few outliers.



These outliers could be a confirmation of production deviations or products from a different kiln site with styles physically similar to the Longquan production.

The small cluster at the top of the general cluster is likely to originate from a different production centre. It is possible that this production centre was located within the province

of Zhejiang but had a different clay source, however, they did seem to adhere to a strict Greenware recipe. When examining the main cluster closely there are smaller clusters visible. These clusters can represent two things;

1. The small clusters represent different kiln sites within the large production centre, where slight differences in clay and glaze preparation could occur.
2. Or, the small clusters represent different batches of Greenware produced at the same kiln site., where every batch could have a slightly different preparation, which could indicate the outliers as 'bad-day products'.

Only chemical fingerprints retrieved from artefacts excavated at kiln sites in the Zhejiang, Longquan area could provide a more conclusive answer.

15. Conclusion

The combination of scientific archaeological data and scientific chemical data opens the door to greater understanding of the excavated Singapore Greenware assemblage. The chemical fingerprinting of the artefacts within this research do show clear clustering that is currently open to a vast array of conclusions, of which most will be premature.

The assemblage tested from the EMP site clearly suggests a location where a vast array of objects has been discarded. The chemical fingerprints show several groupings within the general clustering of the overall Rb-Sr-Zr graph. This confirms some of the conclusions formulated about this site by others (Miksic & Low 2004).

The SCC comparison shows clear clustering within the hard paste setting. At present, there is no more information available about the SCC site besides the information mentioned and discovered within this research. The tight hard paste clustering suggests that the SCC site was the place or near a place where specific hard paste Greenware objects were collected. Whether this was for sale or for collection/utilitarian use that will remain unknown as the chemical data does not provide any interpretations of the anthropological kind. The probability that the hard and soft paste wares both came from the same kilns is particularly

significant, because it resolves a major question, which is bedevilled ceramic analysts for decades.

The objective and subjective comparative analysis implies that the researchers and connoisseurs can accurately identify artefact to their provenance. In this research, they identified three provinces and some even ceramic production centres. Their knowledge is invaluable to archaeological analysis and will likely always be the basis of any scientific comparative research. The finer detail of kiln sites within a production centre is likely only to be identified by scientific research.

The main conclusion of this research must be shrouded in questions and options for future research. However, this research will likely not be able to give many more answers toward a better understanding of the excavation sites themselves or their context within the large site of 14th-century Singapura. This research does provide an insight into the possibilities of using EDXRF for chemical fingerprint analysis. These chemical fingerprints do show significant clustering, which likely can be further utilised in the provenance research of Greenware. The identification of the individual clusters will need to be done by testing artefacts found in the known Greenware production centres and preferably with artefacts from individual kiln sites.

With regard to the conclusions of this thesis on 14th-century Singapore and its suppliers of Greenware; it seems almost certain that Pancur, Singapura, Temasek, Ta-Man-Shi or whichever name one prefers to use for 14th-century Singapore, had several providers. I would say with a high degree of certainty that traders from at least three Chinese provinces exported their Greenware to Singapore. The likelihood that the Greenware from Zhejiang, Fujian and Guangdong were first 'collected' in one location before they were shipped or 'trans-shipped' to Southeast Asia seems rather small as this would entail loading and unloading of ships, which in turn mean a high chance of breakage of the wares. Land transportation seems nullified as well as the chances of breakage due to this type of

transport are high. The known land-based silk route was virtually abandoned when maritime shipping became a viable option.

The small cluster below the major cluster could represent another ceramic provider, but there is not enough data to state this with any degree of certainty. To be able to draw more definite conclusions concerning the uncovered data in this research it is necessary to test Greenware artefacts from kiln sites from the three Chinese provinces mentioned.

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