Research Article

Geochemistry of the *Soll* Facies of the Lower Globigerina Limestone Formation, Malta.

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Summary. *Rock from the "soll" seams of the Lower Globigerina Limestone Formation is distinguishable from "franka" because of its characteristic honeycomb weathering. However, when freshly quarried, "soll" limestone, an inferior building material, is not so readily recognizable. This study was aimed at characterizing "soll" on the basis of its inorganic geochemistry. It is shown that "soll" has a significantly higher silicon content than normal "franka". The silica content of "soll", which is a measure of its clay content, is typically less than 5%. Therefore, the traditional belief that "soll" is a limestone which is rich in clay is untenable. The silicon content may be useful in differentiating "soll" limestone from "franka". Besides that in silicon, other elemental anomalies are found associated with the "soll" facies of the Lower Globigerina and also with "soll ahdar", a stratigraphically related grey-green variety.*

Keywords: Globigerina, limestone, geochemistry, silica, weathering, Malta

The sedimentary rock outcrops of the Maltese Islands are largely composed of limestones and marls deposited in layer cake succession during the Tertiary. The stratigraphic pile is extensively faulted. Listed in order of deposition, the geologic formations are: Lower Coralline Limestone, Globigerina Limestone, Blue Clay, Greensand and Upper Coralline Limestone (Spratt, 1843; Murray, 1890). The Globigerina Limestone Formation is exposed over large areas of central and southern Malta and of Gozo and shows marked variations in thickness, ranging from about 20m to 210m. The formation consists of yellow to pale grey, fine-grained biomicritic limestones, composed almost wholly of the calcitic tests of globigerinid planktonic foraminiferans.

Lithologically and palaeontologically, the formation is divided into Lower, Middle and Upper Members, with a conspicuous phosphatic conglomerate (or "nodule") bed marking the base of each member (Rizzo, 1932; Pedley et al, 1978; Zammit Maempel, 1977). The Upper Member of the Globigerina Limestone displays three major layers: an upper and lower layer consisting of brown-weathering freestone, separated by a seam of blue-grey marls. The stone from the Upper Globigerina weathers very badly and is not considered suitable for masonry work. The Middle Member (Maltese name: *il-qarghija; il-bajjad)* consists mainly of white to pale grey marly limestones with numerous nodules. The lower section of this member contains local layers of chert. Generally, this stone is not useful for building purposes. The Lower Member (Maltese name: *franka*) is composed of massive bedded, pale yellow limestone which, generally, weathers well on exposure. Marine macrofossils may be locally abundant and well preserved, usually as brown-coloured internal moulds. This division of the Globigerina Limestone provides most of the stone employed in the building industry in Malta and represents an important and unique mineral resource. The size of this resource is practically unknown.

The building quality of stone from a given quarry is generally dependent on the stratigraphic level from which the stone is derived. Typically, going from top to bottom, a layer of fractured stone (Maltese: *qxur*) is encountered just beneath the topsoil. The first seam of good quality building stone, or *franka*, which may be several metres thick, generally underlies this fractured rock. Eventually, a layer of limestone, known in Maltese as *gebla tas-soll*, is found. This layer can vary in thickness from about 0.3m to 1.5m or more, depending on the locality. Other discrete *soll* seams may be encountered further down the quarry face. Eventually, the *franka* is permanently replaced by badly-weathering limestone. At this stage, the quarry is usually abandoned. *Soll* limestone is not appropriate for building purposes, since it weathers badly on exposure. A study of the mechanical and physical properties of Globigerina Limestone (Cachia, 1985) has shown that *soll* limestone is less porous and more dense than *franka*. Also, in the dry state, *soll* limestone is mechanically stronger although it becomes much weaker when wet. *Soll* limestone is considered suitable for use only in foundation work.

Soll outcrops exposed at old quarry faces are fairly easy to recognize by their cavitational weathering which produces conspicuous and characteristic honeycomb erosional features. These outcrops frequently occur as horizontal beds with fairly well-defined boundaries with the normal *franka*. A survey of the geological literature reveals that the *soll* seams of the Lower Globigerina have typically been recognized from their peculiar weathering style. Thus Pedley et al (1978), refer to the presence of characteristic honeycomb weathering in Lower Globigerina and they ascribe them to syndepositional trace fossils. It is notable that the descriptive term *soll*, unlike the term *franka*, has not been employed in the literature in order to characterize this limestone facies. "*Soll*" seems to be a term strictly used by masons and quarrymen, and presumably refers to any Malta limestone which weathers by cavitation and flaking. Thus, it probably would also be used by the trade to describe limestone derived from the Middle Globigerina which also weathers badly and which Rizzo (1932) described as being only useful for "foundations and inside thick masonry". In his work, Rizzo never uses the descriptor *soll* either for the facies as it occurs in the Lower Globigerina or for any other rock unit of similar physical properties belonging to the Globigerina Formation.

The inferior qualities of *soll* limestone as a construction material have been traditionally ascribed to a high clay content in this rock. However, no published data exists which supports this belief. Rather, the lithological similarity between *soll* and *franka* limestone, as perceived by visual examination, has probably served to prevent the recognition of the *soll* as a distinct facies in the Lower Globigerina.

The present study was aimed at determining whether *soll* limestone is significantly different from normal *franka* on the basis of its inorganic geochemistry. Apart from the obvious geological interest in establishing such a difference, there is another, possibly more practical, objective which the study addressed. As yet, there exists no conclusive test, either chemical or physical, which can be used to identify whether a sample of globigerina limestone is of the *soll* or normal *franka* type. Freshly-cut stone slabs, small hand specimens or drill cuttings of *soll* limestone are difficult to distinguish from *franka* by visual inspection, even though experienced, local quarrymen claim to be able to distinguish between stone slabs by drenching them with water and observing the sheen produced at the wet stone surface.

Being able to distinguish objectively between *soll* and *franka* is clearly important in technical and legal cases of litigation over the nature of a given construction material. It is also very important to be able to distinguish between these two types of stone when the samples are in the form of cuttings from exploratory drilling aimed at assessing the size of the mineral reserve of Globigerina limestone.

In the first phase of this preliminary study, samples of Lower Globigerina limestone representing both *soll* and *franka* were taken from a site in a quarry which possessed clearly visible *soll* horizons. The rocks were quantitatively analyzed for the six elements calcium, magnesium, carbonate carbon, chlorine, phosphorus and silicon. In the second phase of the study, another set of samples representing both limestone types were analyzed for aluminium and iron, in addition to silicon. A number of these samples were taken from the same quarry as before but other limestones were also analyzed which, although deriving from the Lower Globigerina, included a type of "*soll*" that was different in having a unique depositional style and a grey-green colour.

Materials and Methods

Sampling Sites

The state-owned Tar-Robba quarry at Mqabba was selected for sampling. Figure 1 shows the site plan of the quarry. The quarry is still active, rock cutting taking place in the north eastern sector. Samples were taken from the western and north eastern faces of the quarry. Rock from the western face was last quarried several decades ago and the rock face here is clearly indented with marks made by tools which were in use at the time. In this area of the quarry, two exposed *soll* seams are easily distinguished by their typical honeycomb weathering. The quarrying in the north eastern sector revealed that the lower *soll* seam, of which only about 50 cm were exposed at the western face of the quarry, had already reached a thickness of 30m and presumably continued further down to an unknown extent.

During the first phase of the study, ten samples were collected from a column of rock located in the western quarry face. The column was about 500cm high and included the two *soll* zones. These *soll* zones were separated by a vertical distance of about 260cm. Five samples were taken from the *soll* seams and another five were taken from the *franka*, as shown in Table 1. For the second phase of the study, seventeen limestone samples were analyzed (Table 2). Seven of these were obtained from the same site as employed in the first phase, three from the upper *soll* seam, and four from the contiguous *franka*. A further six samples were collected from the north eastern face of Tar-Robba quarry, three from the lower *soll* zone and three others from the *franka* above.

Sample Descriptor	Height above footpath (cm)	Rock Type
W1S	25	*soll*
W2S	50	*soll*
W3	100	*franka*
W4	150	*franka*
W5	200	*franka*
W6	250	*franka*
W7S	300	*soll*
W8S	350	*soll*
W9S	400	*soll*
W10	450	*franka*

Table 1. Samples taken during the first phase of the study.

The authors based the classification of these latter six samples on advice from quarry personnel because at this particular quarry face, the *soll* seams, being freshly cut, were not yet marked by weathering. The other four limestone samples were obtained from a road cut in Msida next to the Birkirkara Road bridge. The limestone exposed in this area is considered as completely unsuitable for building purposes. At this site, two sedimentary facies could be distinguished: massively

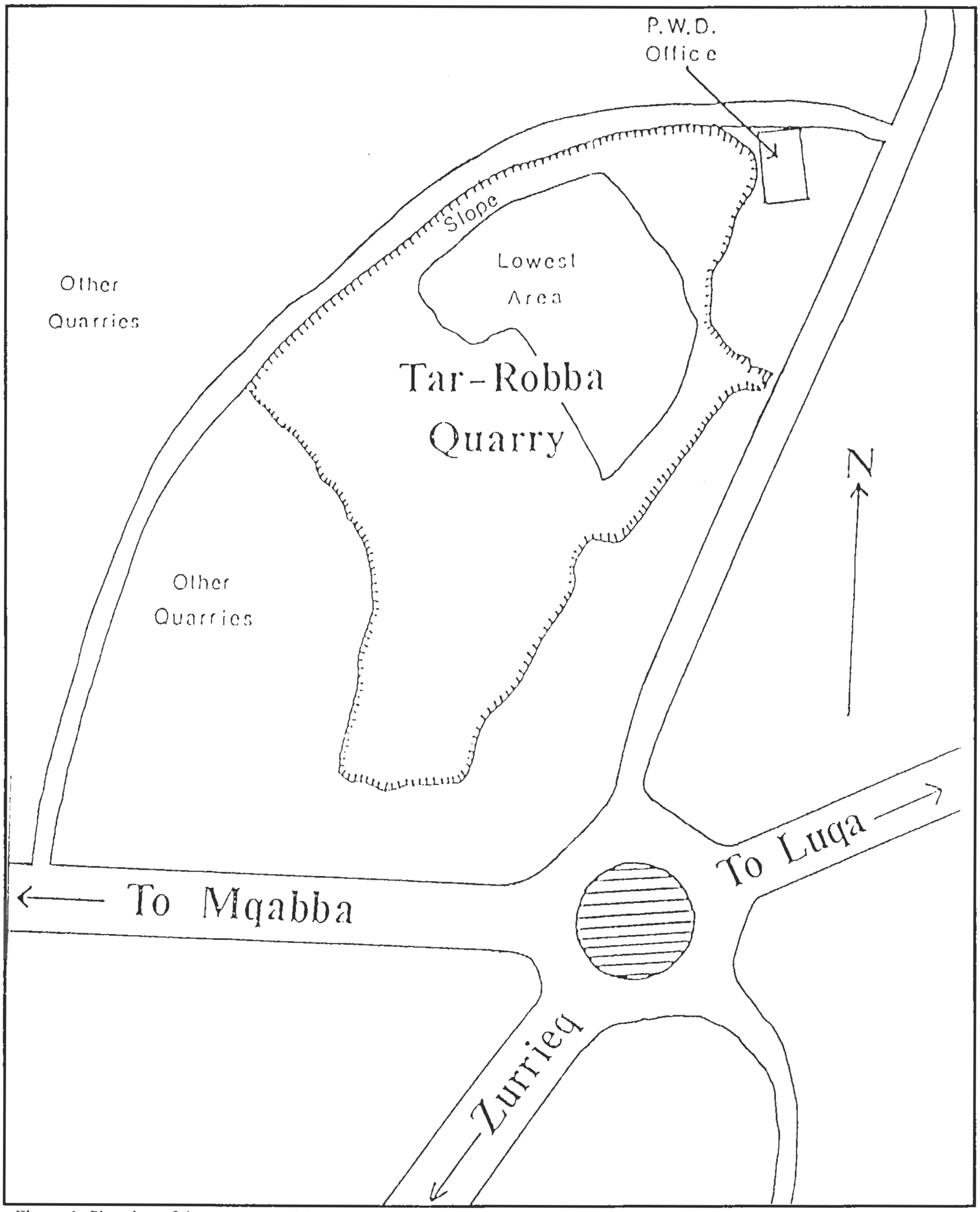

Figure 1. Site plan of the state owned Tar-Robba quarry at Mqabba.

bedded, yellow to white, limestone similar to the fresh *soll* exposed at Tar-Robba and a greyish-green facies occurring as discrete lenticular deposits, about 1m wide on their long axis by 0.5m. The dark coloured lenses were embedded in the yellow-white limestone as local occurrences. Two rock samples were analyzed from each of these two facies. Quarrymen do not probably regard this grey-green limestone as *soll* since it is so obviously different in colour from stone that has been traditionally called so. We propose to refer to this particular facies of the Lower Globigerina Limestone as *soll ahdar* (green *soll*) in view of its colour and its stratigraphic position within the *soll* zone.

Sampling Protocol

Sampling was performed by removing a layer of rock, about 2cm thick and about 40cm^2 in area, from the surface of the outcrop. This material was discarded and about 100g of rock cuttings were collected from the exposed area. This procedure was adopted in order to avoid sampling rock surfaces which may have been affected by environmental factors. The rock cuttings were placed in labelled polythene bags, pending analysis.

Analytical Techniques

Deionized water was used as solvent and analytical grade reagents were employed without further purification. A Varian Model AA1275 atomic absorption spectrophotometer was employed for calcium and magnesium determination. Double beam spectrophotometry was performed with a Shimadzu/Bausch and Lomb Spectronic 210UV model.

Sample	Height above footpath	Provenance
		Mqabba
W11	100 cm	Western face
W12	200 cm	Western face
W13	250 cm	Western face
W14S	300 cm	Western face
W15S	350 cm	Western face
W16S	400 cm	Western face
W17	450 cm	Western face
	Relative height	
E1S	0 m	Eastern Face
E2S	1 m	Eastern Face
E3S	15 m	Eastern Face
E4	30 m	Eastern Face
E5	32 m	Eastern Face
E6S	35 m	Eastern Face
		Msida
M1SA	0 cm	grey green facies
M2SA	75 cm	grey green facies
M3S	125 cm	yellow facies, proximate to green facies
M4S	200 cm	yellow facies

Table 2. Samples taken during the second phase of the study.

Single beam spectrophotometry was performed using a Milton Roy Spectronic 20. Calcium and magnesium were determined by a method based on atomic absorption spectrophotometry. Phosphorus was determined by the molybdenum blue photometric method using ascorbic acid as reducing agent (Jeffery and Hutchinson, 1981). The carbon dioxide content was found by determining the loss in weight on treating the rock with perchloric acid (Cumming and Kay, 1956). Chlorine was determined gravimetrically.

Two methods were employed for the determination of silicon. In the first phase of the study, a technique based on the absorbance of the silicomolybdate chromophore was employed (Jeffery and Hutchinson, 1981). In the second phase, silicon was determined gravimetrically by volatilization as the tetrafluoride. Total iron and aluminium were determined spectrophotometrically after suitable oxidation and complexation with 8-hydroxy-7-iodo-5-quinolinesulphonic acid (ferron) (Cumming and Kay, 1956).

Results and Discussion

The results obtained in the first phase of this investigation are collected in Figure 2. The data is presented in terms of the mass percentage of the element in the oxide form, except for chlorine.

These results show that both *franka* and *soll* Lower Globigerina limestones consist primarily of calcium carbonate. Statistical correlation tests (*e.g.* the Spearman rho) performed on the carbonate and calcium values show that these values correlate well at the 95% confidence limit. This abundance of calcium carbonate in Lower Globigerina limestone is to be expected from a rock which is, petrologically, a foraminiferal biomicrite. Moreover, previous analyses of Lower Globigerina limestones for calcium carbonate concur with our results (e.g. Lehmann, 1980).

In order to assess whether the individual elemental contents obtained for the limestone in the *soll* horizons were significantly different from corresponding values for *franka*, the statistical, non-parametric, Mann Whitney U test was applied to the values obtained. The results of such an assessment show that at the 95% confidence limits, *soll* limestone has a statistically significant higher content of silicon (Figure 2). The calcium, magnesium, carbon, chlorine and phosphorus content of both types of limestone are not significantly different (Figures 2 and 4)

Furthermore, it can be shown that the mean mass ratio of magnesium to calcium for *soll* and *franka* are, respectively, 0.089 and 0.113 and that such ratios for the different limestone types are significantly different at the 95% confidence limit.

Silicon, in sedimentary rocks, may be present as the dioxide in one or more of its various geologic forms, *e.g.* silica, quartz, chert etc. The element may also occur in aluminosilicates, which in sedimentary materials, are frequently, although not uniquely, represented by the clay minerals. Both these two types of silicon-containing minerals have been reported in Maltese globigerina limestones (e.g. Murray, 1890; Formaggio, 1972). However, there are no reports on the relative abundance of such silicates in the *soll* and *franka* facies of the Lower Globigerina Formation.

The foregoing results suggested further investigations into the possibility that the silicon anomaly of the *soll* facies would correlate with other anomalies in the limestone. It was argued that if the higher silicon content in *soll* was primarily due to the presence of clay aluminosilicates, then the silicon anomaly would correlate with an aluminium anomaly, and possibly also with one in iron. This led to the second phase of the study. The results obtained are shown in Figure 3. From the silicon mass fractions, it is again obvious that higher silicon contents are associated with the *soll* limestones.

This conclusion is confirmed when the experimental results are statistically analysed. The mean percentage silica of *soll* found in Tar-Robba is 2.94 %. Comparision of this value with that obtained previously (4.38 %) suggests that the gravimetric technique tends to underrate the silica content. In fact, the individual *soll* and the *franka* values displayed in Figure 3 are consistently lower than the corresponding values shown in Figure 2. This is an important consideration to be borne in mind.

The limestone samples from the Msida site are even richer in silicon than those from Tar-Robba. The values

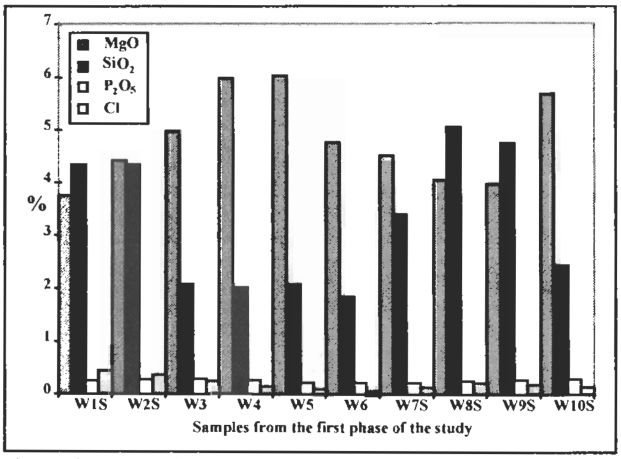

Figure 2. Magnesium, silicon, phosphorus and chloride level in samples from the first phase of the study.

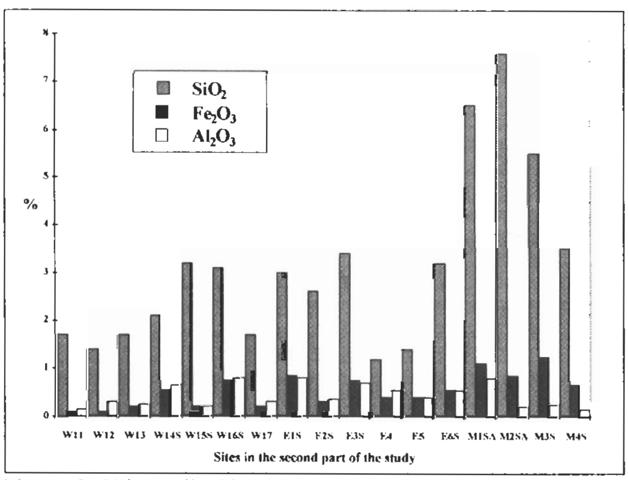

Figure 3 Silicon dioxide, iron(III) oxide and aluminium oxide content in samples from sites of the second phase of the study.

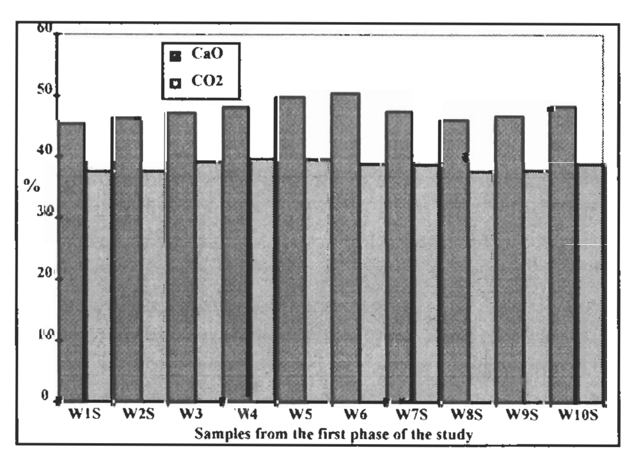

Figure 4. Calcium and carbonate content in samples from the first phase of the study.

show that the grey-green *soll* facies is richest in silica while the Msida yellow *soll* has a similar content to that of Tar-Robba. It is notable that sample M3S, which derives from a region which is stratigraphically intermediate between green *soll* and the usual yellow variety has an intermediate silica content.

Comparision of the aluminium content of the various types of limestones does not reveal any clear-cut anomaly associated with *soll* (Figure 3). Thus, for example, except for M1SA, all the other Msida *soll* samples, while exhibiting pronounced silicon levels, have aluminium contents typical of normal Tar-Robba *franka*. When the aluminium content of all samples are statistically analyzed by the Mann-Whitney U test, no anomaly associated with *soll* emerges, although if the analysis is performed on the Tar-Robba subset, the *soll* seems to have significantly more aluminium. However, even for these limestones, the aluminium oxide content is not statistically correlated to the silicon dioxide content on the basis of the Spearman rho and the Kendall tau tests.

These observations suggest that the main silicon minerals which are responsible for the higher content of the element in Tar-Robba and Msida *soll* facies are probably not aluminosilicates but silicon dioxide phases, possibly including amorphous silica and cryptocrystalline quartz.

The total iron content (Figure 3) of the global set of samples is statistically higher for the *soll* limestones according to the Mann Whitney test. The same conclusion holds true if the Tar-Robba samples are considered on their own. Also, the iron(III) oxide and the silicon dioxide anomalies are statistically correlated with 95% confidence when the samples are considered globally but with just under 90% confidence when the Tar-Robba limestones are considered on their own. Moreover, for the Tar-Robba limestones, the aluminium oxide and the iron(III) oxide contents are also correlated. In the Msida samples, there are no correlations between any pair of elements.

If the contents of silicon dioxide, aluminium oxide and iron(III) oxide are plotted on a three-dimensional scatter plot (Figure 5), we find that the *franka* samples define a compact group in a distinct region of the plot. The *soll ahdar* samples plot away from this *franka* pole while the yellow *soll* samples form a rather diffuse group in the intermediate region of the scattergram, closer to the *franka* than the *soll ahdar* region. From Figure 5, one concludes that sample M3S, stratigraphically proximate to but outside the green limestone lens in the Msida outcrop is closer in lithology to *soll ahdar* than to the yellow variety, in spite of its colour.

Conclusions

We conclude that the differentiation of the *soll* facies from the *franka* of the Lower Globigerina Limestone Formation, as represented at Tar-Robba outcrop, does have an inorganic geochemical basis. *Soll* limestone has a notably higher content of silicon and a less marked but significantly higher content of total iron and aluminium than *franka*.

The higher silicon content of *soll* is probably largely due to the presence of silica cement and cryptocrystalline quartz in these rocks although the correlated iron and silicon anomalies could suggest a more abundant presence of glauconite-derived limonite or other authigenic ferruginous minerals. Only a detailed petrographic analysis of this limestone facies could help decide this point.

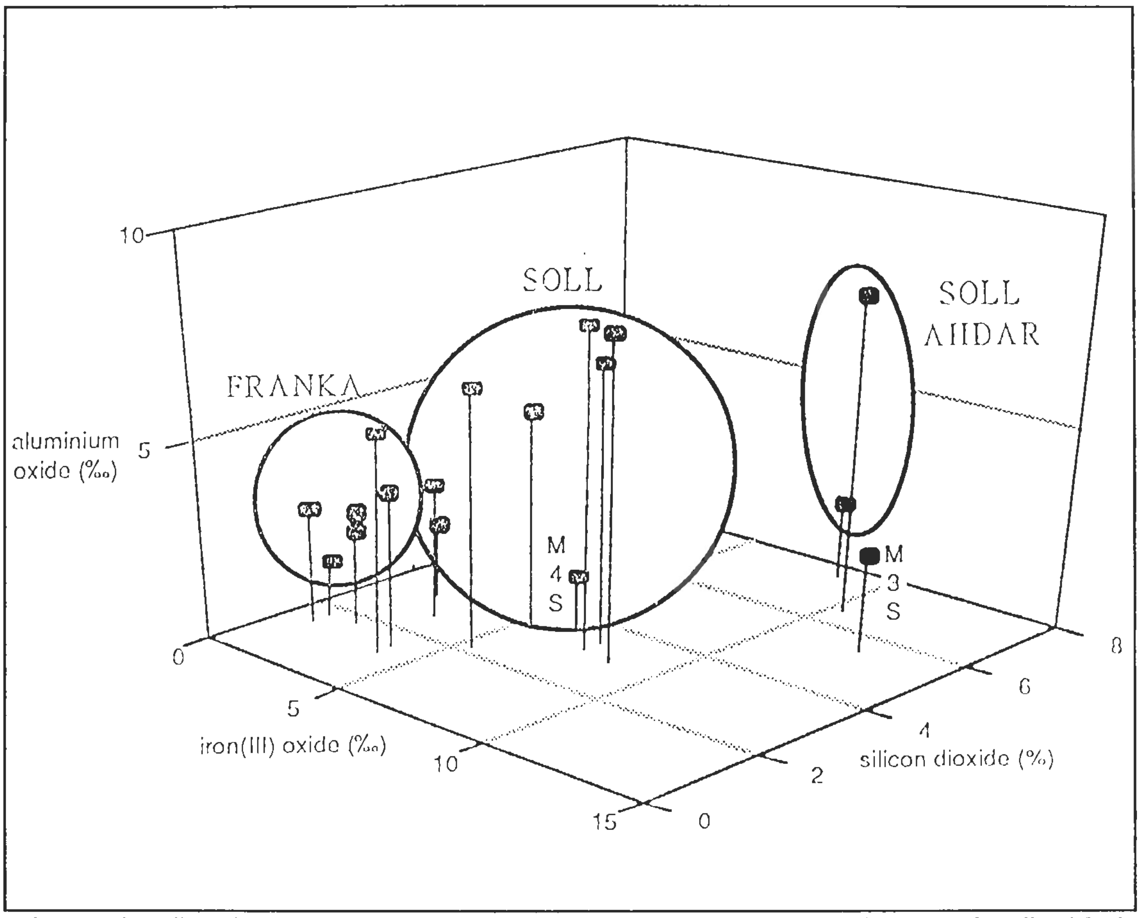

Figure 5. Three-dimensional scatter plot of silicon dioxide, aluminium oxide and iron(III) oxide content for soll and franka samples

To our knowledge this is the first published report on the silicon content of *soll* limestone. In spite of the total lack of experimental information about *soll*, the stone has always been regarded as one containing a high percentage of clay (e.g. Aquilina, 1987). Indeed, although this work does show that there is more silicon in *gebla tas-soll* than in *franka*, the description of the stone as one containing a high percentage of clay is clearly inappropriate in view of the fact that its silica content is less than 5-10%. Petrologically, *soll* is better described as a fairly clean biomicritic limestone and certainly not as a marl.

It does not seem likely that the difference in abundance of siliceous minerals between *soll* and *franka* is large enough to account for the exceptional manner in which *soll* limestone weathers when exposed to the atmosphere. The much reduced resistance to weathering of *soll* probably results from textural differences in the rock caused by a changed depositional environment of the Lower Globigerina sediments brought about by some alteration in the biological activity at the sediment-water interface. This could have been due to increased bioturbation resulting from, perhaps, a reduction in the height of the water column due to a minor regressive shift of the shoreline.

The discrete lenticular occurrences of grey-green limestone found embedded in the yellow coloured *soll* of the Msida outcrop have been shown to be richer in silicon and iron minerals than the surrounding bedrock of *soll*. Our proposal that this grey-green facies be considered as *soll ahdar* seems justified in that the term attempts to take into consideration both the similarities with the normal yellow variety of the rock but also emphasises the facies-difference. There is clearly no difficulty at all in distinguishing *soll ahdar* limestone from the *franka* of the Lower Globigerina.

Finally, although a silicon anomaly has been established for the *soll* at two of its outcrops, one cannot extrapolate confidently this conclusion to all *soll* occurrences of the Maltese Islands. Clearly, a much larger geochemical survey needs to be undertaken. In our view, such a survey would provide basic information which would be required for an eventual proper assessment of the resource potential of the Lower Globigerina.

Note

This paper is dedicated to the memory of Mr Christopher Zammit who has since lost his life in a traffic accident.

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