Journal of Archaeological Science: Reports 22 (2018) 179–192

Contents lists available at ScienceDirect

Journal of Archaeological Science: Reports

journal homepage: www.elsevier.com/locate/jasrep

Greco-Roman mineral (litho)therapeutics and their relationship to their microbiome: The case of the red pigment *miltos*

E. Photos-Jones[a,b,*], C.W. Knapp[c], D. Venieri[d], G.E. Christidis[f], C. Elgy[e], E. Valsami-Jones[e], I. Gounaki[d], N.C. Andriopoulou[f]

[a] *Analytical Services for Art and Archaeology (Ltd), Glasgow G12 8JD, UK*
[b] *Archaeology, School of Humanities, University of Glasgow, Glasgow G12 8QQ, UK*
[c] *Civil and Environmental Engineering, University of Strathclyde, Glasgow G1 1XQ, UK*
[d] *School of Environmental Engineering, Technical University of Crete, 73100 Chania, Greece*
[e] *School of Geography, Earth and Environmental Sciences, University of Birmingham, Edgbaston, Birmingham B15 2TT, UK*
[f] *School of Mineral Resources Engineering, Technical University of Crete, 73100 Chania, Greece*

ARTICLE INFO

Keywords:
Greco-Roman mineral medicinals
Lithotherapeutics
Miltos
Minerals
Microbiota
Nanoparticles
Bioactivity

ABSTRACT

This paper introduces a holistic approach to the study of Greco-Roman (G-R) lithotherapeutics. These are the minerals or mineral combinations that appear in the medical and scientific literature of the G-R world. It argues that they can best be described not simply in terms of their bulk chemistry/mineralogy but also their ecological microbiology and nanofraction component. It suggests that each individual attribute may have underpinned the bioactivity of the lithotherapeutic as an antibacterial, antifungal or other. We focus on *miltos*, the highly prized, naturally fine, red iron oxide-based mineral used as a pigment, in boat maintenance, agriculture and medicine. Five samples (four geological (from Kea, N. Cyclades) and one archaeological (from Lemnos, NE Aegean)) of *miltos* were analyzed with physical and biological science techniques. We show that: a. Kean *miltos* and Lemnian earth/*miltos* must have been chemically and mineralogically different; b. Lemnian *miltos* must have been more effective as an antibacterial against specific pathogens (Gram + and Gram − bacteria) than its Kean counterpart; c. two samples of Kean *miltos*, although similar, chemically, mineralogically and eco-microbiologically (phylum/class level), nevertheless, displayed different antibacterial action. We suggest that this may constitute proof of microbial ecology playing an important role in effecting bioactivity and, interestingly, at the more specific genus/species level. From the perspective of the historian of G-R science, we suggest that it may have been on account of its bioactivity, rather than simply its 'red-staining' effect, that *miltos* gained prominent entry into the scientific and medical literature of the G-R world.

1. Introduction

Greco-Roman (G-R) medical and other texts (for example, Theophrastus *On Stones*; Dioscorides *De Materia Medica* Book V; Pliny *Natural History* Book 35) list not only plants but also minerals and animal products as therapeutic agents, albeit the last two in fewer numbers. These therapeutic materials derived from specific localities and had many and diverse applications in fields not obviously related to medicine, such as pigments, washing powders, textile treatment, or agriculture. Curiously, the basis on which minerals built up their multiple market applications has largely gone unquestioned by modern scholarship; perhaps it was deemed idiosyncratic of the G-R world. Nevertheless, questions remain: was the *same* mineral/mineral combination extracted from each locality and used in each market application and how was 'same' to be assessed?

The study of the minerals industry of the G-R world is still in its infancy. This is primarily on account of the inherent difficulty in attributing what are largely descriptive terms to inorganic substances with precise chemical compositions and crystal structure. G-R minerals nomenclature incorporates both the natural (the raw material) and the processed (the marketable product) and does not always separate between the two. In other words, the G-R texts present the reader with the mineral's commercial or group name, meaning that many different mineral varieties were traded and exchanged under the same name. In an attempt to navigate around this network of complex information, scholars have traditionally assumed a best-fit approach. For example, *lithargyros* has been translated as 'litharge', meaning the mineral PbO. That is despite Dioscorides' (De Materia Medica V.87) and Pliny's

* Corresponding author at: Analytical Services for Art and Archaeology (Ltd), Glasgow G12 8JD, UK.
E-mail address: effie.photos-jones@glasgow.ac.uk (E. Photos-Jones).

https://doi.org/10.1016/j.jasrep.2018.07.017
Received 7 April 2018; Received in revised form 7 July 2018; Accepted 16 July 2018
2352-409X/

(Natural History 33.35) efforts to make it clear to their readers that *lithargyros* existed in three varieties, acknowledging, implicitly, both different method of manufacture as well as a different chemical product.

We therefore argue that in order to understand the nature of the G-R minerals and the properties underpinning their diverse applications, they should be studied 'at source' and based on geological samples. This does not mean that we are confident that ancient miners worked the exact same mineral as the ones we have sampled. Rather, when ancient miner and today's geoarchaeologist confront the same type of 'deposit', in the same locality, there is sufficient confidence that ancient and modern observations will coincide, albeit expressed differently. Having established the need to examine minerals at source, it follows that there is a need to put in place a methodology for their characterisation. In this paper we argue that, apart from their mineralogy and geochemistry, it is vital to study their microbial ecology as well because of the link between microorganisms and their environment, as discussed below.

We focus here on a single G-R mineral, *miltos* (*ruddle*, *L. rubrica*), the naturally occurring, highly prized, fine-particled red iron oxide 'pigment' of antiquity (Cherry et al., 1991; Photos-Jones et al., 1997; Lytle, 2013). *Miltos* first appears in the literature in the 4th century BC (Caley and Richards, 1956 translation of Theophrastus *On Stones*, 52–54) but it actually features in early writing (Linear B script) on Mycenaean clay tablets dated to the late 2nd millennium BC (Blakolmer, 2004). *Miltos*, unlike 'common' ochres (Theophrastus *On Stones*, 51) or '*haematitis lithos*' (hematite) (Dioscorides *De Materia Medica* V.114), had very specific places of origin, including the Cycladic island of Kea, the island of Lemnos in the NE Aegean and Sinope/Cappadocia in Turkey (Fig. 1a and b). The Sinopic *miltos* was actually produced in Cappadocia but shipped from Sinope (Theophrastus, *On Stones*, 52). *Miltos* was primarily characterised by its colour, red, which had considerable staining power (Aristophanes, *Ecclesiazousae*, 387). It had diverse applications, as a pigment, as a cosmetic,[1] in ship maintenance,[2] agriculture[3] and

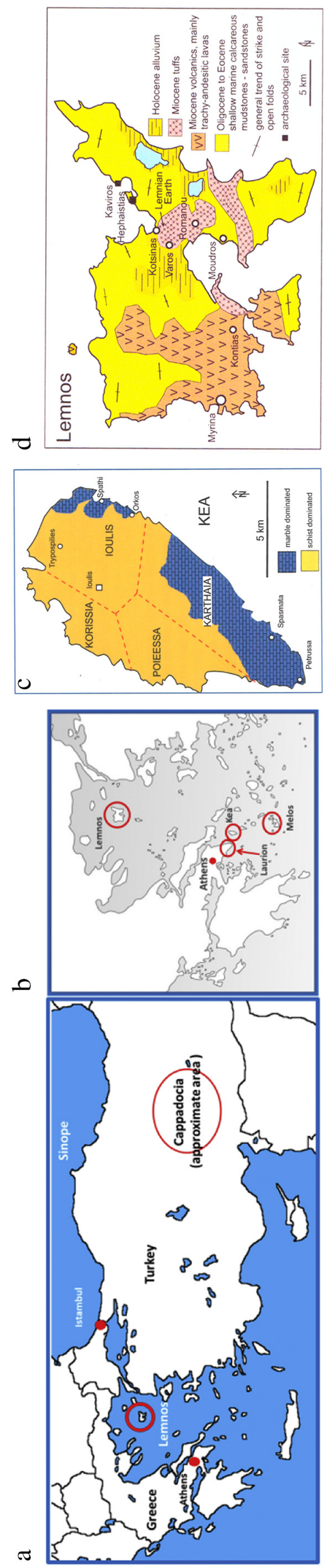

Fig. 1. a. Map of Greece and Turkey with localities mentioned in the text. b Map of Attica and the Aegean with localities mentioned in the text. c Map of Kea with approximate outline of the realm of each of the four city states in the 4th century BC and localities of extraction of *miltos* (after Photos-Jones and Hall, 2011, Fig. 45). d. map of Lemnos showing locality of Kotsinas, place of extraction of Lemnian Earth (after Photos-Jones and Hall, 2011, Fig. 31). Petroussa is the area between localities 4 and 5 on this map.).

[1] As a cosmetic miltos appears in Herodotus (*Hist.* 4.191; 7.69) in reference to body paint amongst African (Libyan and Ethiopian) tribes. Actors used it (Athenaeus 5.197f–198a), as did Aristophanes himself who appears to have made use of it instead of a mask. In 4th century BC Athens, women normally applied red plant-based cosmetics (ἔγχουσα) rather than (red) minerals, but it appears that also some men did opt for *miltos*, dispersed in olive oil, to rub their bodies with, a practice rather frowned upon by Xenophon (*Oeconomicus* 10). Hannah (2005, 204) offers an explanation for this rather socially unacceptable practice '*Whether as a dry powder or as blended into scented oil, its purpose must have been to improve the appearance of the skin of the wearer… by creating the impression of a sun tanned healthy male without the chore of exercise at the gymnasium*'. Olive oil is a good dispersant (a liquid carrier to prevent agglomeration) for fine iron or lead oxide particles (Arulmozhi and Mythili, 2013); thus Kean *miltos* mixed with olive oil would have spread evenly on the skin to create a good 'suntan' effect. In architecture, or for joinery work, *miltos tektoniki* was used, with places of origin, reported, i.e. Egypt and Spain (Dioscorides, V.96).

[2] Inscription IG II^2 1128 found in the Agora of Athens concerns a decree dated (*c.* 360 BC) issued by Athens to the three city states of Kea (Ioulis, Korisseia and Karthaia (Fig. 1b)) requiring them to export *miltos* in its entirety from their respective mines exclusively to Athens, even bearing the charges for the transport. The tone is severe and there were penalties imposed. This has been considered rather an odd 'demand' given that Athens had access to the large iron oxide deposits of Laurion (see Geology section) and therefore the meaning of this inscription has been interpreted in different ways. Lytle (2013) has concluded that Kean *miltos* must have been used in boat maintenance, as an antifouling agent, and to a lesser extent as a decorative paint.

[3] Lytle (2013) presents new evidence associated with the use of *miltos* in agriculture. In the late 4th century BC, *miltos* is reported to have been mixed with pitch (*miltopissa*) in an inscription from Eleusis (*IG II*^{2} 463.90) (Lytle 2013, 537). In a 3rd century AD agricultural manual, the author recommends that Lemnian *miltos*, in a paste with water, is smeared around the roots of plants in an attempt '*to prevent trees and vines from being harmed by worms or anything else*' (Lytle, 2013, 538) and in particular fig trees (Lytle 2013, 539); *miltos* is also placed with olive oil lees (*amurgo*) around roots (Lytle 2013, 539). Separate and