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PREHISTORY OF CLAY MINERALOGY – FROM ANCIENT TIMES TO AGRICOLA

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ABSTRACT
The prehistory of clay mineralogy is highlighted from the beginnings in ancient Greece to the mineralogical works of Agricola, in particular his famous handbook of mineralogy, entitled *De natura fossilium* (1546). Starting with a few scattered hints in the works of Archaic and Classic Greek authors, including Aristotle, the first treatment of clays as a part of mineralogy is by Theophrastus. This basic tradition was further supplemented by Roman agricultural writers (Cato, Columella), Hellenistic authors (the geographer Strabo and the physicians Dioscorides and Galen), the Roman engineer-architect Vitruvius, and finally summarized in Pliny's encyclopedia *Naturalis historia*, which has become the main source for later authors, including Agricola. It is shown to what extent Agricola's work is just a great summary of this traditional knowledge and to what extent Agricola's work must be considered as original. In particular, Agricola's attempt to a rational, combinatorical classification of "earths" is recalled, and a plausible explanation is given for his effort to include additional information on Central European clay deposits and argillaceous raw material occurrences. However, it is shown that – in contrast to common belief – Agricola was not the first to include "earths" in a mineralogical system. This had been done almost one thousand years earlier by Isidore of Seville.

KEYWORDS: clay mineralogy, history, clays, earths, ochre, ruddle, Agricola, Aristotle, Theophrastus, Vitruvius, Pliny, Dioscorides, Isidore of Seville.

INTRODUCTION

Clay mineralogy is usually considered to be a young science (Bergaya et al., 2006), and it is true that the scientific investigation of clays and clay minerals was for a long time much impeded by their variable composition and texture, complicated chemistry, unknown structure and small crystal size. Therefore, there are many reasonable arguments in favoúr of the opinion that scientific clay mineralogy has only been possible since the discovery of X-ray diffraction by Laue and coworkers (1912) and the development of more sophisticated analytical methods at the beginning of the 20th century, see (Konta, 1957; Bergaya et al., 2006). Nevertheless, about one hundred years earlier, at the beginning of the 19th century, the scientific interest in clays had already experienced a subtle renaissance, represented for example by the electrophoretic experiments of Reuss (1809; see Bergaya et al., 2006), and by many passages in Goethe's geological-mineralogical writings (1785-1824; see Pabst, 2000).

Much earlier, however, it has been acknowledged that clays, or "earths", should have their place as a distinct class of materials in systematic mineralogy. And it is clear that empirical knowledge concerning clays and clay minerals, or "earths", accompanied the history of mankind from its very dawn. In this contribution an attempt is made to highlight the "prehistory" of clay mineralogy from its beginnings in ancient Greece to the mineralogical works of Agricola, in particular his famous handbook of mineralogy, entitled *De natura fossilium* (1546). It will be shown to what extent Agricola's work is just a great summary of traditional knowledge and to what extent Agricola's work must be considered as original. In particular, Agricola's attempt towards a rational (combinatorical) classification of "earths" will be recalled, and a plausible explanation will be given for his intense effort to include information on Central European clay deposits and argillaceous raw material occurrences. However, it will be shown that, in contrast to common belief, Agricola was not the first to include "earths" in a mineralogical system, but that this had been done almost one thousand years earlier by Isidore of Seville in his *Etymologiae*, also called *Origines*, written in the years between 620 and 636 A.D.

FROM ARCHAIC GREECE TO THEOPHRASTUS

ARCHAIC GREEK AUTHORS

In archaic Greece "earths" or "clays" are mentioned only with respect to their main application in ceramics (bricks) and, exceptionally, as pigments (ochre and ruddle), see (Lenz, 1861). Homer's *Iliad* (8th century B.C.) contains the first written reference to the work of the potter (κεραμευς), who operates the potter's wheel (τροχος) and the common custom to drink wine from earthen jars (εκ κεραμων).

Moreover, both the *Iliad* and the *Odyssey* inform us that certain ships were coloured with ruddle, i.e. red ochre (νεες μιλτοπαρηοι). The potter is also mentioned by Hesoid (8th-7th century B.C.) in *Opera et dies* (εργα και ημεραι). Further, Herodot (484 – post 430 B.C.), in his *Historiae* (Ιστοριες αποδεξις) explains that the walls of Babylon are built from earth (γη), which is dug, shaped into the form of bricks and burnt in ovens (kilns, furnaces). In the walls these bricks are glued together with warm asphalt as a mortar. Herodot also mentions the Egyptians' use of "stamping earth" (σεμαντρις) to seal the papyrus labels used to mark holy bulls and gives some details on the construction of pyramids, including a pyramid made of bricks. Xenophon (430 – post 355 B.C.), in his *Anabasis*, confirms Herodot's description of the Babylonian walls as "made of bricks (πλινθος οπτη) lying in asphalt" and adds the information that the walls of Larissa (a town near the Tigris river) are made of bricks (πλινθοι κεραμιαι) but based on natural stone (κρεπις λιθινη), while the walls of Mespila consist of a natural limestone base (κρεπις λιθου ξεστου κογχυλιατου), on which the brick wall (πλινθινον τειχος) is built (Lenz, 1861).

ARISTOTLE

At the end of the third book of his *Meteorology* Aristotle (384-322 B.C.) outlines a "general theory" of bodies "quarried" (ορυκτα) and bodies "mined" (μεταλλευτα). He writes (here and in the following the symbol "[…]" denote omitted text passages from the original translations): "[…] there are two exhalations, one vaporous the other smoky, and there correspond two kinds of bodies that originate in the earth, things quarried and things mined. The heat of the dry exhalation is the cause of all things quarried. Such are the kinds of stones that cannot be melted, and realgar, and ochre, and ruddle, and sulphur […], most things quarried being either coloured lye or, like cinnabar, a stone compounded of it. The vaporous exhalation is the cause of all things mined – things which are either fusible or malleable such as iron, copper, gold. All these originate from the imprisonment of the vaporous exhalation in the earth, and especially in stones. Their dryness compresses it, and it congeals […]. Their matter was that which might have become water, but […] the evaporation congealed before water was formed. […] This is the general theory of all these bodies […]" (Aristotle, 1995). In the following fourth book he gives a more detailed description of liquid and solid bodies (organic and inorganic) on the basis of this "general theory" and his basic theory of the four "elements" (earth, water, air, fire) resulting from the four "causes" (two active ones – the hot and the cold, two passive ones – the dry and the moist). These two small passages contain the main part of what is extant from Aristotle's view on mineral substances and demonstrate that inorganic matter, at least from the chemical and mineralogical point of view, was only of peripheral interest in Aristotle's philosophy and science of nature. Whenever clay is mentioned (mainly in the connection "potter's clay") it is only as a paradigmatic example for Aristotle's general theory of matter as mixtures of elements. In this sense it demonstrates a view detailed in other Aristotelian writings (*Physics*, *On the Heavens*, *On Generation and Corruption*). Concerning ceramic processing and clay products Aristotle writes in the fourth book of the *Meteorology*: "Bodies that are soft but not liquid do not thicken but solidify when the moisture leaves them, e.g. potter's clay in process of baking […]. Those bodies which have first been thickened or hardened by cold often begin by becoming moist: thus potter's clay at first in the process of baking steams and grows softer, and is liable to distortion in the ovens for that reason. Now of the bodies solidified by cold which are made up both of earth and water but in which the earth preponderates, those which solidify by the departure of heat melt by heat when it enters into them again; this is the case with frozen mud. […] Mud and earth, too, melt. Of the bodies which are solidified by dry heat some are insoluble, others are dissolved by liquid. Pottery […] cannot be dissolved. Natron and salt are soluble by liquid, […] water and any of its varieties melt them but oil does not. […] If a body contains more water than earth fire only thickens it: if it contains more earth fire solidifies it. Hence natron and salt and stone and potter's clay must contain more earth. […] Pottery consists of earth alone because it solidified gradually in the process of drying. Water cannot get into it, for the pores were only large enough to admit of vapour escaping: and seeing that fire solidified it, that cannot dissolve it either" (Aristotle, 1995). It is evident that Aristotle did not distinguish between the individual types of "earth" or "clay". He uses the word in a very general sense, although he – as a physician and the son of a physician at the Macedonian king's court – was certainly aware of the use of different clays in medicine for example. For Aristotle solidification and melting are essentially processes of eliminating or adding the element "water" (or the cold and moist principles). For him, "bodies are formed by heat and cold […]." Aristotle continues by explaining solidification in more detail: "Of all the bodies that admit of solidification and hardening, some are brought into this state by heat, others by cold. Heat does this by drying up their moisture, cold by driving out their heat. Consequently some bodies are affected in this way by the defect of moisture, some by defect of heat: watery bodies by defect of heat, earthy bodies of moisture. Now these bodies that are so affected by defect of moisture are dissolved by water, unless like pottery they have so contracted that their pores are too small for the particles of water to enter." (Aristotle, 1995). Obviously, Aristotle tries to explain two different ways of liquefaction, melting and dissolution, in a common theoretical framework – an attempt which must appear futile from the viewpoint of modern science. Nevertheless, a fruitful, unifying concept in Aristotle's theory is his view of the porous microstructure of materials. Furthermore, it has to be emphasized that Aristotle had a relatively clear conception concerning the mechanical properties and rheological behavior of materials (he fully recognized e.g. the brittle nature of pottery and stone in contrast to metals and plastic and viscous materials). His examples of inorganic materials include realgar, ochre, ruddle, sulphur, cinnabar, salt, natron, ice, gold, silver, copper, iron (steel), tin, lead, as well as stone (including the mysterious "millstone"), pottery and glass, but Aristotle himself made no attempt to present a systematic classification of mineral substances.

THEOPHRASTUS

Theophrastus' (372-287 B.C.) *De lapidibus* (Περι λιτηον, written around 321-315 B.C., i.e. shortly after Aristotle's death) is the first monothematical treatise on mineralogy and deals with "earths" or "clays" (i.e. argillaceous raw materials) for the first time as a part of mineralogy (Mieleitner, 1922, Theophrastus, 1965). Among the distinctive properties of stones in general Theophrastus mentions colour, hardness-softness, and smoothness. He reports that yellow ochre (ωχρα) and red ochre (μιλτος) are found in mines (μεταλλον), the latter also in iron ore quarries (σιδεριον), and that both are earth-like. According to Theophrastus the latter is found everywhere (e.g. on the islands of Lemnos and Ceos and in Cappadocia, from where it is shipped via the town of Sinope) and both can be used in painting, the former replacing orpiment (αρσενικον); there are three natural types of red ochre (ruddle) – a dark red, a light red and an intermediate one. Theophrastus also reports that red ochre (mainly coloured by hematite) can be produced artificially by heating yellow ochre (mainly coloured by limonite) in covered vessels sealed with loam (πηλος) – the longer the heating the more intense (darker) the red colour. Melian earth (μηλιας) from the island of Melos, used by painters, is characterized by Theophrastus as soft (mild), rough and meagre, Samian earth (σαμια) from Samos (which Theophrastus describes as mined, i.e. not quarried), used by fullers for cleaning cloth (σμεχειν → σμεκτις), is described as unctuous, hard (tough) and smooth. Also Cimolian earth (κιμωλια) from Cimolos is reported to serve "for another purpose" than for painting (more than 350 years later, this is interpreted by Pliny as if Cimolian earth had the same purpose as Samian earth, viz. cleaning cloth – i.e. Pliny, and after him others, including Agricola, identify Theophrastus' "Cimolia" with what later became "fuller's earth", i.e. bentonite or clay with a high smectite content), see Beneke, Lagaly (2002) and Robertson (1986). As a further substance for cleaning cloth Theophrastus mentions "Tymphaic earth" (τυμφαικη) or gypsum (γυψος) and in this connection he comments upon the preparation of gypsum powder by burning and the use of plaster, which results in a hard mass after being mixed with water shortly before use, e.g. for gluing together stones in walls or as wall surface coatings. All this will reoccur, mediated by Pliny and others, in Agricola's work.

HELLENISTIC AND ROMAN AUTHORS

VITRUVIUS

Apart from two marginal notes on clay seal stamps in Cicero's works (Lenz, 1861) and a few passages in Cato's (234-149 B.C.) *De re rustica* (also called *De Agricultura*), where he mentions broken stone (cementum) and lime (calx or calx cocta) as building materials, tiles (tegulae), quartz pebbles (silex), the preparation of lime by calcination in a furnace (fornax calcaria), loam (lutum) and the use of chalky earth or red ochre (terra cretosa vel rubricosa) for wall paints (Cato, 2008; Lenz, 1861), the first important Roman author treating earths and clays in greater detail is Vitruvius (1st century B.C.). In his famous work *De architectura* (written after 27 B.C.) he gives a detailed account of building materials (carved blocks – saxa quadrata, broken stones – caementa, fired bricks – coctus later, unfired bricks – crudus later) and explains that unfired bricks must not be produced from sandy (arenosus) or stony (calculosus) clay (lutum), neither from loose sand (sabulo), because they would decay in the rain, and the straw (!) would not adhere in them (Vitruvius, 1953, 2004, 2008). It is reported that they must be made from whitish loam (terra albida cretosa), red ochre (rubrica) or binding sand (masculus sabulo), and in order to achieve uniform (i.e. sufficiently slow) drying without the danger of cracks they must be produced in autumn or spring (because the sun is not too strong in these seasons). The preparation of mortar (from lime / calx and sand in ratios from 1:2 to 1:3) is reported to require careful selection of the sand types (sea sand, river sand, or sand from pits), taking into account their salt and water content (Vitruvius, 1953, 2004, 2008). According to Vitruvius, pumice (spongia sive pumex) is a naturally burnt mixture of tuff (tofus) and earth (terra). In connection with paints he mentions yellow ochre (sil) and red ochre (rubrica, occurring at many places, but high quality types are rare, e.g. from Pontus near Sinope, in Egypt, the Balearic islands and the island of Lemnos), "paraetonium" (according to Pliny a "chalk" from Libya – the difference between "chalk" and "clay" was not clear to the ancient authors and not even to Agricola), "melium" (from the island of Melos, according to Pliny a white clay), green earth (creta viridis) from Smyrna, orpiment (auripigmentum) and realgar (sandaraca) from Pontus (Vitruvius, 1953, 2004, 2008). We learn from Vitruvius that water pipelines are made either of cemented walls or assembled from lead or fired clay pipes. A whole section in *De architectura* is devoted to the powder of Puzzuolo (Puteoli), which Vitruvius describes as a kind of "sandy powder which does remarkable things": mixed with lime and broken stone it not only strengthens buildings, but even makes these ingredients solidify and harden under sea water, so that offshore dams can be built with it. Further, Vitruvius makes an attempt – based on the peripatetic four-element-theory – to explain why this powder is found only close to Mt. Vesuvius and similar places, in close relation to thermal waters and Pompeian pumice or "sponge stone" (spongia), see (Vitruvius, 1953, 2004, 2008).

OTHER ROMAN AND HELLENISTIC AUTHORS BEFORE PLINY

Among the Roman agricultural writers it is mainly Columella (1st century A.D.) who, in *De re rustica* (written around 50 A.D.) treats earths in some detail with respect to their use in agriculture. He mentions tuff (tophus), meagre gravel (glarea) and unctuous earth (pinguis gleba), further chalky soil (cretosa humus), potter's clay or "argilla" (creta qua utuntur figuli quamque nonnulli argillam vocant), coarse sand (sabulo), and ruddle / red ochre (rubrica). He points out that neither pure clay nor pure sand or gravel result in a good soil (for plant growth), but only an appropriate mixture of the two. In connection with ruddle he explains that it is inappropriate for growing vine, because in wet weather it is too sticky, in dry weather too hard (Columella, 2008).

A few scattered hints concerning ancient occurrences and deposits of argillaceous raw material can be found in Strabo's (64 B.C.-19 A.D.) *Geografica*. The Greek author mentions "fiery mud" coming out of the earth (πηλος διαπυρος), red ochre (μιλτος), Sinopian earth (Σινωπικη γη), and the (erroneous) fact that Puzzuolo (Puteoli) has a port with walls made of a mixture of lime and sand (η αμμος), see (Strabo, 2005). He further mentions the volcanoes Mt. Vesuvius and Mt. Aetna and the fertility of the soil around them (good for growing vine), the purple-coloured Armenian earth (η σανδυξ – according to (Lenz, 1861) a red ochre) from the gold mines near Cambala and the Sinopian red ochre (Σινοπικη μιλτος) from Cappadocia (which is reported to be comparable in quality only with Iberian red ochre).

The medical writer Dioscorides (40-90 A.D.), a Greek physician and pharmacologist, gives a more detailed account of earths in *De materia medica* (written around 77 A.D.). He mentions the Armenian earth (αρμενιον), yellow ochre (ωχρα) from Attica, which he describes as "very light, soft and appropriate for firing", further Sinopian ruddle (μιλτος σινοπικη), architects' or carpenters' ruddle (η τεκτονικη μιλτος) from Egypt and Carthago (which is reported to be of lower value than Sinopian ruddle), and Spanish ruddle, which is reported to be artificially produced by firing yellow ochre (Dioskurides, 1902). Further, Dioscorides describes Lemnian earth (λημνια γη) as mined on the island of Lemnos, then "mixed with goats' blood, shaped and labeled with a stamp showing a goat" (Lenz, 1861). He mentions orpiment (αρσενικον), realgar (σανδαρακη) as occuring in the same mines in Pontus and Cappadocia, different types of clay (μιλτος - this word was used exclusively for "red ochre" by previous authors). It is reported that for medical use the cleavable (σχιστη), white, odorous, very astringent, non-sticky one is preferred, which consists of hairy particles (τριχιτις). As further medicaments Dioscorides mentions "dry-burnt" lime (ασβεστος – made from sea shells or marble; asbestos in the modern sense is called λιθος αμιαντος by Dioscorides), and gypsum (γυψος). In another context, Dioscorides mentions "vineyard earth" or "pharmacitis" (αμπελιτις γη) from Syria, used for dying hair and as an insecticide, which he describes as black (similar to charcoal), easily cleavable, glossy and melting (i.e. dissolving) in oil, according to (Lenz, 1861) either a bituminous clay or a type of coal. Dioscorides' *De materia medica* became the foremost classical source of modern botanical terminology and the leading pharmacological text for sixteen centuries (it describes approx. 1000 "simple drugs", among them 600 of plant origin). Similar information can be found in the books of Galen (129-199 A.D.), another Greek physician and one of the most distinguished physicians of antiquity (Encyclopaedia Britannica, 1980). The influence of these and other medical writings on Agricola's work cannot be overestimated (see below), since during his stay in Italy (1524-1526) Agricola was actively engaged with Andreas Asulanus in the edition of Galen's Complete Works published by Manutius in Venice (1525). Shortly before, the works of Aristotle and Theophrastus, as well as Dioscorides' *De materia medica* (1499), had been published there, followed a few years later by the works of Hippocrates (1526) and Paulus Aeginatus (1528), where Agricola was again among the editorial assistants (Krafft, 2006).

PLINY

Pliny's (23-79 A.D.) 37-volume *Naturalis historia* (written before 77 A.D.) is the most complete encyclopaedia of Antiquity and has become the major source for natural sciences for more than 1000 years (Encyclopaedia Britannica, 1980). The mineralogical knowledge is contained mainly in books 33–37, with a few facts mentioned elsewhere. For "earths" and argillaceous raw materials books 33 and 35 are the most interesting (Plinius, 2007, Pliny, 2008). A detailed comment on all relevant passages in this work is beyond the scope of the present contribution. Therefore we restrict ourselves to only a few keywords, with special regard to information which is not extant from previous authors. In his text he mentions gypsum (gypsum), mortar (maltha), limestone (calx), lime (calx viva), burnt lime (calx recens), marble (marmor), potter's clay (argilla) and the use of earthen (fictilis) pipes as water ducts. In book 33 he explains that crucibles for melting metals are made of a white clay (tasconium), because "other earths do not withstand the air stream, the fire and the fiery metal" (Plinius, 2007, Pliny, 2008). He also mentions the fact that clay (argilla) is used for iron welding, and further orpiment (auripigmentum), realgar (sandaraca), minium (minium) from silver-mines (used as a pigment), ruddle or red ochre (rubrica, Greek miltos), cinnabar (cinnabaris) and Sinopian earth (sinopis) for painting, yellow ochre (sil) from gold- and silver-mines ("the best […] from Attica"), also used for painting (the dark one for shadows, the light one for light in paintings; for wall paintings mixed with marble powder, because "marble withstands the influence of fresh lime coatings"), see (Plinius, 2007, Pliny, 2008). In book 35 Pliny enumerates the natural colours used by painters: Sinopian earth, red ochre (rubrica), Paretonian earth (paraetonium, "after the place in Egypt where it is found"), Melian earth (melinum, after the island of Melos), Eretrian earth (eretria, after the place where it is found); Samian earth is reported to be too unctuous for painters (Samian earth is dug in cracks intersecting rocks, and when touching the tongue it is astringent). Sinopian earth (sinopis) is reported to come from the Pontus and "named after Sinope, a town in Pontus", further from the Balearic islands and Africa; the best, however, coming from Lemnos and Cappadocia, "where it is dug out of the earth", i.e. mined, not quarried – three types: a more red / dark red one, a less red / light red one; the one from Lemnos is estimated higher than the other types and is closest to cinnabar (minium); the commercial product is stamped (sealed), therefore called sealed earth (sphragis); cinnabar (minium) is sometimes mixed with it and it is praised in medicine; found in iron mines (quarries ?); ochre (yellow ochre ?) is made from ruddle (here Pliny seems to misinterpret Theophrastus' information) by firing it in vessels sealed with loam (the stronger the fire the better the colour). In book 35 Pliny also gives two opinions on the invention of sculpture (plastica): according to the first, it had been invented by Butades, a potter in Corinth: he made a sculpture out of clay (argilla) and fired it with other pottery (cum ceteris fictilibus); according to the other opinion, sculpture had been invented on the island of Samos. Pliny tells us that Butades also mixed clay with red ochre (rubrica) or made sculptures with red clay (rubra creta) and that the art is called "plastica", the artisan "plastes" (this reoccurs later in Agricola's work). Pliny also compares cast gypsum (plaster) products and sculptured products made of clay without firing with sculptures of fired clay (fictilis), painted with minium (here apparently a lead-containing glaze). He mentions fired clay vessels (figlinarum opus), roof tiles (imbrex), bricks and pottery made on the potter's wheel (rota), and the potters' club (collegium figulorum). He reports that Samian and (in Italy) Arretinian tableware are in high esteem, facts that are later repeated in Agricola's work. Probably with reference to Vitruvius, Pliny reports that "an apparently unimportant component of earth, called powder (pulvis), is found on the hills of Puzzuolo (Puteoli) and is used to construct dams against the sea waves; in water it is changed into a indestructible stone which gets harder with each day, especially when mixed with the broken stone (caementum) of Cumae" and adds other examples of earths that can change into stone, see (Plinius, 2007, Pliny, 2008). Pliny also reports that form walls (paries formaceus) are built by pressing earth between two wooden boards, which harden to such a degree that rain, wind and fire cannot destroy them and they become harder than broken stone (caementum). As an example he adds the fact that Hannibal had built earthen watchtowers in Spain (also this detail will later be repeated by Agricola and – together with many other similar cases – reveals Pliny as his main source). Pliny is fully aware of the different uses of earths or clays as building materials. He says that houses are built by coating the wooden skeleton with loam or by constructing with loamstones (later crudus) and describes the raw materials and processes in brick production in a similar way as Vitruvius: "bricks are made neither from sandy (sabulosus) nor from gravelly (arenosus) nor from stony (calculosus) earth (solum), but from argillaceous, whitish or red earth (ex cretoso et albicante aut ex rubrica) or at least from sandy, hard (i.e. clayey) earth (ex sabuloso masculo); the best time for making bricks is in spring, because those made in summer are liable to cracking; the earth from which bricks are made must be completely wet".

With respect to applications of earths in medicine Pliny describes Samian earth (two types: kollyrion – fresh and soft, aster – lumpy and white, both types are heated and washed), Eretrian earth (two types: white and ash-grey, tested with respect to softness), Chian earth (white, medical properties similar to Samian earth), earth from Selinus (milky-white and easily dissolved in water, with milk used for wall coatings), pnigitis (very similar to Eretrian earth, but occurring in larger lumps, sticky, with properties like Cimolian earth, but weaker) and ampelitis (similar to asphalt, soluble in wax and oil, while retaining its dark color; as a medicament softening and dividing, also used for colouring hair). As an extra group he adds "several types of white clay" (creta); although for physicians two types of Cimolian earth are important (the white one and the purplish one), Cimolian earth is mainly used for treatment of cloth. Pliny is apparently the first who gives a recipe for cloth treatment: according to the "fullers' law", set under the censors Caius Flaminius and Lucius Aemilius, the cloth has first to be washed with Sardian earth (creta), then treated with sulphur; true and precious colours are softened by Cimolian earth, while for white clothes claystone (saxum) is better (however, the latter can be applied only after the sulphur treatment; it is deleterious to other colours); in Greece they use Tymphaeian gypsum instead of Cimolian earth. As a result of Pliny's authority all these informations, including many details not mentioned here (e.g. silversmiths' chalk = creta argentaria) and the less obvious canonical part of mineralogy (e.g. metals, slags and gems, glass production, pumice and obsidian), found their way into many later works, including those of Isidore and Agricola.

ISIDORE OF SEVILLE – THE UNKNOWN MINERALOGIST

Isidore of Seville (Isidorus Hispalensis, 560-636) was a theologian (later he became archbishop of Seville and entered Catholic hagiography as Saint Isidore) and encyclopaedist. His main work *Etymologiae* (written in the period between 621-636) "became one of the most studied works during the Dark and Middle Ages" (Encyclopaedia Britannica, 1980). In the sixteenth book of the *Etymologiae* (Isidore, 2000, 2008, 2009), entitled *De lapidibus et metallis* (Stones and metals), Isidore summarizes the mineralogical knowledge of his time, presenting a classification containing powders and lumps of earth (De pulveribus et glebis terrae, chapter 1), matter solidified from water (De glebis ex aqua, chapter 2), common stones (chapter 3), rarer stones (chapter 4), marbles (chapter 5), precious stones (chapters 6-15), glass (chapter 16), metals (chapter 17-24), followed by three chapters on measures and their labels. Although the information given by Isidore is obviously based on the aforementioned tradition, his treatment of the subject and his subdivision into chapters is in many respects original and to a certain degree even independent of Pliny. In particular, the treatment of earths in a separate chapter is new, the fineness of earths is emphasized as their main characteristic feature ("clay or loam is called limus because it is fine", i.e. of small particles size), and definitions are given for powders (pulvis, i.e. "what can be whirled up by the wind and transported") and lumps of earth (gleba, i.e. "a single agglomerate of many powder particles glued and held together"). In other words, Isidore distinguishes, for the first time in history, clearly between "bound earth" (terra ligata, i.e. lumps) and "free earth" (terra soluta, i.e. powders). On the other hand – again in contrast to Pliny – Isidore makes no attempt to be exhaustive; his examples of earths (taken from previous sources) are relatively few, including only potter's clay (argilla, from which "vessels – vasa – are made", i.e. a kaolinitic or plastic clay), chalk (creta, possibly in the meaning of "clay" in general), Cimolian earth (creta Cimolia, "used for cleaning cloth", i.e. fuller's earth, a smectitic clay), silversmith's chalk (creta argentaria, i.e. a chalk which has been used for cleaning silver even in modern times) and Samian earth (terra Samia, probably a kaolinitic clay), which he describes as "glutinous (glutinosa), white (candida) and smooth (lenis) to the tongue, used for the preparation of small vessels (vascula) and in medical applications (medicamenta)". The chapter on earths further mentions ash (cinis), sand (sabulum), silt (lutum) as examples of powders, as well as the very special Puteolian powder (pulvis Puteolanus), "collected near the hills of Puzzuolo (Puteoli) in Italy and used in coastal dam construction". Isidore reports that "when thrown into water it is immediately changed into stone", an obvious citation from Vitruvius and / or Pliny. In full accord with Plinian tradition, he adds in his chapter on earths a remark on the "four types of sulphur"; similarly, in the following chapters he closely follows Pliny (and / or Vitruvius) in his remarks on gypsum, limestone (calx), lime (calx viva), pumice (pumex), glass (vitrum), obsidian and the mysterious "millstone" (cotis). However, in contrast to Pliny (and Vitruvius), Isidore does not mention the earths used by painters, carpenters, and in brick production. The last three chapters of the sixteenth book of Isidore's *Etymologiae* have a remarkable parallel in Agricola'a late metrological works. There is, however, no direct indication that Agricola knew Isidore's work, although it is now known that more than 1000 manuscripts existed in medieval monastery libraries (Möller, 2008) and it is known that several printed versions were available already in Agricola's time (the work was published for the first time in 1472 by Zainer in Augsburg, immediately followed by editions in Strassburg (1473), Köln (1478), Basel (1489) and two editions in Venice (1483 and 1493), see (Möller, 2008). Agricola's possibly independent efforts in this direction may readily be explained by the confusion in science and business of his time brought about by the use of different measures, as well as by the practical necessity of knowing the ancient measures to restore ancient knowledge, especially in medicine and pharmacology (i.e. for the preparation of medicaments; it is well known that the realization of this program of a unification of measures has been started not earlier than 200 years after Agricola, viz. during the French revolution, and has been accomplished as late as in the second half of the 20th century, resulting in the International System of Units).

AGRICOLA – HIS SOURCES AND ORIGINAL CONTRIBUTIONS

The main sources of Agricola's mineralogical works are well known, mostly from his own explicit quotations of the cited authors, in accordance with Agricola's words "Pliny gives credit openly and frankly to those whose writings he uses, and likewise I shall give credit by name to those whom I quote". These main sources are Aristotle (384–322 B.C., *Meteorology*), Theophrastus (372–287 B.C., *De lapidibus*), Vitruvius (1st century B.C., *De architectura*), Columella (1st century A.D., *De re rustica*), Dioscorides (40–90 A.D., *De materia medica*), Pliny (23–79 A.D., *Naturalis historia*), and Galen (129–199 A.D., *De compositione medicamentorum secundum locos*, *De simplicium medicamentorum temperamentis et facultatibus*). Other ancient authors are mentioned only marginally, e.g. Strabo (64 B.C.–19 A.D., *Geografica*), and the only medieval author quoted in *De natura fossilium* (1546) is Avicenna (980-1037, *De congelatione et conglutinatione lapidum*, *Canon medicinae*). Isidore is not quoted, and it seems that Albert the Great (Albertus Magnus, 1200-1280), although cited by Agricola e.g. in his early mineralogical dialogue *Bermannus* (1530), is not recognized by Agricola as a serious or reliable source – probably due to his proximity to medieval Alchemy (Agricola, 1957).

Agricola's biography has been thoroughly investigated, and the reader may refer to many studies elucidating the relations between his life and work (Kořan, 1956; Kettner, 1957; Parma, 1957; Engewald, 1994; Krafft, 1994; Majer, 1994a, 1994b; Richter, 1994; Krafft, 2006). The editions of Agricola's works, from the first editions to the currently available ones, have been carefully compiled in (Prescher, 1994). His handbook of mineralogy *De natura fossilium* (1546), see (Agricola, 1546, 1955, 1958, 2006), has to be viewed as complementary to another work concerning a second group of "subterranean bodies" (corpora subterranea), viz. those flowing out of the earth by themselves (i.e. without the necessity of being quarried or mined, e.g. mineral waters or crude oil), which appeared in print together with the first work in a joint volume (*De natura eorum quae effluunt ex terra*, 1546). The mineral raw materials that have to be dug (i.e. quarried or mined) are called *fossile* and subdivided into non-composites (concreta ex partibus sui similibus substancia) and composites (concreta ex partibus sui dissimilibus substancia), the non-composites into simple ones and mixed ones. While Book I gives a general introduction explaining this classification scheme, composites and mixed non-composites make up the major part of Book X (although "earths" can also be part of "composites" an adequate interpretation of this complicated last book cannot be given in a few sentences and is beyond the scope of this contribution). The simple non-composites are then divided into earths (terrae, Book II), "congealed juices" (succi concreti, Books III and IV), stones (lapides, subdivided into common stones / lapides / Book V, precious stones / gemmae / Book VI, marbles / marmora / Book VII, rocks / saxa / Book VII) and metals and related materials (metalla et rei metallicae, Books VIII and IX).

The second book of *De natura fossilium* (1546) begins with a detailed criticism of the traditional classification of earths according to their utility and uses. Based upon the aforementioned literature sources, but supporting his arguments by contemporary examples (mainly from Central Europe), Agricola mentions the agricultural use of certain earth types for supporting plant growth and farming, applications of Lemnian, Samian, and Armenian earths in medicine, the use of clay by potters and sculptors, of red ochre by carpenters, of Paraetonian and Melian earth by painters, of Cimolian earth by fullers, and of *creta argentaria* by silversmiths. His main counter-argument is that some earths are used by different professions, further that "this classification does not consider the true nature of the earths and fails to distinguish sufficiently one earth from another" and, therefore, "the expert in natural history cannot use this classification". However, Agricola accepts "the common practice of giving locality names to earths because of the lack of another name" and he believes "that earths from one locality may be worthless while similar ones from another are valuable".

As an alternative to common practice, Agricola proposes a new classification scheme based on properties ("qualities"). In particular, according to Agricola there are four basic "quantifiable" properties, each subdivided into three degrees. These are:

- Meagre – unctuous – intermediate,
- Porous – dense – intermediate,
- Soft – hard – intermediate,
- Smooth – rough – intermediate.

By mutual combination of these properties (in hierarchical sequence from top to bottom) Agricola determines the theoretically possible number of earth species to be 81 (i.e. three-to-the-fourth power). The three degrees (two extremes and one intermediate) are an evident tribute to peripatetic tradition (Aristotle and Theophrastus), but the combinatorical approach (Stoyan, Stoyan, 1994) is new and may be called rational. Apart from the "quantifiable" properties (properties of degree) Agricola acknowledges the existence of other "non-quantifiable" properties ("either-or" properties). These properties, which are related directly to the human senses and may be traced back to presocratic (specifically atomistic) traditions (Pabst, 1994) are colour (variable), taste (sweet, oily, sour, oily-sweet or oily-sour), odour ("agreeable and pleasing" versus "disagreeable and foul" and form (related to the tactile sense, e.g. tabular versus non-tabular). In contrast to the former they do not define new species but only new varieties.

Concerning the amount of information on earths contained in the second book of *De natura fossilium* (1546), Agricola's work doubtlessly exceeds everything that has been written and published on that subject before. Among the most interesting details are his comments on marl (marga). According to Agricola, marl, which is "sometimes used by farmers, is usually hard" (i.e. marl in the sense of an impure, lime-containing clay or loam, the German "Mergel"). On the other hand, Agricola reports that "the word *marga* is derived from the marrow of bones, for sometimes the water which flows from marls is as white as marrow and hence has been given this name by the Germans and the Gauls who speak the same language. The white solution is called *Steinomarga* by our miners. Marl is sometimes found along fractures and joints of rocks but more commonly enclosed within the rock itself. The variety of marl found in mines and quarries is not used by farmers since it occurs in small quantities." It is evident from the context (etymology and reported typical occurrence) that here marga is used – for the first time in the literature – in the sense of nacrite (i.e. the German "Steinmark", a word originating in medieval miners' jargon), which is known to occur in the Krušné hory mountains region and which Agricola probably came to know from his own experience in this area (Horní Slavkov?) or at least from discussions with miners in Jáchymov (Joachimsthal).

In contrast to former discussions of earths (except for Vitruvius in some respects), Agricola is the first who tries to explain the technological properties of clay products (e.g. earthenware vessels) on the basis of their classifying properties. In connection with earths used by potters and sculptors he writes: "Porous clays are rarely used and those which are incoherent are valueless. Unctuous and dense clays are used to make crucibles and scorifiers which are not affected by fire and are used in refining ores and metals. These clays are also used to make vessels which neither absorb nor exude liquids. Vessels that do not absorb liquids are made not only from dense clays but also from sand which is mixed and burnt. These vessels such as those from Waldenburg are in great demand by pharmacists for holding liquids and syrups since they last longer than others; they also withstand fire for long periods of time. Some containers that absorb and even exude liquids are made from unctuous porous clays. These are the only pottery which is not completely burnt. They cannot stand a high heat." Similarly, in another context he writes: "fuller's earths are unctuous but, having been dried over a fire, become acrid and, because of this, possess the power to clean cloth. Many of these earths derive their names from islands and countries. Cimolian earth, also called *smektis*, because it cleans so well, comes from Cimolos, one of the Cyclades islands. Fuller's earths are found today in many parts of Germany; a gray variety at Kaaden (Kadaň), Bohemia." The end of this passage shows Agricola's ability of clearly recognizing smectite-containing earths – the "gray variety at Kaaden" is the bentonite of the Rokle deposit type, the major Czech bentonite deposit of today. These two examples may suffice for demonstrating Agricola's attempt to explain the technological behavior and applicational performance of clay products in terms of their basic (classifying) properties. Moreover, the two text passages show that Agricola tries to combine traditional knowledge (based on, or directly copied from, the aforementioned ancient authors) with examples of contemporary production and deposits in Central Europe. The latter had become a necessity not only because of the exhaustion of many classical deposits mentioned by ancient authors (mainly on the Greek Isles and in Asia Minor), but also because of the blocking of trade routes due to the expansion of the Turkish (Ottoman) Empire in Agricola's time. Figures 1 and 2 give an overview of the deposits and occurrences of clays and other argillaceous raw materials mentioned by Agricola. Apart from the information taken over directly from ancient authors, the preponderance of Central European sites is obvious.

SUMMARY AND COMPARATIVE DISCUSSION

The prehistory of clay mineralogy contains everything that has been part of the mineralogical tradition concerning earths and argillaceous raw materials before the establishment of clay mineralogy as a scientific discipline (which may be dated at best into the 19th century). We have recalled some of the most prominent early statements, with special regard to Theophrastus, Vitruvius, Pliny, Isidore and Agricola. To the best of our knowledge, Isidore of Seville has never been taken into account in any historical sketch of the mineralogical sciences, probably partly because complete editions of Isidore's works have not been easily accessible and partly because the title of his encyclopaedia, *Etymologiae*, is not appropriate to draw the attention of scientists and engineers. A simplified picture of the tradition is as follows: With his small book *De lapidibus* Theophrastus may be considered as the earliest founder of mineralogy as a science and represents the pure Aristotelian (peripatetic) tradition, which is later fully assimilated, elaborated and stringently refined by Agricola. Vitruvius (*De architectura*) gives a few original contributions (e.g. concerning building materials, ceramic processing, and the powder of Puzzuolo), most of which are later integrated in Pliny's encyclopaedic compliation *Naturalis historia*, the most influencial work on natural sciences until the end of the Middle Ages, the main factographical source of Agricola's *De natura fossilium* and without doubt also one of the primary sources for Isidore. With respect to earths and clays Agricola cites Theophrastus, Vitruvius, Columella, Dioscorides, Pliny, Galen, Avicenna and a few others, but not Isidore, although he praises Pliny for giving "credit openly and frankly to those whose writings he uses" and assuring that likewise he will "give credit by name to those whom I quote".

There are many analogies in the mineral systems of Isidore and Agricola. The degree of similarity is higher than between any other mineral system before Agricola (Aristotle/Theophrastus, Avicenna, Albert

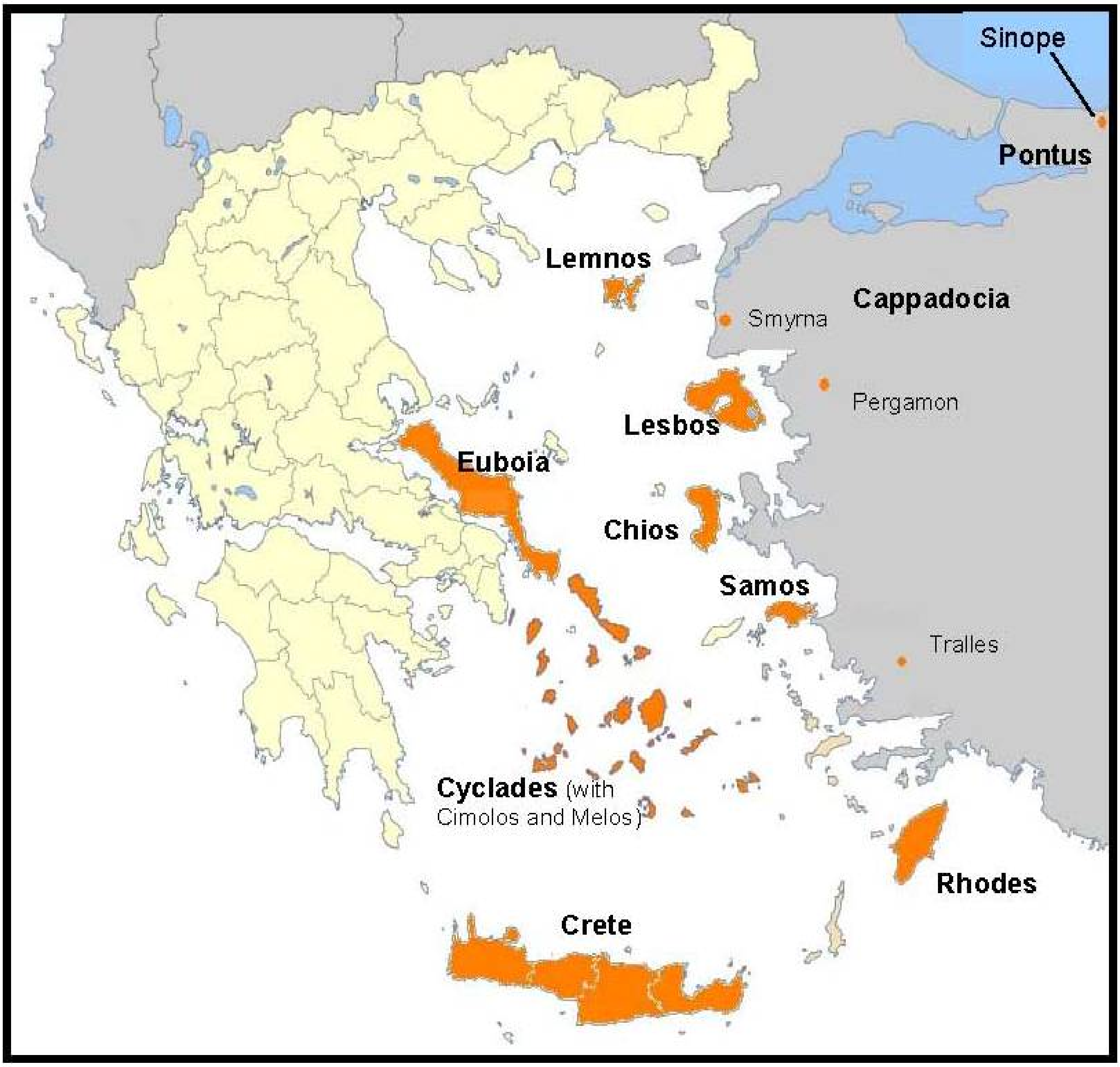

Fig. 1 Deposits and occurrences of earths (clays and other argillaceous raw materials) on the Greek Isles, Crete and Asia Minor, according to Agricola; virtually all information on these sites is directly taken from ancient authors – Agricola never saw these places (Kořánová, 2008).

the Great), see (Prescher, 1958; Prescher and Quellmalz, 1994; Krafft, 2006). Table 1 may illustrate this.

Thus, despite a few significant differences (e.g. glass is an individual class only for Isidore, and the abstract class "mixtures and composites" was introduced by Agricola for the first time), it is evident that Agricola'a mineralogical classification is so closely related to Isidore's, that it is hardly believable to be independent. Of course, Isidore was at best an encyclopaedist, whereas Agricola was a specialist in mineralogy. It is therefore clear that Agricola's work is incomparably more detailed and gives a much more complete account of the previous work of other authors. Nevertheless, the basic idea of this systematic classification, in particular the introduction of "earths" as a separate class (which does not occur in other authors' work before Agricola), must be attributed to Isidore, not to Agricola. Nevertheless, it must be assumed that Agricola did not know Isidore's work directly. His own statements in his works (e.g. the one in the preface of *De Natura Fossilium*, in which he praises Pliny for "giving credit to" his sources) indicate that Agricola truly attempted to cite ("by name") all those whose opinions he adopts. There is no hint of hidden plagiarism in any of Agricola's works. Of course, an indirect influence cannot be excluded – either Isidore's ideas were present in oral late-mediaval educational tradition or Agricola was informed about them by one of his humanist

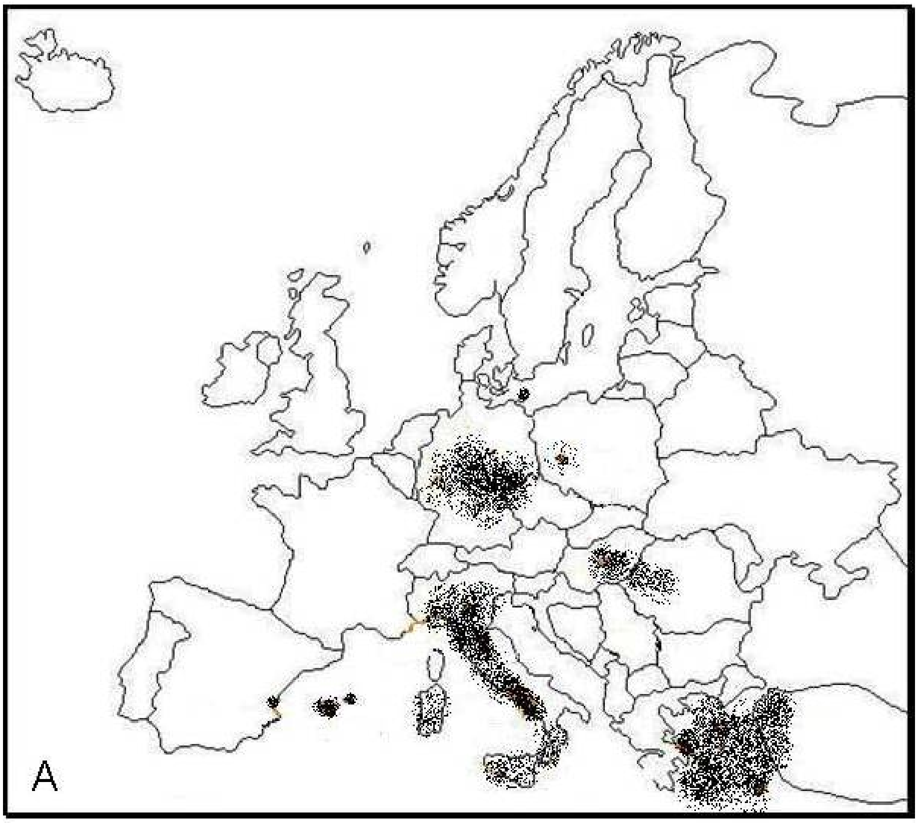

Fig. 2 Deposits and occurrences of earths (clays and other argillaceous raw materials) in Europe (a), mainly Central Europe, essentially Germany and Bohemia (b), according to Agricola; virtually all information on these latter sites is original and has never occurred in the literature before – Central Europe was the main stage of Agricola's life (Kořánová, 2008).

Table 1 Comparison of the sixteenth book of Isidore's Etymologies (written between approx. 620 and 636 A.D.) and Agricola's De natura fossilium (1546).

Isidore	Agricola
Etymologiae liber XVI (~ 620-636 A.D.)	De natura fossilium libri X (1546)
De pulveribus et glebis terrae (powders and lumps of earths) – chapter 1	Terrae (Earths) – book 2
De glebis ex aqua (solids originating from liquids) – chapter 2	Succi concreti ("congealed juices"): macri (meagre) – book 3, pingues (unctuous) – book 4
De lapidibus (stones): vulgaribus (common stones) – chapter 3, insignioribus (rarer stones) – chapter 4	Lapides (stones) – book 5
De gemmis (precious stones) – chapters 6 – 15 *(general introduction and individual "species" according to their color – green, red, purple, white, black, multicolor, crystal-clear, fire-red, golden)*	Gemmae (precious stones) – book
De marmoribus (marbles) – chapter 5	Marmora (marbles) & saxa (rocks) – book 7
De vitro (glass) – chapter 16	-
De metallis (metals) – chapters 17–24 *(introduction and 7 individual species – gold, silver, copper (and bronze), iron, plumbum (album & nigrum, i.e. tin & lead), stagnum (probably a silver-lead alloy), electrum (a gold-silver alloy))*	Metalla (metals) – book 8 *(seven metals – gold, silver, copper, iron, tin, lead, mercury, bismuth)* et rei metallicae (alloys, slags and artificial metallurgical products) – book 9
-	Mista et composita (mixtures and composites) – book 10
De ponderibus (weight measures)	*Treated by Agricola in his metrological works (1533 and 1550).*
De mensuris (volume measures)	
De signis (measure symbols and labels)	

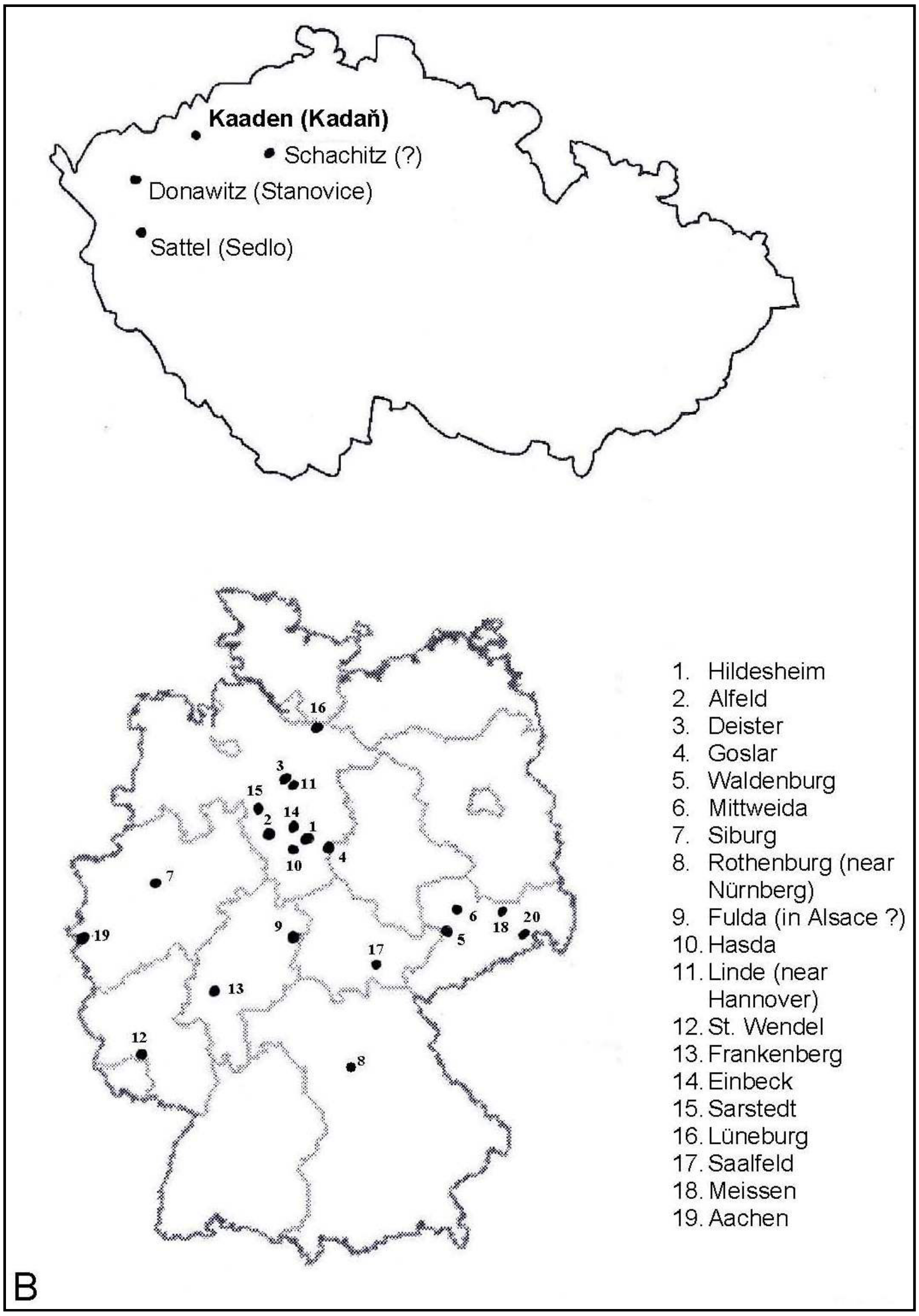

Fig. 2 - continued

contemporaries. Nevertheless, since the non-accessibility of Isidore's work must definitely be excluded (Möller, 2008), the final reason, why Agricola did not appear to be aware of Isidore's work, seems to be a philosophical one (reluctance to accept medieval sources, especially those written by theologians). Isidore was the last western "Father of the Church". He was considered as a popular (and by the official doctrines acknowledged) author and as a main source for early medieval, and partly also scholastic, natural philosophy (Pabst, 1994). Therefore renaissance humanists must have considered his eclectic work with suspicion (Möller, 2008). As an official clerical author (and more than that, as a Saint) from the Dark Ages he could certainly not be en vogue in the new back-to-the-roots movement, which sought to rediscover and restore – without prejudices and unimpressed by authority – the knowledge of Antiquity. An alternative explanation for the evident similarity of Isidore's and Agricola's system may be that both authors used Pliny's *Naturalis historia* as their main factographical source, and that their systematization is indeed an independent development.

Neither Isidore nor Agricola give a general definition of “clay“ or even “earth“. In contrast to Agricola, Isidore makes at least an attempt to clearly distinguish between powders (i.e. systems with freely flowing earth particles) and lumps of earth (i.e. cohesive masses of earth particles). As a consequence of his emphasis on the powder character of earths, Isidore takes also sulphur into this class (for Agricola an “unctuous congealed juice”). Similarly, the “powder from Puzzuolo” is classified by Isidore as an earth (for Agricola not a “single earth” / *concretum ex partibus sui similibus substancia*, but a *compositum / concretum ex partibus sui dissimilibus substancia*, i.e. a mixture of earth with alum, sulphur and / or bitumen). The distinction between argillaceous earths (i.e. clays) and non-argillaceous earths, in particular chalk (creta), was not clear for either of them. The combinatorical classification scheme of earths into 81 (theoretical) species has been proposed by Agricola for the first time. It is, however, based on four property complexes (“meagre-unctuous”, “porous-dense”, “soft-hard”, “smooth-rough”) with qualities of three degrees that are firmly based on Aristotelian tradition and go back to Theophrastus’ *De lapidibus*. Although certain technological characteristics are implied by these “properties” (e.g. “unctuous, dense and soft” imply high plasticity in combination with water, while “meagre, porous and rough” indicate a high content of non-clay minerals or grog / flux particles, using today’s ceramic terminology) the selected properties are in fact to a large degree interrelated, and Agricola has to invoke all his argumentational wit to justify this basic set of properties, which is actually dictated by (peripatetic) tradition. As a consequence, from a modern point of view, Agricola’s seemingly “rational” approach to systematisation and classification loses a lot of its scientific value. With respect to properties it must be taken into account, however, that clays and clay minerals were particularly difficult to classify, so that significant progress in that area had to wait for several centuries. In fact, only when chemistry had become a discipline with well-established analytical methods, it was possible to determine the composition of clays (19th century), and only the availability of X-ray diffraction made structural investigation possible (20th century). Although today the classification of clay minerals and clays is based on chemical composition and crystallographical structure, technological parameters characterizing the rheological behavior of clay-water mixtures and the complex behavior during low- and high-temperature heat treatment (drying and firing) are as important today as they were in Agricola’s time. It is only in the last decades that the qualitative, rough and empirical approach in this area is being replaced by quantitative, precise and standardized measurements. This, of course, is dictated by the necessity to optimize processing and raw materials consumption according to economical and ecological demands, and by the growing insight that natural raw material resources of appropriate quality, even of such apparently common matter as clays and earths, are not unlimited.

Agricola’s main accomplishments in the field of mineralogy (in general, and in particular concerning earths and clays) are the careful compilation of the knowledge of Antiquity, the general emphasis on properties and applications, and the beginning of mapping deposits and occurrences of raw materials and minerals (including earths and clays) in Central Europe. The latter has become a practical necessity due to the fact that many of the classical deposits (e.g. the famous silver mines of Laurion in Attica and many clay deposits on the Greek Isles) were exhausted or the blocking of trade channels due to the expansion of the Turkish (Ottoman) Empire in Agricola’s time prohibited their use. Due to his medical education, philological experience and special professional background Agricola was able to take into account also the medical authors Hippocrates, Galen and Dioscorides, which have been neglected before in the Latin natural science tradition. As a professional physician and pharmacist Agricola felt that in the absence of original ingredients the recipes traded by the ancient authors would become useless. This explains his general effort in collecting and publishing information on the known occurrences and new deposits of analogous or similar raw materials in Central Europe, in particular Germany and Bohemia, the main stage of his life. At the same time it was necessary, of course, to cope with the confusion between ancient Greek or Roman and current Central European weights and measures (this was the impetus for Agricola’s metrological works and his struggle for a unification of the metrological system). Of course, for Agricola a well-founded knowledge of mineral properties was the basic precondition to identify mineral raw materials and find substitutes. Already in his *Bermannus* (1530) he criticizes the physicians of his time for blindly citing dead literal science without understanding the contents (which in the field of mineralogy was indeed a problem at that time), see (Agricola, 1957); on the other hand, in *De re metallica* (1556) he sketches the ideal figure of a miner as a man of universal erudition (Agricola, 1994, 2007). These are two poles of Agricola‘s humanist educational vision: theoretical knowledge is of value only when it can be correctly applied, and men of practice have to be theoretically educated over and above the immediate practical needs. Agricola showed that earths (and clays) were part of the scientific tradition of the western world. At the same time he was interested in their different applications and certainly inclined to use them for medical purposes whenever feasible (last but not least as medicaments against the pest, which was threatening many regions of Central Europe during Agricola’s lifetime (Engewald, 1994)). This connection of careful philological work with the purpose of matching urgent practical needs of his time made Agricola's work, which in its completeness stood absolutely unrivalled in his time and for centuries to come, a foundation on which future generations of mineralogists (and clay scientists) could build.

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