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James Budahn

Anna Avila

Gary Skipp

Joshua Freeman

See next page for additional authors

Authors

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The role of African dust in the formation of Quaternary soils on Mallorca, Spain and implications for the genesis of Red Mediterranean soils

Daniel R. Muhs[a,*], James Budahn[a], Anna Avila[b], Gary Skipp[a], Joshua Freeman[a], DeAnna Patterson[c]

[a] U.S. Geological Survey, MS 980, Box 25046, Federal Center, Denver, CO 80225, USA
[b] Centre de Recerca Ecològica i Aplicacions Forestals, Universitat Autònoma de Barcelona, Bellaterra, Spain
[c] ATA Services, Inc., 165 South Union Blvd., Suite 350, Lakewood, CO 80228, USA

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ABSTRACT

African dust additions explain the origin of terra rossa soils that are common on the carbonate-platform island of Mallorca, Spain. Mineralogical and geochemical analyses indicate that Quaternary carbonate eolianites on Mallorca have a very high purity, usually composed of more than 90% carbonate minerals (calcite, dolomite, and aragonite). In contrast, terra rossa soils developed on these eolianites have lower carbonate contents and contain higher concentrations of quartz and other silicates. Analyses of immobile trace elements indicate that the non-carbonate fractions of the eolianites have distinctive Zr/Hf, La/Yb, Cr/Sc and Th/Ta values that differ from the superjacent terra rossa soils. These observations indicate that even if sufficient dissolution of the eolianite had taken place to create the soils by residual accumulation, immobile element ratios in the soils require an external source. However, Zr/Hf, La/Yb, Cr/Sc and Th/Ta values in the soils fall within the range of values for these element ratios in African dust collected on Barbados and mainland Spain. We conclude that the silicate fractions of terra rossa soils on Mallorca are derived mainly, though not wholly, from far-traveled African dust, and this process may explain the origin of other terra rossa soils found in southern Europe.

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1. Introduction

There has been an increasing awareness in the Earth and atmospheric science communities of the importance of mineral dust. Kohfeld and Tegen (2007) summarize several potentially important impacts of mineral dust on the Earth–atmosphere system. As a component in the atmosphere, dust can change the overall planetary radiation balance through direct radiative effects at both solar (shortwave) and terrestrial (longwave) portions of the spectrum. Indeed, mineral dust was explicitly identified in the 2007 IPCC report as an important component in the global radiation balance (Forster et al., 2007). Phytoplankton blooms that occur after dust-derived Fe fertilization in the oceans can result in significant carbon dioxide drawdown from the atmosphere, creating a reverse greenhouse effect. In addition, dust can have important effects on the biogeochemical cycle of terrestrial ecosystems, adding nutrients to soils and the vegetation they support. Kohfeld and Tegen (2007) also point out that dust can have a significant effect on soil genesis. Numerous recent studies have now documented evidence for far-traveled dust additions to soils (e.g., Birkeland, 1999; Kurtz et al., 2001; Muhs et al., 2007a,b, 2008).

The most important sources of mineral dust in the world today are the Sahara and Sahel regions of Africa (Goudie and Middleton, 2001, 2006; Prospero et al., 2002; Mahowald et al., 2006). Although much dust from Africa is transported westward to the Atlantic Ocean (Prospero et al., 1970; Prospero and Lamb, 2003), a significant portion is transported northward, to the Mediterranean Sea and beyond (see papers in Guerzoni and Chester, 1996). Earth-orbiting satellites have documented that African dust transport is both common and widespread (Figs. 1 and 2). African dust inputs are a significant component of the overall biogeochemical budget in both the Mediterranean Sea (Guerzoni et al., 1999) and southern European ecosystems (Avila et al., 1997, 1998, 2007). Modeling studies by Mahowald et al. (2006) show that while the flux of dust from Africa to Europe during the present interglacial period is significant, it was likely even higher during the Last Glacial period, about 12,000–25,000 yr ago (Fig. 3). Recently, Stuut et al. (2009) presented a hypothetical model of dust transport from Africa to southern Europe within the context of soil zones. They proposed three latitudinal depositional (D) zones wherein dust influences on soils could be identified, going from south to north: D1a, a southern zone where there are discrete soil layers that are

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* Corresponding author. Tel.: +1 303 236 7919.
E-mail address: dmuhs@usgs.gov (D.R. Muhs).

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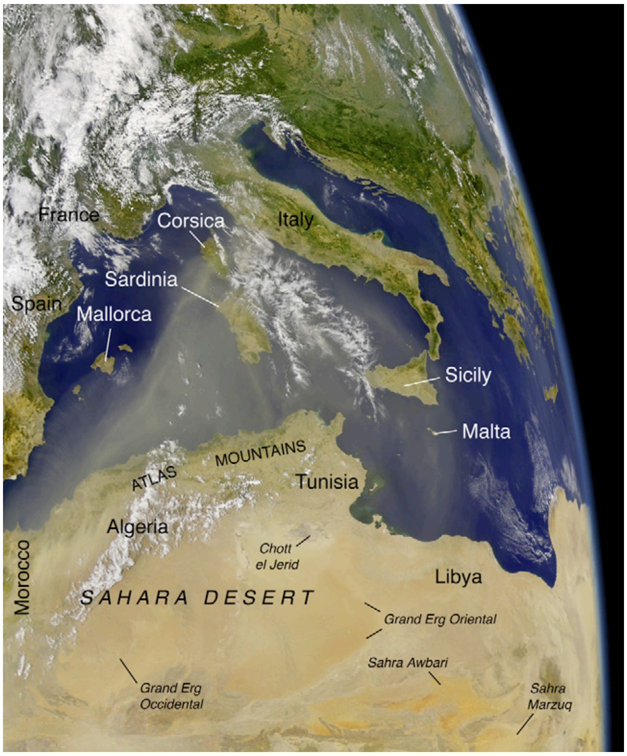

Fig. 1. True-color image showing Saharan dust transport from northern Africa to southern Europe on 19 August 2004. Sea-viewing Wide Field-of-view Sensor (SeaWiFS) image taken aboard the OrbView-2 satellite, courtesy of the SeaWiFS Project of NASA/Goddard Space Flight Center.

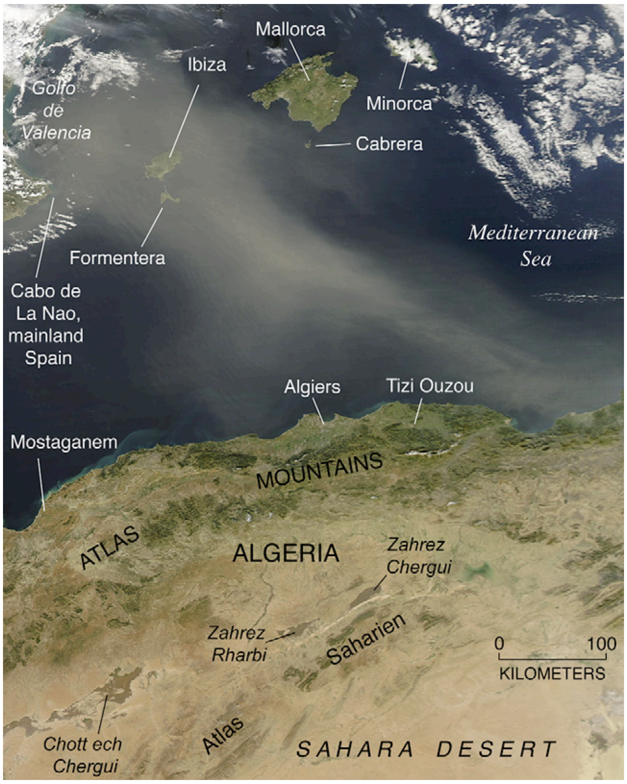

Fig. 2. True-color image showing transport of dust from Algeria to the western Balearic Islands (Ibiza and Formentera) and mainland Spain on 6 December 2003. MODIS image taken from the Aqua satellite, courtesy of Jeff Schmaltz of the MODIS Rapid Response Team, NASA/Goddard Space Flight Center.

eolian-derived; D1b, an intermediate-latitude zone where eolian inputs occur as soil admixtures; and D1c, a northern zone where eolian inputs occur as "fugitive clouds", and identification of eolian inputs to soils presumably would be difficult. To some extent, this latitudinal zonation parallels that of modeled dust fluxes from Africa to Europe presented by Mahowald et al. (2006) and shown in Fig. 3a.

Dust transport from North Africa to the Mediterranean and southern Europe potentially can come from a variety of sources. Gillette (1999) points out that important dust sources in deserts are often concentrated as "hot spots" of particularly favorable conditions for eolian entrainment. These conditions include a lack of vegetation, a source of sand (for saltation-induced impact on finer particles), a long fetch, smooth geomorphic surfaces, disturbed sediment (i.e., loose particles), thick deposits (i.e., no supply limitations), topography with converging winds, dry and uncrusted surfaces, and strong mesoscale winds. Based on satellite imagery such as that shown in Figs. 1 and 2, much of the dust reaching the western Mediterranean and southwestern Europe comes from the Sahara Desert portions of Algeria and Tunisia, south of the Atlas Mountains (Fig. 1). Prospero et al. (2002), using the Nimbus 7 Total Ozone Mapping Spectrometer, confirm that this region is a consistent, year-to-year dust source. They hypothesize that fluvial and playa ("chott") deposits are potential sources of dust in this area.

Our own examination of sedimentary environments in the northern Sahara Desert of Tunisia and adjacent eastern Algeria supports the hypotheses of Gillette (1999) and Prospero et al. (2002). Potential dust sources that we have observed in the field in the Sahara (described in more detail in Ori et al., 2009) consist of dry washes or wadis ("oueds") such as the Oued el Khanga and its tributaries, which drain an enormous part of the Atlas Mountains of Algeria and Tunisia (Fig. 4a). In addition to potential deflation from the loose, dry sediments of its broad channel system (Fig. 4b), south-draining rivers such as the Oued el Khanga deposit sediments onto large alluvial fans to the south of the mountains, with the finest particles ultimately reaching playas, such as the Chott el Gharsa (Fig. 4a). Older Neogene sediments, rich in clays, are currently being eroded by the wind, as evidenced by extensive fields of yardangs (Fig. 4c). Such sediments are also eroded by fluvial/colluvial processes into playas where, if not cemented by crusts, they can be entrained by the wind (Fig. 4d). The Grand Erg Oriental is one of North Africa's most extensive dune fields (Fig. 4e). Crouvi et al. (2008) have shown how eolian abrasion of sand-sized particles in dune fields can generate silt-sized particles that accumulate downwind as loess. In addition, sand from this dune field and other sources can be transported over playa surfaces, such as the Chott el Jerid (Fig. 4f). Saltating sand grains on playas can dislodge and entrain finer-grained particles by impact, allowing them to be transported farther by wind in suspension.

The conceptual models outlined above, along with observational data of dust transport northward from Africa, suggest that southern European soils could be influenced to one degree or another by eolian inputs. In this paper, we test this hypothesis by studying soils and paleosols collected on the island of Mallorca, the largest of the Balearic Islands off the coast of mainland Spain (Fig. 5). Mallorca is well within the zone of northward transport of African dust to southern Europe, based on back-trajectory analyses (Avila et al., 1997, 2007) and examination of satellite imagery (Figs. 1 and 2). Dust additions during rain events on Mallorca have been well documented (Fiol et al., 2005). In addition, because Mallorca is a carbonate-dominated island (Fig. 6), local sources of silicate-rich soil parent material are limited, making it easier to identify exotic components that reflect dust inputs. As pointed out by Yaalon and

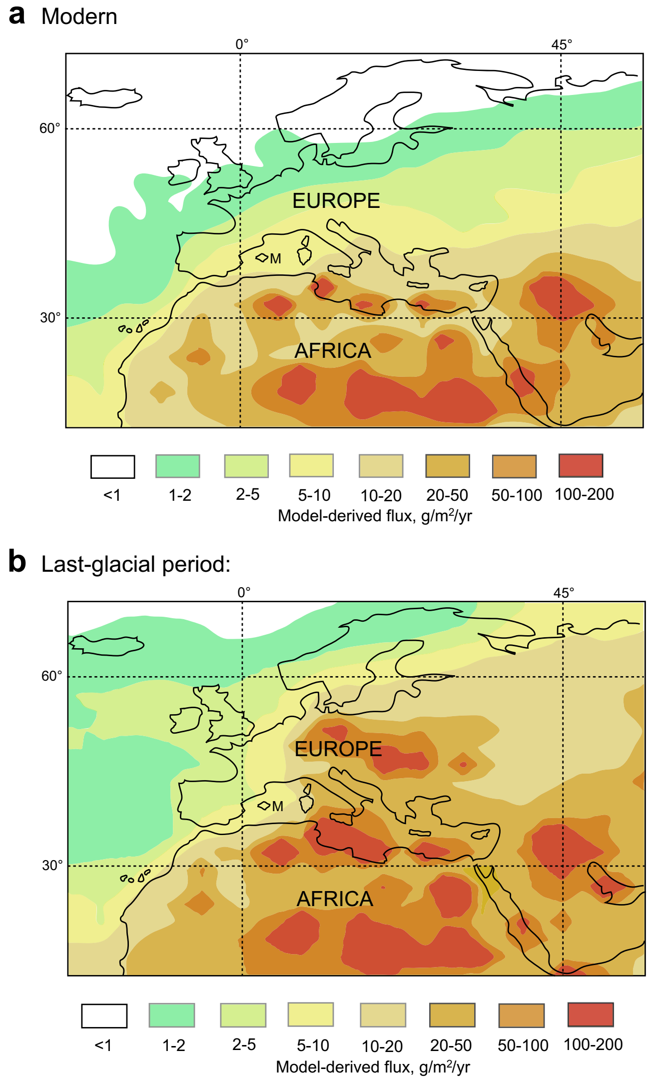

Fig. 3. Model-derived dust flux in northern Africa and southern Europe for (a) modern climatic conditions, and (b) during the Last Glacial period, the latter tuned to the geologic record of dust deposition. Redrawn from Mahowald et al. (2006), their Figs. 2 and 10.

Ganor (1973), Macleod (1980), Jackson et al. (1982), Rapp (1984), Yaalon (1987), and Stuut et al. (2009), study of soils on carbonate terrains in southern Europe, including Mallorca, leads to the issue of dust inputs to the widespread "terra rossa" soils of southern Europe (Fig. 5). The origin of these Mediterranean soils and their relation to dust inputs have been controversial topics, which we review here.

2. African dust to Europe and the "terra rossa problem"

Terra rossa soils are red (2.5YR, 10R) or reddish brown to reddish yellow (5 YR, 7.5 YR), clay-rich soils on carbonate substrates (limestone or dolomite). Commonly, terra rossa soils have sharp contacts with the underlying limestone or dolomite and often fill solution cavities within the carbonate bedrock. They have highly variable thicknesses, depending on landscape position, with thicker soils typically found in low-lying portions of the landscape.

Terra rossa soils are found along many of the continental shores and islands of the Mediterranean basin (Fig. 5), the classical region in which they have been studied. (Kubiëna, 1953; Rapp, 1984; Yaalon, 1997; Durn, 2003). They are also found in parts of Australia, particularly the Mediterranean zone of South Australia (Stace et al., 1968). Although they are typically associated with Mediterranean climates, they have also been described in continuously humid climates in the Western Hemisphere along an irregular latitudinal distribution from Wisconsin (Frolking et al., 1983) and Indiana (Olson et al., 1980) south to Bermuda (Ruhe et al., 1961) and Jamaica (Muhs and Budahn, 2009).

In earlier FAO–UNESCO soil maps of Europe (Dudal et al., 1966), terra rossa soils were classified and mapped primarily as "Red Mediterranean soils" or, in many parts of Spain, as "Reddish Brown Soils" (Fig. 5). Whereas Red Mediterranean soils have well-developed, red, argillic B horizons, Reddish Brown soils may or may not have a B horizon. Later FAO–UNESCO soil classification schemes have eliminated terra rossa soils as a discrete mapping unit, as they fall into a number of new soil taxa, including Cambisols, Luvisols, and Leptosols (in the U.S. Soil Taxonomy, these are Inceptisols, Alfisols, and Entisols), depending on profile thickness, diagnostic horizons and other taxonomic criteria. Nevertheless, some national soil classification systems (Croatia, Italy, and Israel) have retained "terra rossa" as a mapping unit (Yaalon, 1997; Durn, 2003).

The origin of these visually striking soils has been the subject of debate for decades. Durn (2003), Schaetzl and Anderson (2005), and Singer (2007) review the possible modes of origin that have been proposed for terra rossa soils. Singer (2007) distills the concepts into three theories for their origin: (1) the "residue theory," where such soils are derived from residual accumulation of the non-carbonate fraction of the host bedrock; (2) the "ascending sesquioxide theory," where Fe- and Al-hydroxides build up in the soil by capillary ascent from the bedrock; and (3) the "allochtonic accretion theory," where most, if not all, of the soil builds up from outside sediment sources, most commonly thought to be from fine-grained eolian accretion.

By far, the greatest number of terra rossa studies fall into tests of either the "residue theory" or the "allochtonic theory" categories. Residue-theory advocates include Reifenberg (1947), Dudal et al. (1966) and Stace et al. (1968), who simply state that terra rossa soils formed from the host rock, and Barshad et al. (1956) and Moresi and Mongelli (1988), who use mineralogical data to support their claims for a residue origin in Israel and Italy, respectively. Glazovskaya and Parfenova (1974) thought terra rossa soils in the Crimean area of Ukraine formed from residue, but also suggest that some developed by slopewash additions from limestone terrains that are upslope of terra rossa soils. Although most residue-theory advocates were among the earlier investigators of terra rossa soil origins, some adherents remain today. As recently as 1997, Bronger and Bruhn-Lobin (1997, p. 279) stated: "The development of Terrae rossae from material with high carbonate content can result only by carbonate dissolution and accumulation of limestone residue (LR)." Later in the same paper, however, Bronger and Bruhn-Lobin (1997, p. 285) also seem to accept the possibility of eolian additions to terra rossa soils that they studied in Morocco. Schaetzl and Anderson (2005) point out that there are numerous problems with the "residue theory" of terra rossa origin. These include sharp contacts between soil and rock, more mass in the soil profile than can be reasonably accounted for by residual accumulation from bedrock (i.e., a residual accumulation that would require unrealistic amounts of carbonate rock dissolution), particle size differences between soil and bedrock residue, and mineralogical mismatches between soil and bedrock residue.

Supporters of the allochtonic accretion theory have tended to cite eolian particle additions (as opposed to alluvial or colluvial additions) as the primary parent material for terra rossa soils (Yaalon and Ganor, 1973; Rapp, 1984; Yaalon, 1987, 1997). An eolian

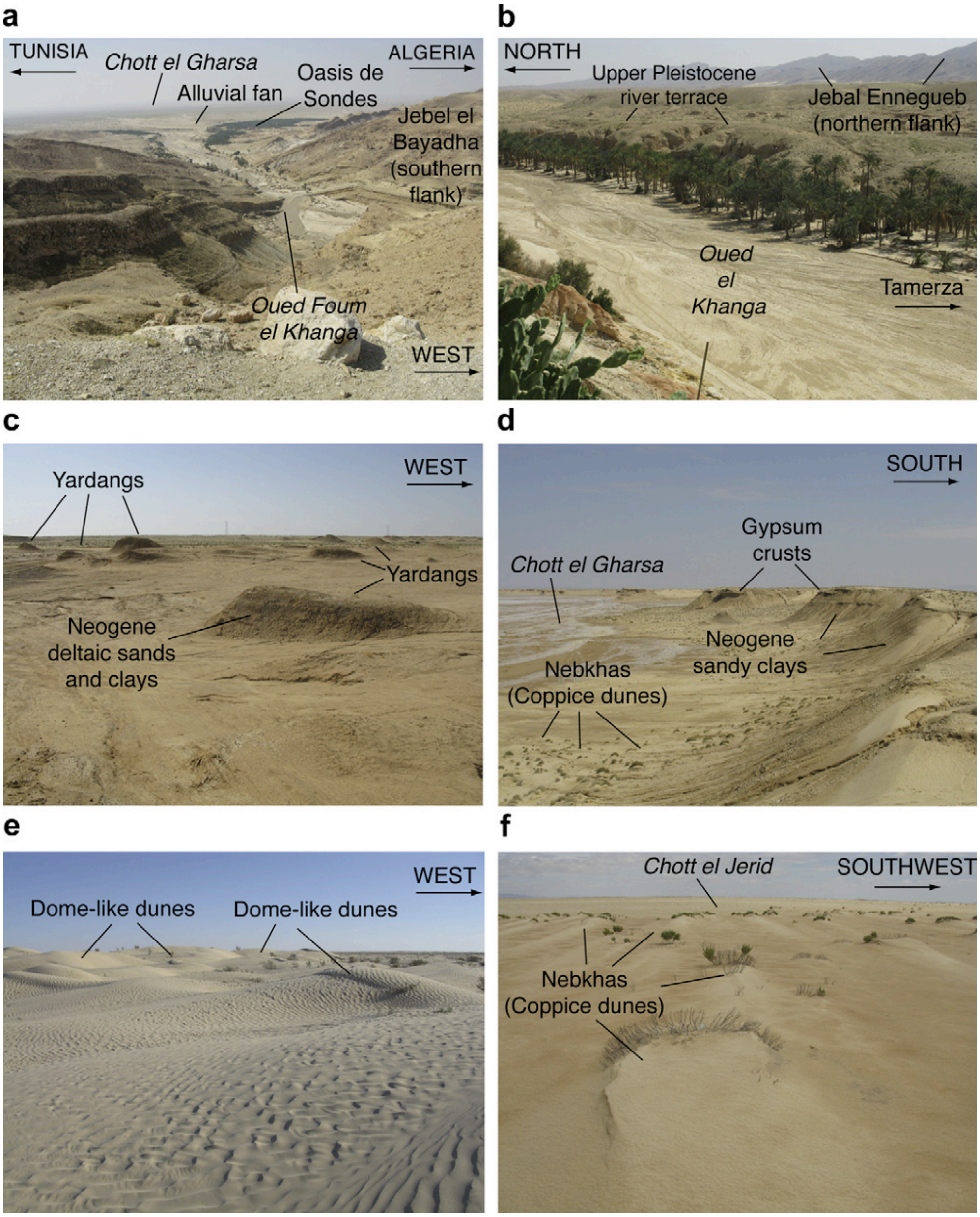

Fig. 4. Possible sources of long-range-transported dust to the Mediterranean and southern Europe from the northern Sahara Desert of Tunisia and adjacent eastern Algeria: (a) fine-grained sediments in dry wash or wadi ("oued") and alluvial fan of Oued el Kranga and Oued Sendess, which drain the Atlas Mountains of Tunisa and Algeria; (b) fine-grained sediments of Oued el Kranga, upstream of (a), near Tamerza, Tunisia; (c) Fine-grained Neogene sediments being eroded into yardangs, northwest of Tozeur, Tunisia; (d) Fine-grained playa ("chott") sediments in Chott el Gharsa, derived from sandy clays of Neogene age, northeast of Tozeur, Tunisia; (e) northernmost part of Grand Erg Oriental (see Fig. 1), ~15 km west of Douz, Tunisia; (f) Fine-grained sediments and coppice dunes in Chott el Jerid (see Fig. 1), ~70 km northwest of Kebili, Tunisia. All photographs by D.R. Muhs.

origin for particles in terra rossa soils has been inferred using a wide variety of analytical tools. Jackson et al. (1982) utilize oxygen isotopes in quartz to support an eolian origin for much of the mass in terra rossa soils of Italy. MacLeod (1980) uses low silicate residue contents in carbonate bedrock and particle size data to infer an eolian origin for terra rossa soils in Greece. Durn et al. (1999) employ particle size analyses, geochemistry and mineralogy to show that the host limestone is not the parent material for terra rossa soils in Croatia and suggest eolian additions as a mechanism for at least part of their genesis. Genova et al. (2001) studied terra rossa soils in Sardinia and conclude from trace element geochemistry that soils must have had eolian and/or alluvial additions from outside the host carbonate rock.

A number of investigators note that the various theories of terra rossa genesis are not mutually exclusive. Kubiëna (1953, p. 214) acknowledges the possibility of fine particle enrichment in terra rossa soils by *both* residue accumulation and eolian additions. Duchaufour (1982, p. 381) states that terra rossa forms by carbonate rock dissolution, but also points out that eolian contamination by wind-blown silt is "very common." Pye (1992) studying red soils on limestone on Crete, reports that although an eolian component is important, in situ weathering of the limestone is actually a greater contributor to the soils. Nihlén and Olsson (1995), also studying terra rossa soils on the island of Crete, use oxygen isotopes in quartz in an approach similar to Jackson et al. (1982). They report oxygen isotope values in soil quartz similar to

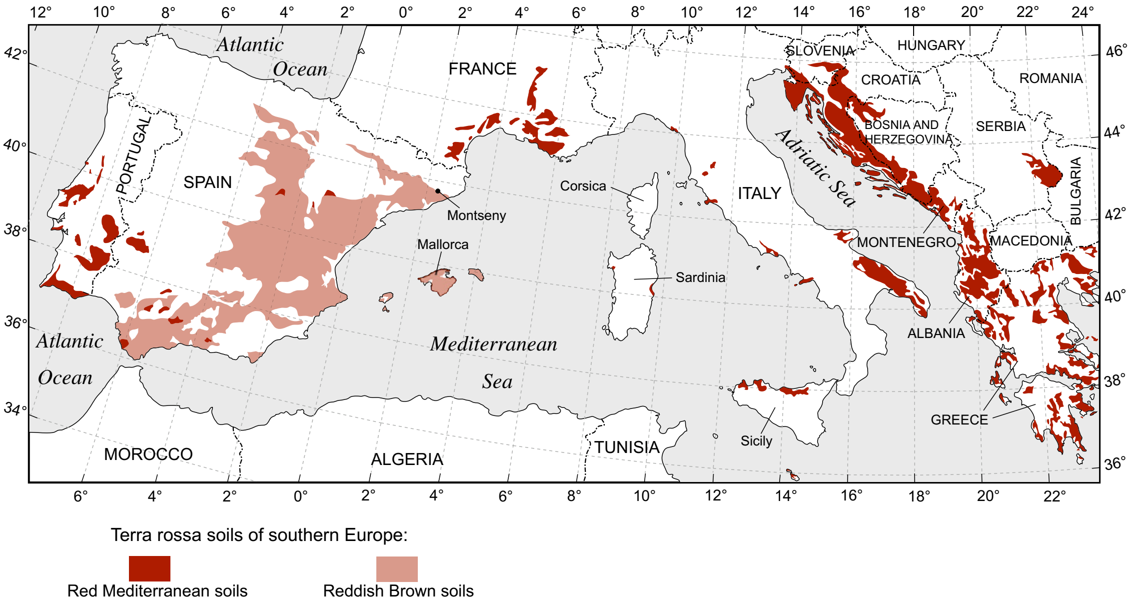

Fig. 5. Map of southern Europe showing the distribution of terra rossa soils (Red Mediterranean and Reddish Brown soils). Redrawn from Dudal et al. (1966).

dust quartz and differing from limestone quartz, but also find that much of the sand fraction in the soils is due to limestone weathering. Delgado et al. (2003) studied terra rossa soils in southern Spain and report mineralogical evidence for both residue and eolian origins for soil particles.

On Mallorca, Dudal et al. (1966) map the modern surface soils primarily as Reddish Brown soils (Fig. 5), but some true terra rossa (Red Mediterranean) soils can also be found and were observed by us in the course of field work. Both Red Mediterranean and Reddish-Brown soils also occur as paleosols in various eolianite

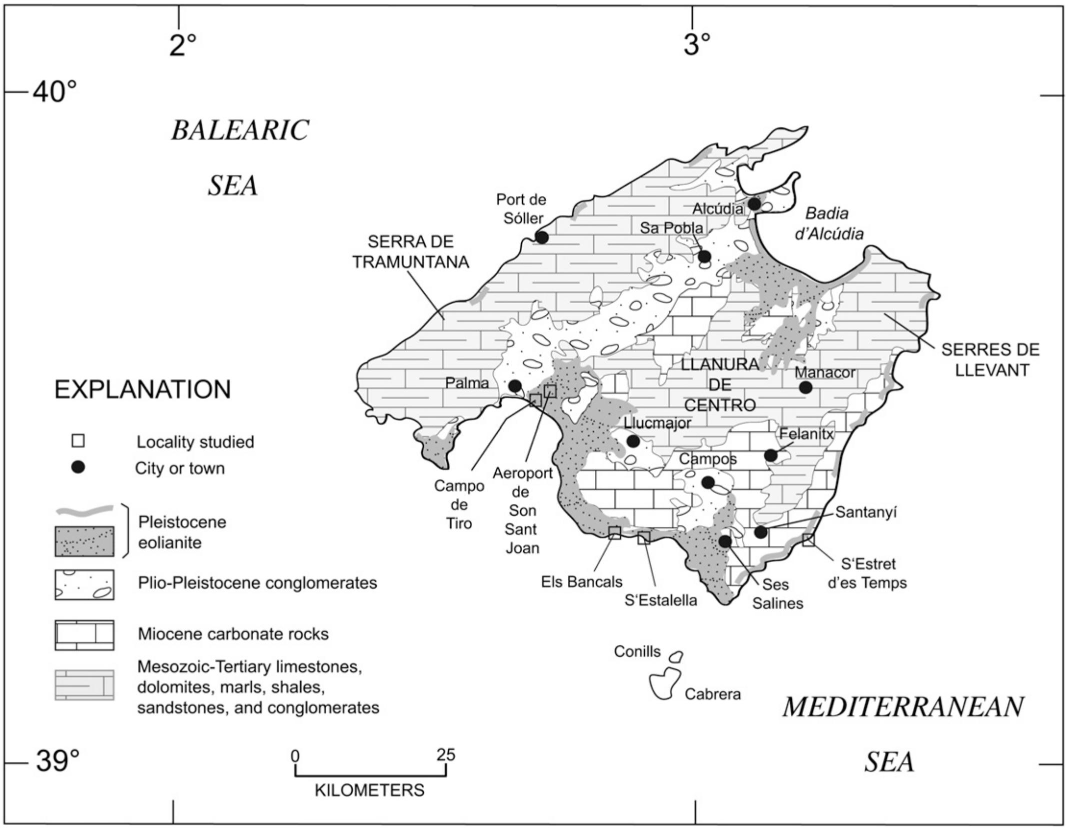

Fig. 6. Geology of Mallorca and location of study sites. Geology simplified from Instituto Geológico y Minero de España (1986, 1987) for bedrock; eolianite distribution simplified from Butzer and Cuerda (1962), González-Hernández et al. (2001), and Fornós et al. (2002).

sections that we studied (Fig. 7). Many of the features that we interpret as paleosols are described as reddish colluvial deposits as well as terra rossa soils (Butzer and Cuerda, 1962; Butzer, 1975; Nielsen et al., 2004). Nevertheless, Butzer and Cuerda (1962) document relatively high silt contents in much of what they describe as colluvial deposits. Silt content is high in modern dust falls that affect Mallorca (Fiol et al., 2005), suggesting that an eolian component is present as silts in the colluvial deposits. Furthermore, although Nielsen et al. (2004) also describe the reddish zones between eolianites on Mallorca as colluvial, they recognize that pedogenesis is an important process in the formation of these features. In addition, Nielsen et al. (2004) hypothesize that eolian silt additions are a part of their origin as well, a hypothesis that is tested here.

3. Bedrock geology of Mallorca

The bedrock geology of Mallorca (Fig. 6) is described by Jenkyns et al. (1990), Gelabert et al. (1992, 2003), and Pomar and Ward (1995). The island has a basin-and-range structure and topography, resulting from Miocene to early Pleistocene extensional faulting (Gelabert et al., 1992; Pomar and Ward, 1995). The mountain ranges (Serra de Tramuntana or Sierra Norte and Serres de Llevant or Sierra de Levante) are horst blocks composed of Mesozoic-to-Tertiary rocks (Gelabert et al., 1992). Another area of upland topography, though of lower elevation, is the Llanura de Centro (Fig. 6). The basins between the southwest-northeast-trending mountains and the Llanura de Centro are floored by Miocene carbonate rocks, primarily reef platforms (Pomar and Ward, 1995) or Pliocene–Pleistocene conglomerates. Overall, much of the island is composed of carbonate rock of Mesozoic or Tertiary age (Instituto Geologico y Minero de España, 1986, 1987). For example, Gelabert et al. (1992) describe the uppermost 2500 m of the Serra de Tramuntana as composed largely of marls, limestones, calcareous breccias, calcarenites, and dolomites, all carbonate rocks, intercalated with thinner beds of conglomerates, sandstones, shales, and gypsum. Similarly, the Serres de Llevant is composed largely of Triassic and Jurassic dolomites, marls, and limestones (Gelabert et al., 2003). Indeed, Rose et al. (1985) estimate that 99% of the island's bedrock is composed of carbonate rocks.

4. Quaternary geology of Mallorca

The Quaternary geology of Mallorca is dominated by deposits that are of alluvial, colluvial, eolian, and marine origin (Butzer and Cuerda, 1962; Butzer, 1975; Cuerda, 1975; Rose et al., 1985, 1999; Hearty et al., 1986; Hearty, 1987; Cuerda and Sacarès, 1992; Hillaire-Marcel et al., 1996; Clemmensen et al., 1997, 2001; Zazo et al., 2003; Nielsen et al., 2004). During interglacial high stands of sea, thin deposits of highly fossiliferous sand and gravel were laid down from what is now near sea level to a few meters above present sea level. These deposits are particularly well exposed in the area just south of Palma (Figs. 8 and 9). Several ages of deposits have been proposed, but many of these age assignments are based, in part, on U-series dating of mollusks (Stearns and Thurber, 1965, 1967; Butzer, 1975; Hearty, 1987; Hillaire-Marcel et al., 1996; Zazo

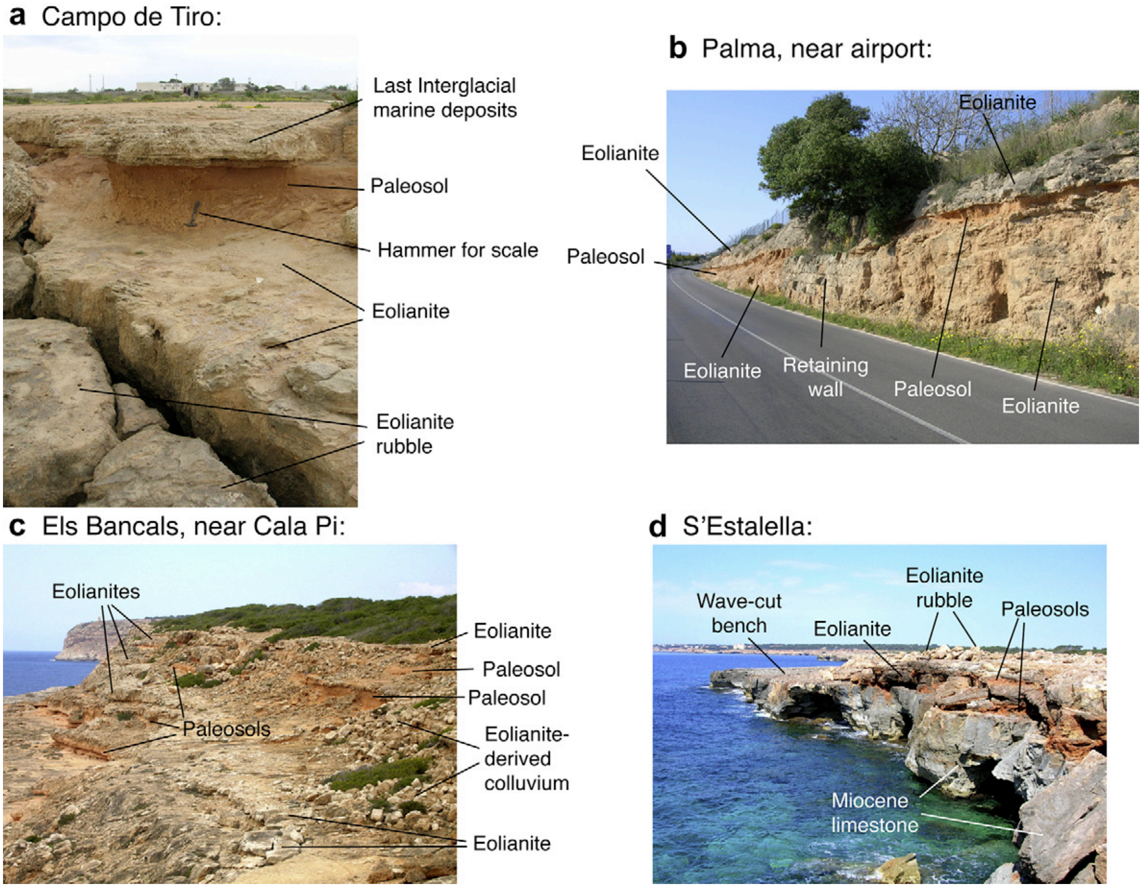

Fig. 7. Soils and paleosols developed on Quaternary eolianites of Mallorca: (a) Campo de Tiro; (b) Palma airport frontage road cut; (c) Els Bancals; (d) S'Estalella. See Fig. 6 for localities. All photographs by D.R. Muhs.

et al., 2003). It has been known for several decades that mollusks are not reliable for U-series geochronology (Kaufman et al., 1971), so these interpretations are uncertain. Instead, Butzer and Cuerda (1962), Butzer (1975), Rose et al. (1985), Hearty et al. (1986), Hearty (1987), and Cuerda and Sacarès (1992) use a blend of elevation data, stratigraphic position, U-series data, aminostratigraphy and paleontological data to infer at least possible correlations of the most recent two sea-level high stands. These are referred to in the older literature as "Tyrrhenian I" and "Tyrrhenian II." Both contain a number of extralimital-southern species of mollusks, notably the Senegalese assemblage that commonly includes the large gastropod *Strombus bubonius*. Tyrrhenian I deposits are thought to date to the penultimate interglacial period (Marine Isotope Stage, or MIS, 7), around 200 ka. Tyrrhenian II deposits are thought to date to the peak of the Last Interglacial period, MIS 5e, around 125 ka.

A single coral U-series age of ~129 ka from Mallorca was reported by Hearty et al. (1986) and supports correlation of Tyrrhenian II deposits to the Last Interglacial period, or MIS 5e. Hearty et al. (1986) and Hearty (1987) use this coral age, amino acid ratios in mollusks, and the extralimital-southern ("Senegalese") faunal aspect of the molluscan assemblages to correlate marine deposits around Mallorca to the Last Interglacial period. Furthermore, Hearty (1987) reports higher amino acid ratios in mollusks at some localities on Mallorca, suggesting marine deposits that date to older interglacial periods.

Whereas low-elevation marine deposits on Mallorca represent interglacial high-sea stands, eolian sands probably represent low stands of sea during glacial periods (Butzer and Cuerda, 1962; Butzer, 1975; Rose et al., 1985, 1999; Clemmensen et al., 1997, 2001; González-Hernández et al., 2001; Fornós et al., 2002; Nielsen et al., 2004). In fact, Butzer (1975) proposes that the Mallorcan Quaternary stratigraphic record can be recognized as a series of continental and marine hemicycles, corresponding to glacial–interglacial cycles. Butzer (1975) considers eolianites to represent glacial periods. Our own observations support the interpretation that eolianites likely were deposited during glacial periods. Eolianites at Els Bancals, S'Estalella, and S'Estret d'es Temps (Fig. 9), as well as other localities on Mallorca, all have high-angle foreset beds that dip landward, yet there is no sand source to build dunes at present. This is a geomorphic setting similar to other island localities, including Puerto Rico (Kaye, 1959), the California Channel Islands (Muhs et al., 2009) and Sardinia (Andreucci et al., in press). As is the case on the California Channel Islands during the Last Glacial period, when sea level was ~120 m lower than present (Fairbanks, 1989; Bard et al., 1990) Mallorca and the other Balearic Islands would have had considerably larger land areas, even to the extent of connecting islands (Fig. 10). Other glacial periods, such as the penultimate glacial period (MIS 6, ~150 ka), probably experienced similar magnitudes of sea level lowering, explaining the origin of older eolianites on Mallorca.

Eolianites on Mallorca have a very high carbonate mineral (calcite, aragonite, dolomite) content, ranging from just over 80% to nearly 100%. This reflects the high carbonate mineral content of modern offshore shelf sands (Fornos and Ahr, 1997) and modern beach sands (Gómez-Pujol et al., 2007) that are analogs to the past sources of eolianites on Mallorca. With the exception of one modern beach on the northwest side of the island, on the flanks of the Serra de Tramuntana, Gómez-Pujol et al. (2007) report that modern beaches around Mallorca have carbonate contents of 88–99%. It is likely, therefore, that former beaches and now-submerged shelves that supplied the source sediments to the Quaternary eolianites were of similarly high carbonate content.

5. Geochemical approach and analytical methods

Our primary method in this study is to use relatively immobile trace elements as "fingerprints" for source materials, following an approach we have used in studying eolian sediments in Alaska, western Atlantic islands, and California (Muhs and Budahn, 2006, 2009; Muhs et al., 2007a,b, 2008). Certain trace elements in rocks, sediments, and soils, such as Sc, Cr, Th, Ta, Zr, and Hf are very

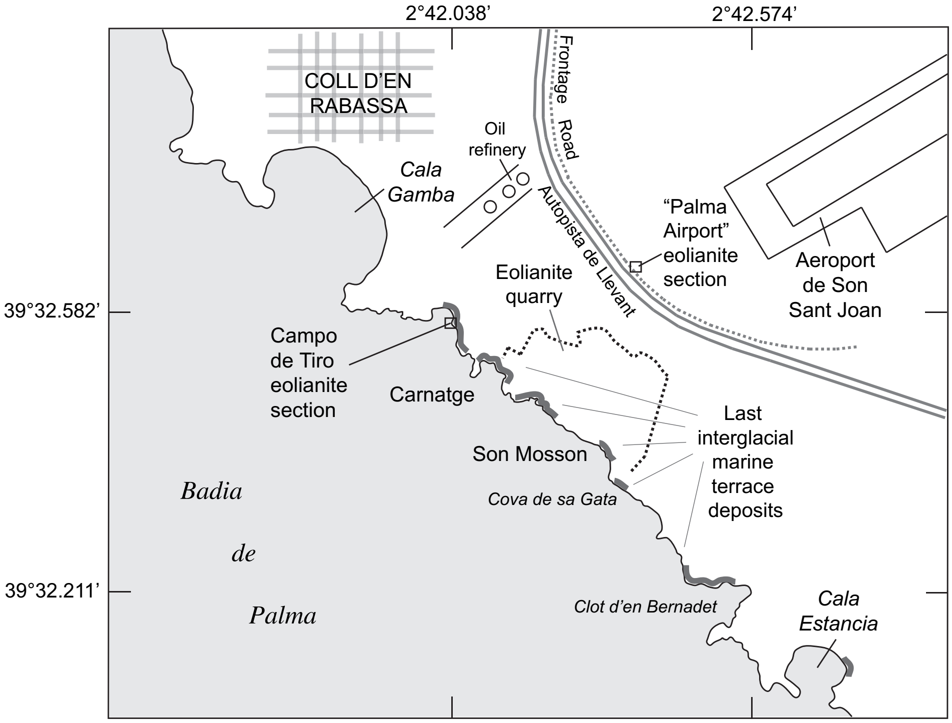

Fig. 8. Distribution of Last Interglacial marine deposits and eolianite sections studied in the Palma area. Marine deposit distribution from Rose et al. (1985).

Fig. 9. Quaternary deposits of Mallorca: (a) eolianites and paleosols at Els Bancals; (b) eolianites at S'Estret d'es Temps; (c) Last Interglacial marine deposits at Campo de Tiro. See Figs. 6 and 8 for localities; all photographs by D.R. Muhs.

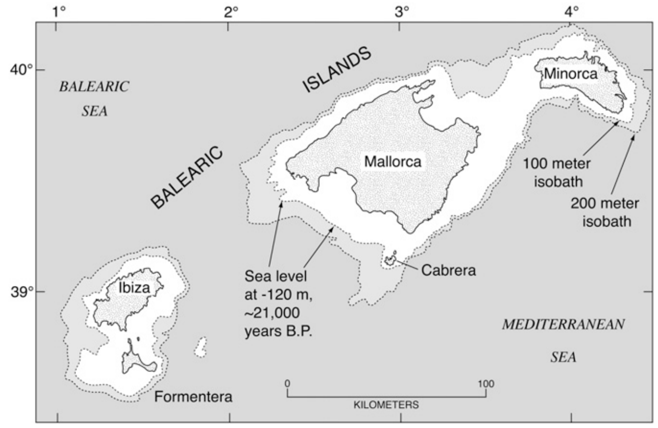

Fig. 10. Mallorca and the other Balearic Islands, showing paleogeography during the Last Glacial period, with a lowered sea level of ∼120 m relative to present (Fairbanks, 1989; Bard et al., 1990).

useful for provenance studies. Suites of these relatively immobile elements, when arrayed on bivariate or ternary plots, show clearly separated compositional fields for sediments derived from oceanic crustal rocks and upper continental crustal rocks (Taylor and McLennan, 1985, 1995; McLennan, 1989). If the particles in soils are locally derived, they should plot within fields similar to the underlying substrate; if externally derived, they may differ from the underlying materials.

The rare earth elements (REE), La through Lu, also can be valuable as provenance indicators. The upper continental crust (UCC) has a distinctive composition with regard to REE (Taylor and McLennan, 1985, 1995; McLennan, 1989). On chondrite-normalized REE plots, sediments from the UCC are characterized by enriched light REE, a negative Eu anomaly, and depleted heavy REE (expressed on plots as a "flat" curve). Loesses and long-range-transported dust from a variety of regions worldwide have a typical UCC composition based on REE abundances (Gallet et al., 1998; Birkeland, 1999; Jahn et al., 2001; Sun, 2002a,b; Muhs and Budahn, 2006; Muhs et al., 2007a; Sun et al., 2007).

Soil samples were collected and subsequently analysed by soil horizon. For bulk mineralogy, samples were pulverized and analysed as random mounts by X-ray diffractometry using Cu radiation. Carbonate content was measured by coulometric titration following the methods outlined in Engleman et al. (1985). This method measures the total amount of carbon in all carbonate minerals, including calcite, aragonite, and dolomite. Major element chemistry was done using wavelength-dispersive X-ray fluorescence. Trace element chemistry, including the REE, was measured on splits of bulk samples using instrumental neutron activation analysis or INAA (Budahn and Wandless, 2002).

6. Geochemistry of African dust

To characterize dust from Africa that could contribute to soils in southern Europe, including Mallorca, we utilize collections from two different localities. One is a suite of African dust samples collected in 1967, 1968, and 1969 by J.M. Prospero (University of Miami) on the island of Barbados. Collection protocols for these samples are given in Prospero and Nees (1977) and typical particle size distributions ($\sim$50% 20–2 μm; $\sim$50% $<$ 2 μm) are given in Prospero et al. (1970). Geochemical analyses of these samples were conducted in the laboratories of the U.S. Geological Survey and results are given in Muhs et al. (2007a). Results of these studies indicate that African dust collected on Barbados has a composition very close to that of average UCC (Taylor and McLennan, 1985). This result is not surprising, because it is likely that dust collected at a distal locality such as Barbados is a mix of source sediments derived from a broad part of the Sahara and Sahel regions of Africa. As such, this fine-grained dust is likely to be representative of the well-mixed dust that reaches southern Europe as well. Nevertheless, because of the possibility of dust with somewhat different compositions reaching southern Europe, we analysed a second suite of African dust samples. These consist of four samples of "red-rain" dust collected in a rural area northeast of Barcelona, Spain (41°46′N; 2°21′E), near Montseny (Fig. 5). Red-rain dust events have been studied in detail at this locality (Avila et al., 1997) and specific dust deposition events have been carefully documented from 1987 to 2002 (Avila et al., 2007). On Mallorca, African dust–rain events also have been documented from an overlapping period, 1982–2003 (Fiol et al., 2005). In the period common to both records (1987–2002), almost all of the African-derived dust–rain events observed are documented at both Montseny and Mallorca (Fig. 11).

Avila et al. (1997, 2007), using air mass trajectory analysis for dust reaching Spain, identify three main source areas in Africa, including Western Sahara, coastal Algeria and Tunisia, and central Algeria. Using a similar approach, Bergametti et al. (1989) identify dust reaching Corsica, not far from Mallorca, as originating from three regions of Africa: Sector 1, including Libya, Tunisia, and eastern Algeria; Sector 2, including western Algeria and Morocco; and Sector 3, a broad area of western Africa south of $\sim$30°N latitude, in general agreement with Avila et al. (1997, 2007). As Goudie and Middleton (2001) point out, specific sources of trans-Atlantic dust are not as well known, but they likely include Western Sahara, Morocco and Algeria, including many of the same regions as the sources of trans-Mediterranean dust.

We selected four of the samples from African dust events at Montseny for detailed geochemical analyses and comparison with the Barbados-derived African dust compositions. For ease of comparison, we present Sc–Th–La and Zr–Sc–Th ternary plots and rare earth element (REE) chondrite-normalized abundance plots. On the ternary plots, African dust collected on Barbados plots in a range that is typical for average UCC (Fig. 12). This range, from a petrological point of view, is felsic and is distinctive from Alaskan loess, derived from intermediate-composition rock sources (Muhs and Budahn, 2006), and New Zealand loess, derived from mafic rock sources (Graham et al., 2001). The African dust samples collected at Montseny plot in the middle of the range of African dust samples collected on Barbados (Fig. 12). On the REE plots, African dust samples collected on Barbados show a typical UCC signature, with enriched light REE, negative Eu anomalies, and a depleted, or "flat" heavy REE portion of the REE curve (Fig. 13). African dust samples collected at Montseny show the same general form of REE abundances as those collected on Barbados. We conclude from these analyses that the composition of far-traveled African dust collected on Barbados does not differ significantly from that collected in Montseny, Spain. Thus, we pool geochemical data from these two collecting localities in comparing the composition of African dust to soils, paleosols, and eolianites on Mallorca.

7. Stratigraphy and paleosol morphology

7.1. Els Bancals

The thickest and most complex section we studied is a sea cliff exposure called Els Bancals (also called "Es Bancals" by Cuerda and Sacarès, 1992), not far from the village of Cala Pi on the southwest coast of Mallorca (N39°21.518′; E02°49.199′). Many workers, including Cuerda (1975), Rose et al. (1985), Hearty (1987), Jenkyns et al. (1990), Cuerda and Sacarès (1992), and Nielsen et al. (2004), have studied the Els Bancals section previously. The section is $\sim$18 m thick and consists of a series of nine carbonate eolianites with intercalated paleosols, all resting on a marine platform cut on Miocene carbonate bedrock that is $\sim$15 m above sea level (Figs. 7, 9, 14 and 15).

The individual eolianite units at Els Bancals are generally between 50 and 200 cm thick and consist of well-cemented carbonate sands. In fact, eolianites are so well-cemented that a rock hammer is required to break off fragments and each eolian unit forms a prominent ledge in the outcrop. The eolianite between depths of 1000 and 1200 cm exhibits high-angle cross-bedding, with foreset beds dipping eastward (Fig. 9). However, the other eolianites were probably originally eolian sheet sands rather than dunes, based on their modest thickness and lack of cross-bedding. Most eolianites at Els Bancals are pink (7.5 YR 8/4, dry), although some are very pale brown (10 YR 8/4, dry). Although all eolian units are composed largely of carbonate grains, cemented by secondary carbonate, the pink (rather than white) color may be due to release of a small amount of Fe that is present as a trace element in dolomite, which is found throughout the section. Almost all the eolianites at Els Bancals contain carbonate rhizoliths (root casts), still in growth position, suggesting that eolian sand accretion took place in the presence of at least sparse plant growth.

Paleosols at Els Bancals are easily recognized in that they do not form ledges like the eolianites, but instead are concave or recessional features owing to a lesser degree of carbonate cementation (Figs. 7 and 9). In addition, they are usually redder than the host eolianite and have reddish yellow (7.5 YR 7/6, 7.5 YR 6/6, dry) colors, with a relatively high proportion of fine-grained particles (silts and clays), particularly in their upper parts. The one exception to this is the patchy modern soil at the top of the section, where thin A horizons have strong brown (7.5 YR 4/6, dry) or dark brown (7.5 YR 3/4, dry) colors. The paleosols sometimes contain angular, colluvial (?) clasts of pink or white eolianite. Finally, an important distinguishing characteristic is that paleosols at Els Bancals have gradual lower boundaries with the underlying eolianite and sharp contacts with overlying eolianite.

It is very difficult to estimate ages for the eolianite-paleosol sequence at Els Bancals, but a careful sifting of the observations

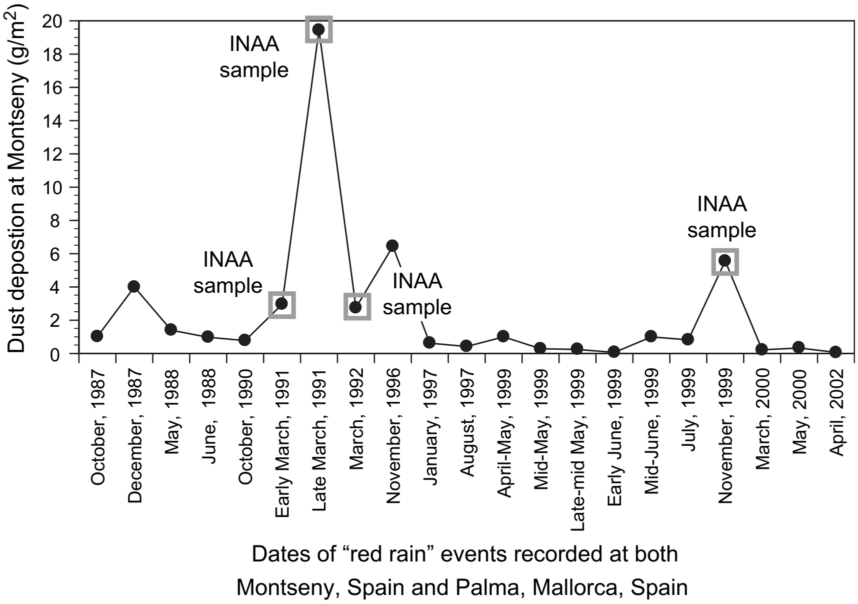

Fig. 11. Dates of "red-rain" dust events recorded at both Montseny, Spain (Avila et al., 2007) and Palma, Mallorca, Spain (Fiol et al., 2005), and dust deposition rates at Montseny. Also shown are samples selected for detailed geochemical analyses using INAA.

and analyses by previous workers gives some limits on possible ages. Cuerda and Sacarès (1992) thought that the marine platform at ~15 m above sea level was cut around the time of the Pliocene–Pleistocene boundary, which would suggest a basal age of ~2.6 Ma. The same workers also report remains and molds of marine shells such as *Monodonta* on top of the marine platform. Cuerda and Sacarès (1992) offer no specific evidence as to why they thought the marine platform ought to date from the Pliocene–Pleistocene boundary. However, these workers also describe a shell-bearing marine conglomerate that was deposited in a cave that cuts into the lower part of the succession at Els Bancals. Hearty (1987) analysed *Arca* shells from this deposit and reports amino acid ratios that are significantly higher than those in *Arca* shells from deposits elsewhere on Mallorca that have a U-series age of ~129 ka on fossil coral. Thus, Hearty (1987) infers that the older *Arca* shells at Els Bancals could correlate with MIS 9, ~300 ka. If this age estimate is correct, then the lower part of the sequence at Els Bancals must be older than ~300 ka. Nielsen et al. (2004) studied the Els Bancals section and used paleomagnetic and optically stimulated luminescence (OSL) methods to estimate ages of some of the units. An eolianite unit approximately midway through the section gives an apparent OSL age of 333 ± 70 ka and the uppermost eolianite unit gives an apparent OSL age of 275 ± 23 ka. These ages are consistent with their paleomagnetic age estimates. Using the OSL ages, the paleomagnetic correlations, magnetic susceptibility measurements, an assumed age of ~400 ka (MIS 11) for a marine deposit that underlies the whole sequence, and correlation to calculated Northern Hemisphere insolation, Nielsen et al. (2004) infer that the units within the Els Bancals section were deposited between ~410 ka and ~260 ka. We agree that the section could indeed represent much of the upper Middle Pleistocene and we further agree with Nielsen et al. (2004) that the eolianites represent glacial periods. What they refer to as reddish colluvial units (and what we interpret as paleosols) represent interglacial periods.

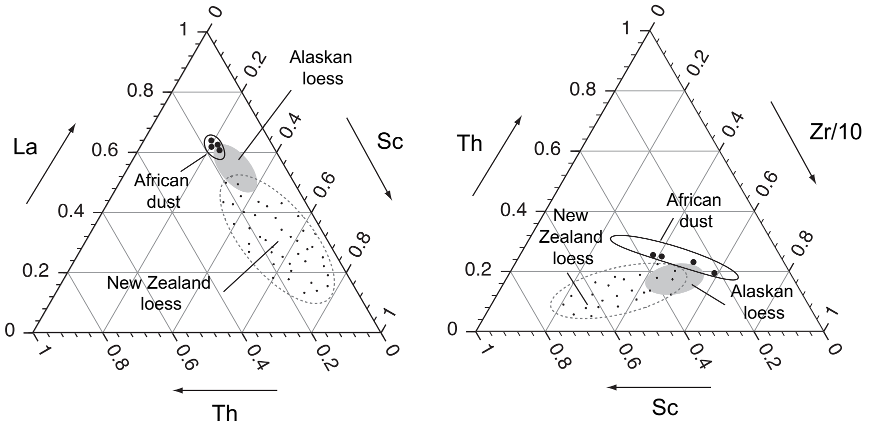

Fig. 12. Ternary plots showing relative abundance of Sc–Th–La and Zr/10-Sc–Th in various eolian materials. New Zealand loess data from Graham et al. (2001); Alaskan loess data from Muhs and Budahn (2006); African dust (collected at Barbados) data from Muhs et al. (2007a). African dust collected at Montseny (samples shown in Fig. 11) are shown as solid black dots.

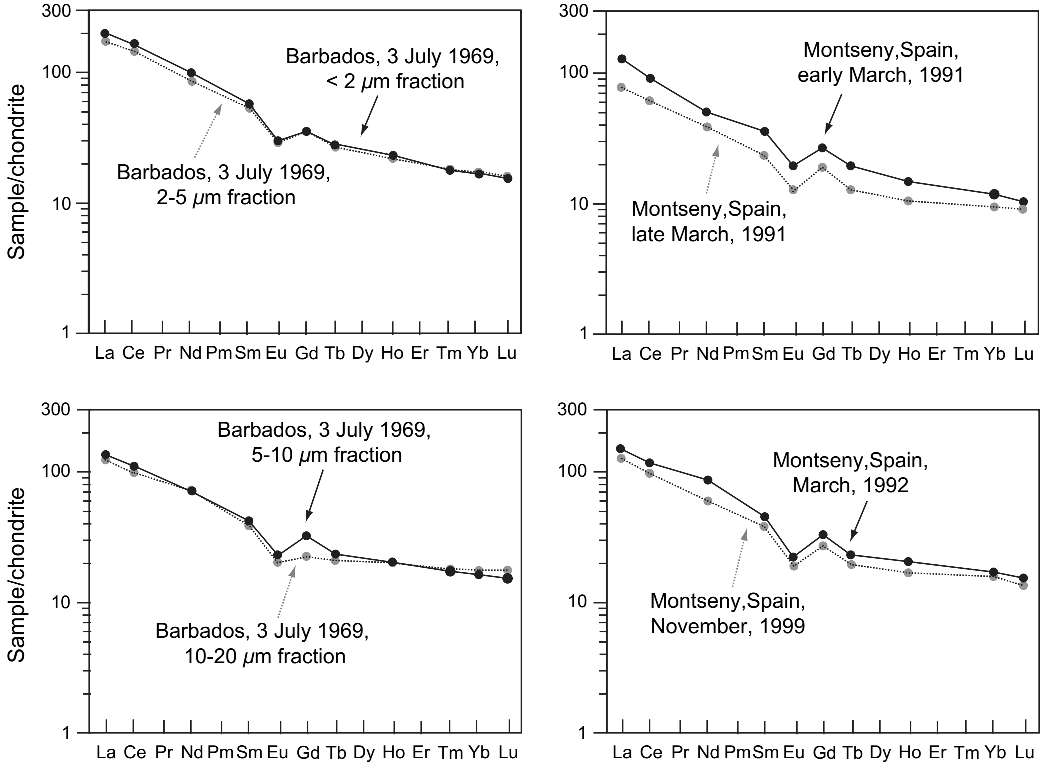

Fig. 13. Chondrite-normalized rare earth element plots of African dust collected on Barbados (data from Muhs et al., 2007a) and at Montseny, Spain (data from this study).

7.2. S'Estalella

A few kilometers to the east of Els Bancals is another coastal exposure called S'Estalella (N39°21.508′; E02°54.049′), previously studied by Butzer and Cuerda (1962), Butzer (1975), Rose et al. (1985), Hearty (1987), and Cuerda and Sacarès (1992). Our measurements and stratigraphic descriptions agree with those of all previous workers except Hearty (1987). The locality we studied is exposed on the middle of three prominent headlands found near the S'Estalella medieval tower. We measured 4.0 m of Miocene limestone exposed above sea level, truncated by a wave-cut bench (Figs. 7, 16 and 17). Both Butzer and Cuerda (1962) and Rose et al. (1985) report marine fossils above the wave-cut bench, but were not observed by us at this locality. The wave-cut bench is capped by a well-developed terra rossa paleosol that is a meter thick. The lower half of this paleosol has cemented soilstones and/or Miocene (?) rock fragments whereas the upper half has platy carbonate layers between zones of cemented clay. Secondary calcite is also pervasive throughout the profile in a complex network. The clay-rich zones are yellowish red (5 YR 5/6, 4/6, and 5/8), whereas the carbonates are white (10 YR 8/1 or 2.5Y 8/2). This well-developed soil is overlain by 0.8 m of eolianite that forms a prominent ledge in the sea cliff. No bedding is apparent in this unit and it has gradual contacts above and below. The eolianite has a thin (~30 cm) but very distinct terra rossa paleosol developed in its upper part, with red (2.5 YR 4/8) or yellowish red (5 YR 5/8) colors in the upper part of the profile and reddish yellow (5 YR 6/6) colors in the lower part. This paleosol is in turn overlain by a light yellowish brown (10 YR 6/4), light brown (7.5 YR 6/4), or reddish yellow (7.5 YR 6/6, 7/6, 7/8), cross-bedded eolianite that is ~2 m thick. The cross-beds are high-angle foresets that dip mostly to the east, in agreement with measurements made by Rose et al. (1985). This eolianite has a thin (~5 cm), patchy, well-cemented, yellowish red (5 YR 5/8) paleosol developed in its upper part. This thin soil is overlain by yet another eolianite, ~40 cm thick. The uppermost part of the section is ~30 cm of patchy, reddish yellow (7.5 YR 6/6) or pink (7.5 YR 7/4) soil mixed with fragments of the underlying eolianite and marine fossils, mostly mollusks. The marine fossils, where we observed them, occur at an elevation of ~9.2 m above sea level, although Butzer and Cuerda (1962) and Cuerda and Sacarès (1992) report them as high as 10–11 m above sea level. The same investigators studied the fossil assemblage in detail. It contains a number of extralimital southern species, including *Strombus bubonius*, *Brachidontes senegalensis*, and *Conus testudinarius*. Butzer and Cuerda (1962), Butzer (1975), Rose et al. (1985), and Cuerda and Sacarès (1992) all interpret the fossil-bearing marine deposits at 9–11 m at this locality to represent a high-sea stand at the peak of the Last Interglacial period, ~125 ka (MIS 5e). Hearty (1987), however, interpreted these fossils as storm deposits set down at sometime during the Holocene. It is difficult to imagine this mechanism, however, as the fossil assemblage contains at least six extralimital southern species of mollusks (Cuerda and Sacarès, 1992). Thus, reworking by Holocene storm waves would require transport of the major part of a Last Interglacial fossil assemblage from a seaward locality that no one has yet identified in the area. Furthermore, Hearty (1987, his Fig. 4) describes ~9 m of Miocene limestone exposed above sea level, whereas we and all other workers have measured only 4 m. We agree with the interpretations of Butzer and Cuerda (1962), Butzer (1975), Rose et al. (1985), and Cuerda and Sacarès (1992) that the section below the uppermost marine deposits represents a major portion of the upper Middle Pleistocene. The fossils at ~4 m above sea level overlying the wave-cut platform on Miocene limestone (not observed by us) were correlated to the "Tyrrhenian I" by Butzer and Cuerda (1962) and Rose et al. (1985). Thus, these deposits may represent the penultimate interglacial period (MIS 7, or ~200 ka), as proposed by Butzer (1975). The uppermost, fossil-bearing unit at ~9–~11 m above sea level represents the peak of the Last Interglacial period (MIS 5e,

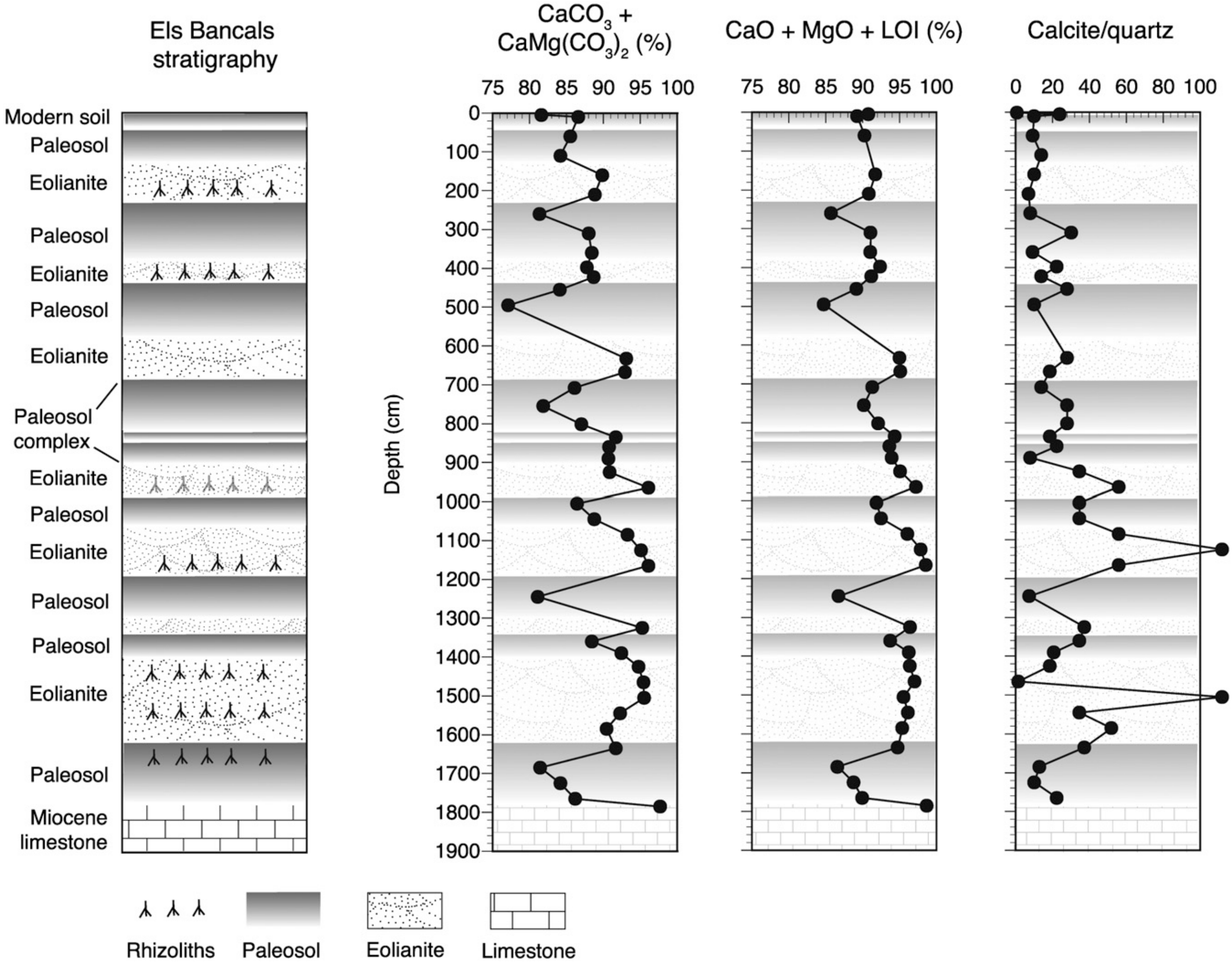

Fig. 14. Stratigraphy of the Els Bancals section, total carbonate and CaO + MgO + LOI contents, and ratio of calcite-to-quartz XRD peak heights.

or ~125 ka). If these correlations are correct, most of the package of eolianites and paleosols at S'Estalella represents a series of eolian and soil-forming events during the penultimate glacial period (MIS 6), between ~200 ka and ~125 ka.

7.3. Campo de Tiro-Palma airport road cut

We studied two eolianite sections southeast of the city of Palma, one in a road cut exposure near the airport and the other on the coast at a locality called Campo de Tiro (Figs. 7, 9, and 18–20). To our knowledge, the specific section we examined near the airport has not been studied previously, but it may be roughly equivalent to a section containing eolianites described as being ~500–600 m landward of Campo de Tiro, studied by Hillaire-Marcel et al. (1996) and Zazo et al. (2003; their Fig. 14). At Campo de Tiro, Hillaire-Marcel et al. (1996) and Zazo et al. (2003) report four marine units that truncate the oldest of three eolianite (dune) units. Their eolianite or dune (D) units are, from oldest to youngest, D1, D2 and D3. Rose et al. (1985), Hearty (1987), Hillaire-Marcel et al. (1996) and Zazo et al. (2003) interpret the marine units at Campo de Tiro as dating to the Last Interglacial period (MIS 5e). This interpretation is supported by amino acid ratios in *Glycymeris* shells that are similar to those from Son Grauet, a locality a short distance south of Campo de Tiro (Hearty, 1987). Marine deposits at Son Grauet have a U-series age of 129 ± 7 ka on coral (Hearty et al., 1986). Thus, the D1 eolianite unit that is truncated by the marine beds likely dates to the penultimate glacial period, or MIS 6. A few hundred meters shoreward, Zazo et al. (2003) describe the D1 eolianite as overlain unconformably by a younger eolianite, D2. Eolianite unit D2 is described as having a red paleosol developed in its upper part. Zazo et al. (2003) speculate that this paleosol could have formed during the Last Interglacial period. Whatever its age, this paleosol is in turn overlain by the youngest eolianite, D3. In places farther inland, however, eolianite unit D1 is exposed at the surface and hosts a red soil in its upper part.

We studied the Campo de Tiro eolianite section and our observations are consistent with those of Rose et al. (1985) and Zazo et al. (2003). We also suspect that the exposure we studied near the Palma airport contains some, if not all, of the same eolianite units (D2, red paleosol, D3) described by Zazo et al. (2003) as occurring landward of Campo de Tiro. Because of both quarrying of eolianite and building construction, we were unable to trace eolianite units from the coast to the airport road cut. We hypothesize that the eolianite units we observe at the Palma airport section are stratigraphically younger than the eolianite at Campo de Tiro (D1), but this interpretation is tentative.

The paleosol developed on what we interpret to be the "D1" eolianite at Campo de Tiro is clay-rich and has strong brown (7.5 YR 5/6) to reddish yellow (7.5 YR 7/6) colors whereas the eolianite is pink (7.5 YR 8/4) and is a well sorted, bedded and weakly cemented

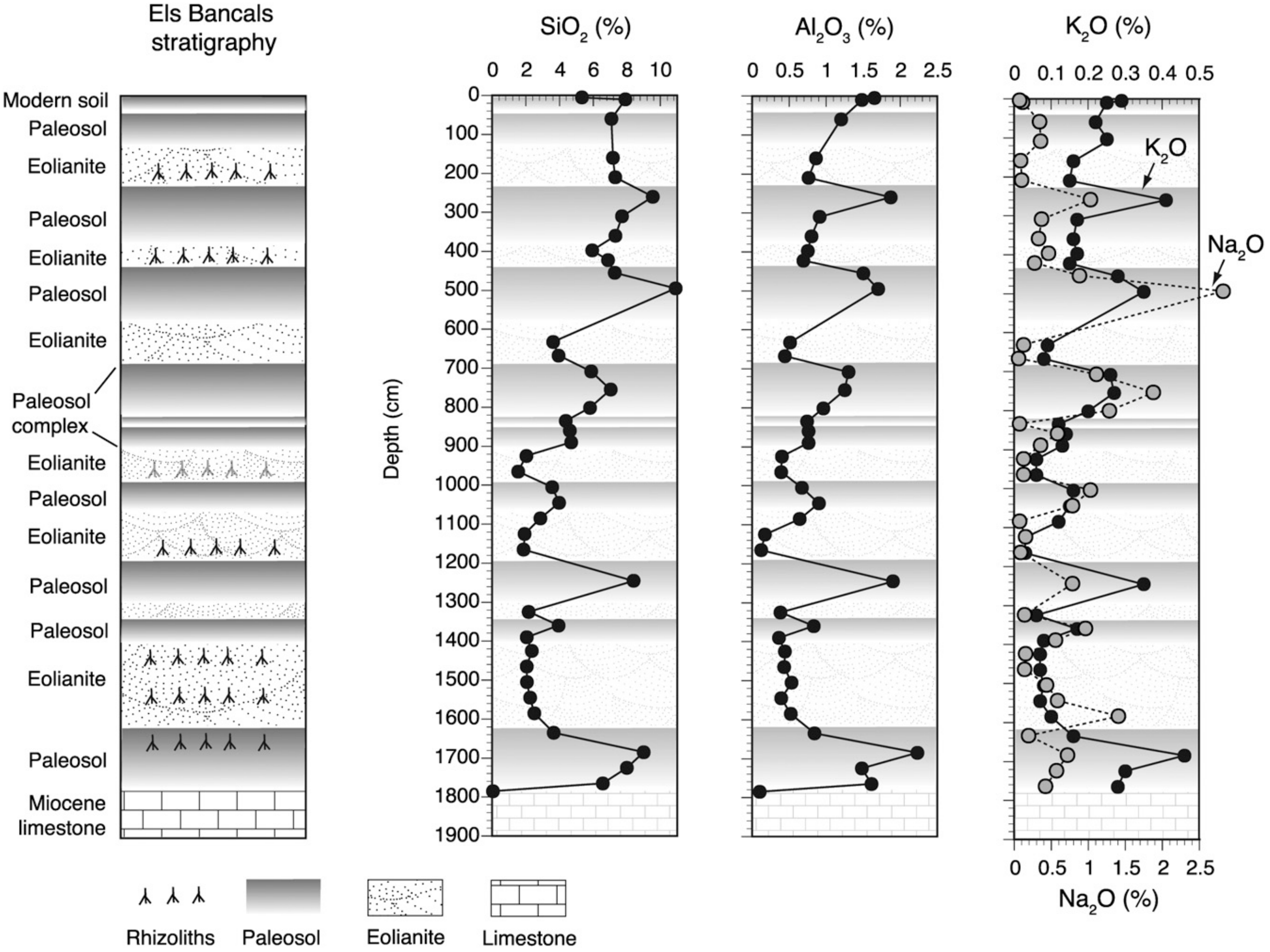

Fig. 15. Stratigraphy of the Els Bancals section and SiO_2, Al_2O_3, K_2O, and Na_2O contents.

sand. The paleosol developed on eolianite at Campo de Tiro is overlain by $\sim$0.5 m of highly fossiliferous marine deposits of Last Interglacial age, including abundant *Acar* sp. shells. The Last Interglacial marine deposit is in turn capped with what appears to be a stripped reddish clay and thin calcrete. At the Palma airport road cut, the lowermost paleosol studied is developed on what we refer to as unit Qes_2 (Fig. 18). This paleosol is pink (7.5 YR 7/4) in its lower parts and is redder in its upper horizons. It contains a stone line in its lower part and also hosts land snails. The paleosol above it, developed in what we refer to as eolianite unit Qes_3, is a very light pink color (7.5 YR 8/4) and also contains land snails in its lower part. In the three uppermost eolianites exposed in the Palma airport road cut, dips are to the southeast (Fig. 18). High-angle dips in the upper part of unit Qes_2 and most of unit Qes_3 are interpreted to be foreset beds, suggesting paleowinds from the northwest.

8. Mineralogy and major element geochemistry

8.1. Els Bancals

At Els Bancals, eolianites are dominated by carbonate minerals, primarily calcite, but also smaller amounts of dolomite. Quartz is present in small amounts in all eolianites as well. The dominance of calcite over dolomite is seen not only in the XRD peak heights but also the CaO/MgO ratios, which range from 28 to 60, with most ranging from 40 to 60. Nevertheless, the presence of dolomite indicates that sediments derived from the island bedrock make a contribution to the offshore sediment source for eolianite, in addition to skeletal debris or bioclastic grains that accumulated during Quaternary time. Total carbonate content in the eolianites, as measured by coulometric titration, ranges from 88 to 96% (Fig. 14). Total carbonate content, as estimated by XRF (CaO + MgO + LOI, where "LOI" is loss-on-ignition at 900 °C) is slightly higher (91–98%). Calcite-to-quartz XRD peak height ratios and CaO + MgO + LOI values parallel those of total carbonate content throughout the section.

In contrast, carbonate concentrations in the paleosols are much lower than in the eolianites. Total carbonate content in the paleosols, as measured by coulometric titration, ranges from 77 to 92%, with the modern soil at the top of the section having a much lower carbonate content of 35%. These carbonate concentration values are paralleled by the CaO + MgO + LOI values and calcite-to-quartz XRD peak height values, which are much lower in the paleosols than in the eolianites (Fig. 14). A notable trend is that the carbonate content decreases gradually upward from eolianites into the overlying paleosols, particularly for the units between $\sim$1350–1500 cm and $\sim$1000–1200 cm. This is consistent with the increasing degree of soil profile expression upward in each stratigraphic unit, as described earlier.

Other major element trends at Els Bancals show a reciprocal relation to the carbonate trends in the paleosols and eolianites. Concentrations of SiO_2 (representing quartz, feldspars, and clay

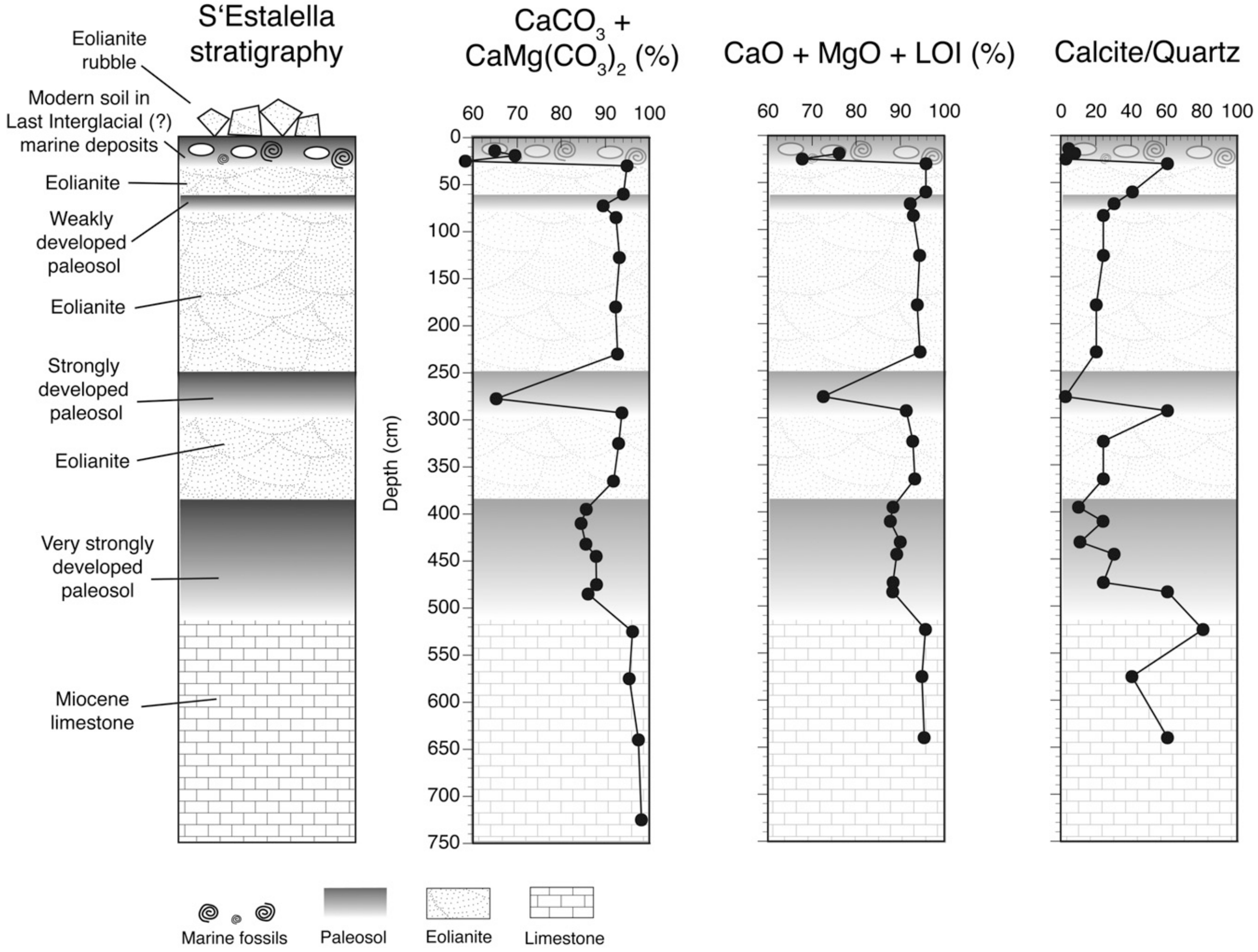

Fig. 16. Stratigraphy of the S'Estalella section, total carbonate and CaO + MgO + LOI contents, and ratio of calcite-to-quartz XRD peak heights.

minerals) are highest in paleosols and lowest in eolianites (Fig. 15). This depth trend is paralleled by concentrations of Al_2O_3 (representing feldspars, micas and clay minerals) and Na_2O and K_2O (representing feldspars and micas). XRD analyses indicate that relative amounts of feldspars are generally higher in paleosols compared to eolianites. Although we did not attempt XRD analyses of clay separates, bulk XRD analyses nevertheless indicate the presence of a probable kaolinite-like mineral in several of the paleosols, including those at ~1390–1360 cm, 1045–1005 cm, 925 cm, and 495–455 cm. This mineral shows a peak at ~11.7° 2-theta ($d = 7.55$ Å) in bulk, air-dry XRD analyses. Its peak position at an angle lower than 12.4° 2-theta ($d = 7.13$ Å), which is the kaolinite position, suggests the possibility that the mineral is halloysite (Moore and Reynolds, 1997). On the other hand, this mineral does not give peaks at 20° and 35° 2-theta, expected if it is halloysite. Thus, we regard this identification as tentative.

8.2. S'Estalella

Eolianites at S'Estalella, unlike those at Els Bancals, consist almost entirely of calcite with very small amounts of quartz. Total carbonate content, as measured by both coulemetric titration and XRF (CaO + MgO + LOI) is >90% in all eolianites (Fig. 16). Dolomite is absent in all units at S'Estalella, except in the modern soil, where it was unexpectedly detected. The lack of dolomite is reflected in the CaO/MgO values of these eolianites, which range from 44 to 95. Quartz contents are highest in the paleosols, although carbonate contents in these soils are still relatively high (59–94%). As expected, therefore, SiO_2, Al_2O_3, and K_2O are highest in the paleosols, where quartz feldspars and clay minerals were detected by XRD (Fig. 17).

8.3. Campo de Tiro-Palma airport road cut

Eolianites at the Campo de Tiro-Palma airport road cut localities (Fig. 18) have a mineralogy that differs slightly from both Els Bancals and S'Estalella. Similar to the section at Els Bancals, these eolianites contain calcite, dolomite and quartz. In addition, however, the Campo de Tiro and Palma airport eolianites also contain aragonite, and the uppermost unit at the Palma airport road cut also contains high-Mg calcite. Total carbonate contents in the Campo de Tiro and Palma airport eolianites range from 82 to 93% by coulemetric titration and 90 to 96% by XRF (CaO + MgO + LOI) (Fig. 19). With the presence of high-Mg calcite and dolomite, Palma airport eolianites have CaO/MgO ratios that are lower than those at S'Estalella, and range from 10 to 38. The paleosol at Campo de Tiro contains calcite, dolomite, aragonite, and quartz, but also feldspar and possibly mica. Hence, there are enrichments of SiO_2, Al_2O_3, and K_2O in this soil compared to the eolianite that hosts it (Fig. 20). Paleosols at the Palma airport road cut are not particularly well-developed, especially the uppermost one, which shows only a slight depletion of carbonate and enrichment of SiO_2, Al_2O_3, and K_2O

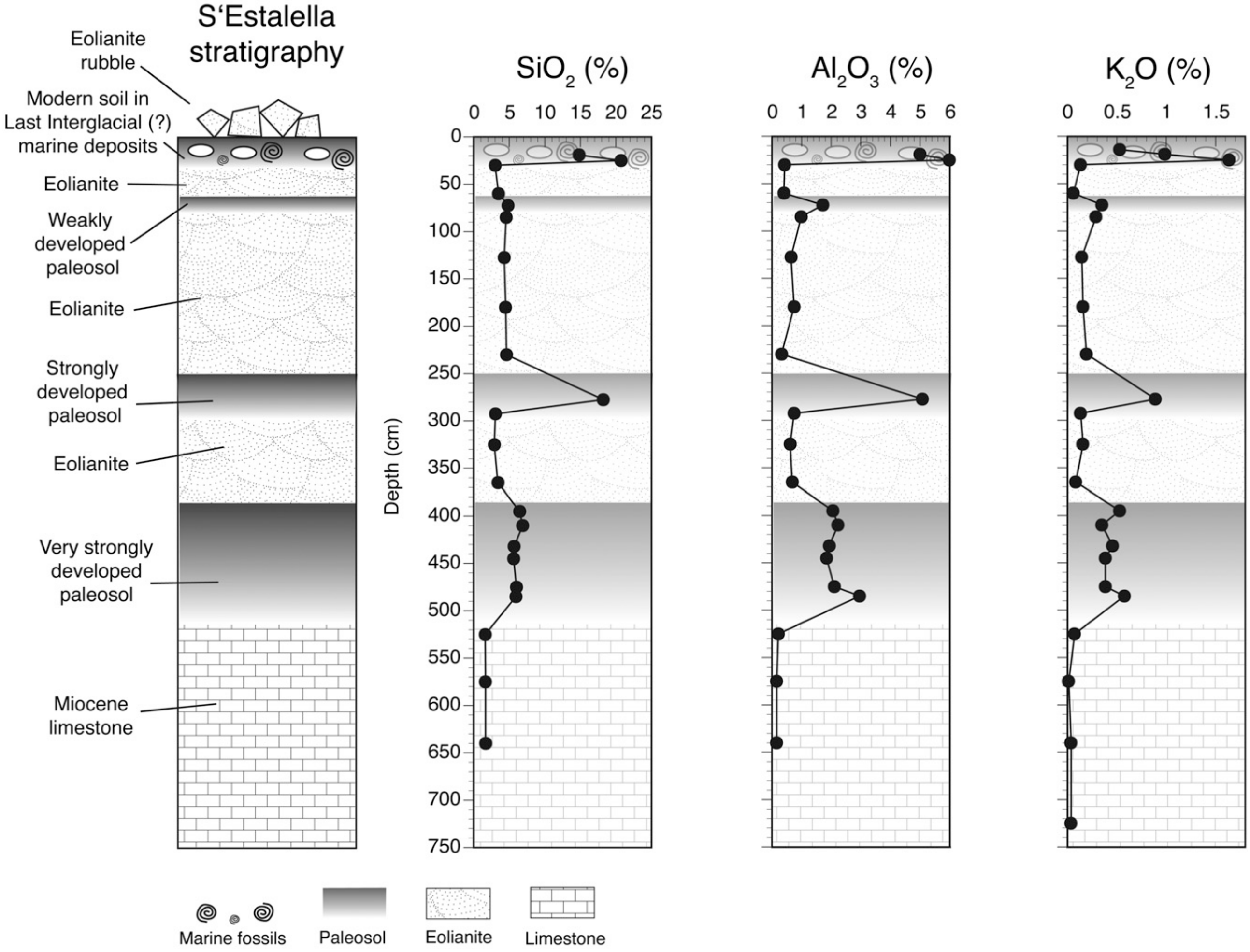

Fig. 17. Stratigraphy of the S'Estalella section and SiO_2, Al_2O_3, and K_2O contents.

(Fig. 20). The lower paleosol in this section is better expressed morphologically and also shows more carbonate depletion (Fig. 19). In addition to quartz enrichment, this paleosol also contains feldspars and clay minerals, based on XRD, and the presence of these minerals is reflected in enrichments in SiO_2, Al_2O_3, and K_2O.

8.4. Si/Al values in Mallorcan eolianites and paleosols

At all three sections, Si/Al values differ between eolianites and paleosols (Fig. 21). At Els Bancals, with the exception of the eolianite between 1400 and 1600 cm, eolianites have higher Si/Al than the paleosols that are developed on them. Similarly, at the S'Estalella and Camp de Tiro/Palma airport sections, Si/Al values in paleosols are relatively low whereas Si/Al in eolianites and the underlying limestone are significantly higher. Furthermore, Si/Al values in eolianites have the same or higher Si/Al values than UCC, which averages 3.83 (Taylor and McLennan, 1985). The higher Si/Al values probably reflect the abundance of quartz over feldspar in the non-carbonate fraction of the eolianites. Si/Al values have significance for paleosol origins, which we discuss later.

9. Trace element geochemistry of Mallorcan eolianites, paleosols, and African dust

An important observation to note in the results of mineralogy and major element geochemistry discussed above is that although the non-carbonate component of Mallorcan eolianites is low, it is not zero. As discussed earlier, a traditional hypothesis for the origin of terra rossa and similar soils is accumulation of the insoluble residue as carbonate dissolution proceeds. A key topic we address in this section, therefore, is whether the composition of the non-carbonate component of the eolianites is identical to that of the paleosols. We recognize that, despite the present semiarid Mediterranean climate of Mallorca, past climates could have been more humid and chemical weathering could have been significant at those times. Indeed, the very hypothesis being tested here *requires* that chemical weathering has taken place in the paleosols. Thus, in comparing compositions of the non-carbonate component of the eolianites with soils, we use only high-field-strength elements that are the least mobile in soils, including Zr, Hf, La, Yb, Th, and Ta. All these elements have been used previously in investigations of soil genesis where the possibility of intense chemical weathering (such as that in humid tropical climates) is a concern (Kurtz et al., 2001; Muhs et al., 2007a; Muhs and Budahn, 2009).

9.1. Els Bancals

In order to portray down-section variation in immobile element composition, we chose four key element ratios that have been used in previous investigations as good discriminators for parent materials. The Zr/Hf ratio is useful because both elements are found almost exclusively in one mineral, zircon. Thus, large variations in Zr/Hf in sediments would indicate derivation of zircons from plutons with different sources. The only disadvantage in using this

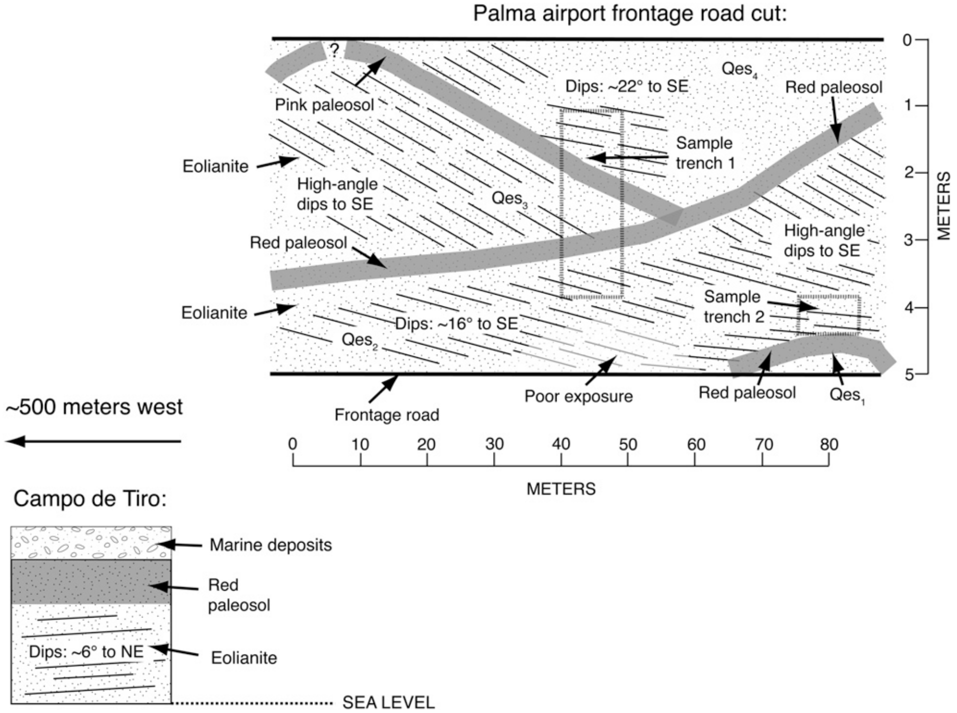

Fig. 18. Stratigraphy of the Campo de Tiro and Palma airport frontage road cut sections.

ratio is that concentrations of Zr in some of the eolianites are low enough that INAA is not able to yield reliable data. For those units in which this is a concern, we have not calculated this ratio and those data points are not displayed in our figures. Intervals where sample points have no data are indicated by dashed lines. At Els Bancals, Zr/Hf is highest in eolianites and lowest in soils (Fig. 22). Several eolianites have Zr/Hf values greater than 50. In contrast, all paleosols have Zr/Hf values that fall within the range of African dust, which is 30–50.

A measure of the enrichment of light REE to heavy REE is the La/Yb ratio. Sediments derived from the UCC, such as many loesses (Gallet et al., 1998; Jahn et al., 2001; Sun, 2002a,b; Muhs and Budahn, 2006; Sun et al., 2007) and African dust (Muhs et al., 2007a) are enriched in light REE. At Els Bancals, La/Yb is highest in soils and lowest in eolianites (Fig. 22). African dust has La/Yb values that range from 8 to 16 and all paleosols in the section fall within this range. In contrast, all eolianites in the lower part of the section (below ~900 cm depth) have La/Yb below the range of African dust. In the upper part of the section, La/Yb in eolianites are within the range of African dust, but are significantly lower than La/Yb in the soils developed in their upper parts. Furthermore, many of the eolianite–paleosol couplets display gradual increases from the eolianite upward into the soil, reaching a maximum value just below the contact with the next-youngest eolianite. This suggests a possible scenario of increasing accumulation of African dust in the soil as pedogenesis proceeds. Such a pattern is consistent with the observation of increasing soil morphological expression and decreasing carbonate content upward through a given profile, as noted earlier.

Both Cr and Sc are found in a variety of primary, rock-forming, Fe-bearing minerals as well as several clay minerals. Muhs and Budahn (2006) found that Cr/Sc was an effective discriminator for fluvial silts from different drainages in Alaska that were hypothesized sources of loess. At Els Bancals, Cr/Sc is consistently higher in eolianites than in paleosols (Fig. 22). Moreover, many paleosols have Cr/Sc values that plot within (or nearly within) the range of values for African dust.

The Th/Ta ratio is a sensitive indicator of sediment origins, even for materials that are all derived from the UCC. For example, Muhs et al. (2007a) reported significantly different Th/Ta values for fine-grained African dust (<20 μm) and the fine-grained portion of loess (<20 μm) from the lower Mississippi River valley. At Els Bancals, Th/Ta values are lowest in paleosols and highest in eolianites (Fig. 22). As with Zr/Hf and La/Yb, all Th/Ta values in paleosols at Els Bancals fall within the range of African dust whereas many (though not all) eolianites do not.

9.2. S'Estalella

Immobile trace element ratios at the S'Estalella section display many of the same trends seen at Els Bancals (Fig. 23). It was not possible to compute Zr/Hf for many of the eolianite samples at S'Estalella because of the methodological problems alluded to earlier. However, those eolianites for which we do have data (as well as the underlying Miocene limestone) have Zr/Hf that are significantly greater than in the paleosols, similar to Els Bancals. Furthermore, Zr/Hf in the paleosols all fall within the range of African dust. For La/Yb, many of the eolianites have values that plot within the lower part of the range of African dust, but all values are significantly lower than what is observed in the paleosols. La/Yb in paleosols at S'Estalella, with one exception, all fall within the range of African dust. Similar to Els Bancals, eolianites and Miocene limestone at S'Estalella have Cr/Sc values that are consistently higher than in the paleosols. Although none of the paleosols have Cr/Sc values that are within the range of African dust, all are just slightly greater than this range. Th/Ta values in eolianites and Miocene limestone at S'Estalella are mostly higher than the range for African dust and are higher than Th/Ta in the paleosols, except for the thin paleosol at ~60 cm. Other than this paleosol, Th/Ta values in the paleosols all fall within the range of African dust.

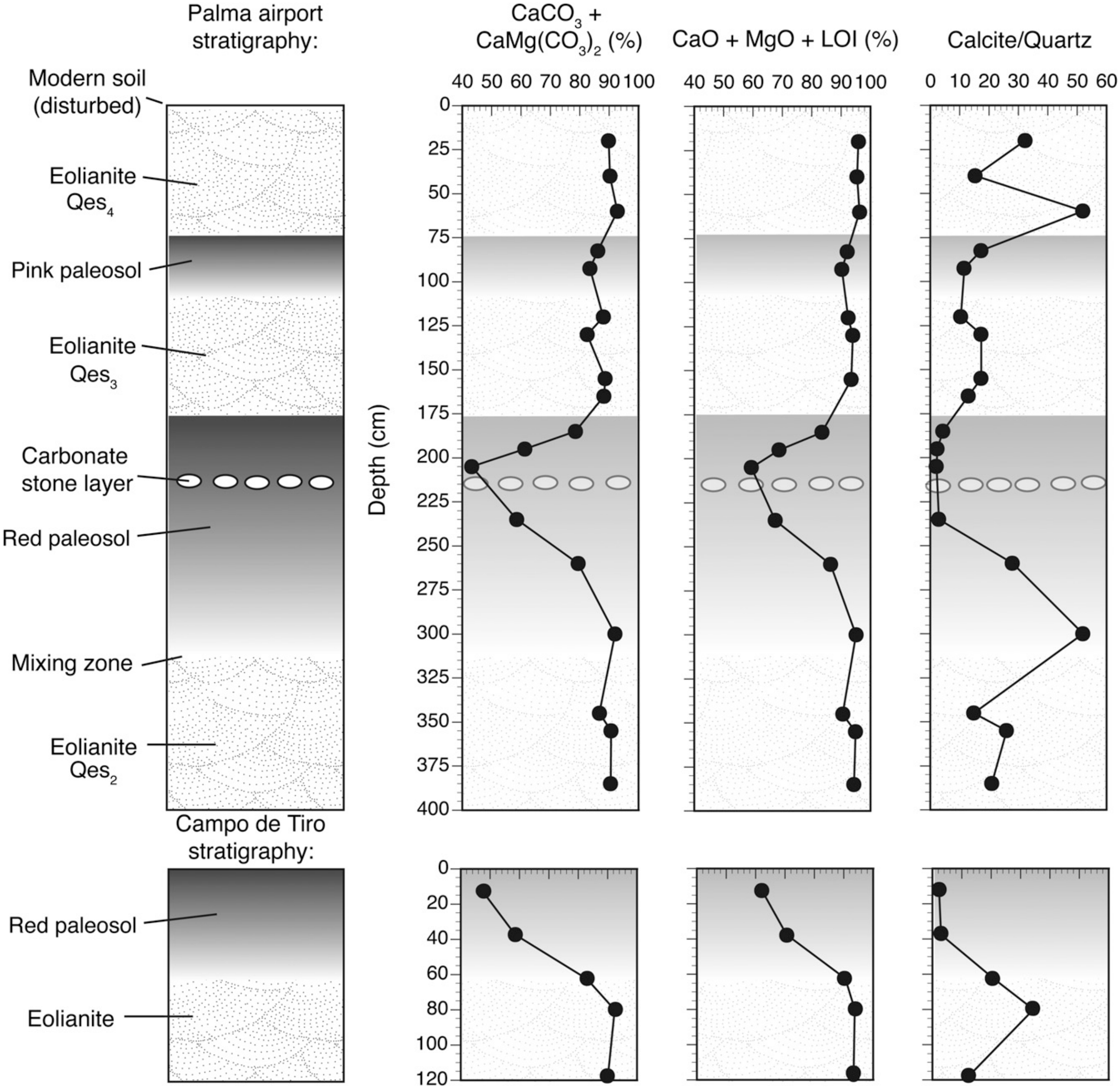

Fig. 19. Stratigraphy of the Campo de Tiro/Palma airport road cut sections, total carbonate and CaO + MgO + LOI contents, and ratio of calcite-to-quartz XRD peak heights.

9.3. *Campo de Tiro-Palma airport road cut*

Trace element ratios at the Campo de Tiro-Palma airport localities mostly show similarities to the sections at Els Bancals and S'Estalella (Fig. 24). The paleosol at the Campo de Tiro locality has Zr/Hf, La/Yb, Cr/Sc, and Th/Ta values that all fall within the range of African dust, and all values in the paleosol differ from those in the underlying eolianite. Unfortunately, we were unable to obtain precise estimates of the concentrations of Zr for many of the eolianite samples at the Palma airport section. For the two eolianite samples where we had success, however, one has a Zr/Hf value well above those found in the paleosols and one is slightly greater. Both paleosols have Zr/Hf within the range of values for African dust, however. For La/Yb, samples at all depths in the section, whether eolianites or paleosols, have values within the range of African dust, but both paleosols have higher La/Yb, similar to Els Bancals and S'Estalella. Cr/Sc at the Palma airport section is also similar to the other two sections inasmuch as all three eolianites have significantly higher values than do the two paleosols. Furthermore, both paleosols have Cr/Sc that fall clearly within the range of African dust, whereas the eolianites do not. Th/Ta at the Palma airport section also displays a trend similar to Els Bancals and S'Estalella. Eolianite units Qes_2, Qes_4, and the upper part of Qes_3 have Th/Ta significantly higher than the paleosols. Nevertheless, the middle and upper parts of Qes_3 have Th/Ta similar to the paleosols. Both paleosols have Th/Ta that fall within the range of African dust.

9.4. *Summary: trace element geochemistry*

Finally, we also present the trace element geochemistry of Mallorcan paleosols and eolianites for all three sections as bivariate plots of Zr/Hf vs. La/Yb and Cr/Sc vs. Th/Ta (Fig. 25). The Zr/Hf

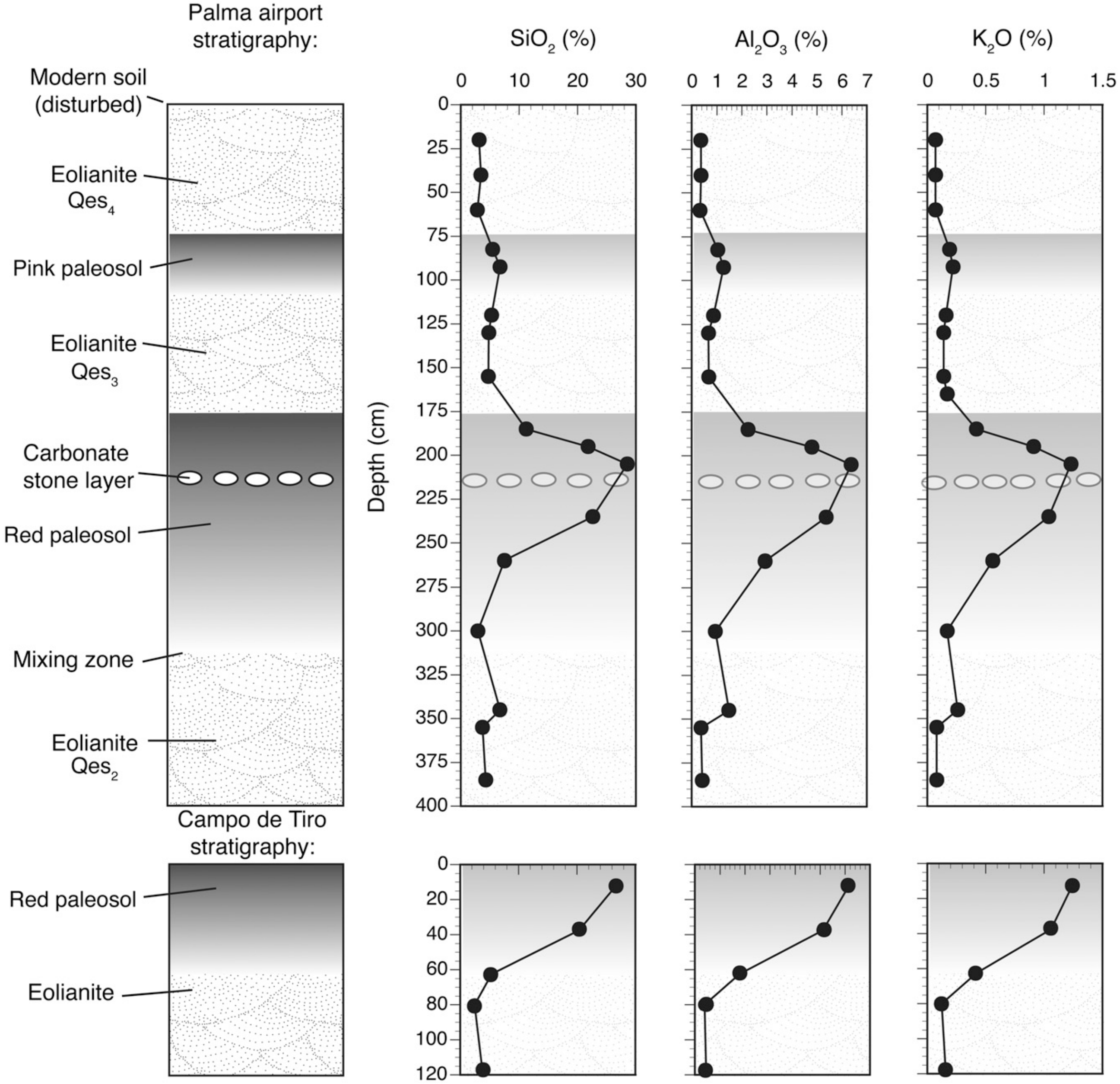

Fig. 20. Stratigraphy of the Campo de Tiro/Palma airport road cut sections and SiO_2, Al_2O_3, and K_2O contents.

vs. La/Yb fields for African dust and eolianite at Els Bancals and Palma airport/Campo de Tiro show some overlap. In all others, however, the trace element ratios define compositional fields that are distinct for Mallorcan eolianites and African dust. The plots are done separately for each section because eolianite composition varies from locality to locality. For Zr/Hf vs. La/Yb, paleosols plot mostly within the African dust field. For Cr/Sc vs. Th/Ta, some paleosols fall between the African dust and eolianite fields whereas others fall entirely within the eolianite field.

10. Discussion

Mallorca has a rich record of multiple periods of eolian sand deposition and lithification into eolianite, punctuated by periods of stability and pedogenesis. Lithification of the eolian sands into eolianite is thought to occur relatively soon after deposition (Clemmensen et al., 1997). Our findings indicate that eolianites on Mallorca have a very high carbonate content, ranging from 88 to 96%, with most being >90%. Such high carbonate contents would permit rapid lithification, as envisioned by Clemmensen et al. (1997). In addition, however, these eolianites can be regarded as limestones of considerable purity. Furthermore, existing stratigraphic relations and numerical dating by previous investigators indicate that the eolianites of Mallorca are of Quaternary age, and some of them are almost certainly of Middle and Late Pleistocene age. Lack of beach sources during the present interglacial period suggests that most of the eolianites were probably deposited during glacial periods.

Although the pink, red, or reddish-brown materials that are intercalated with the eolianites have previously been interpreted as colluvial deposits (e.g., Butzer and Cuerda, 1962), our results suggest that these features are buried soils or paleosols. We

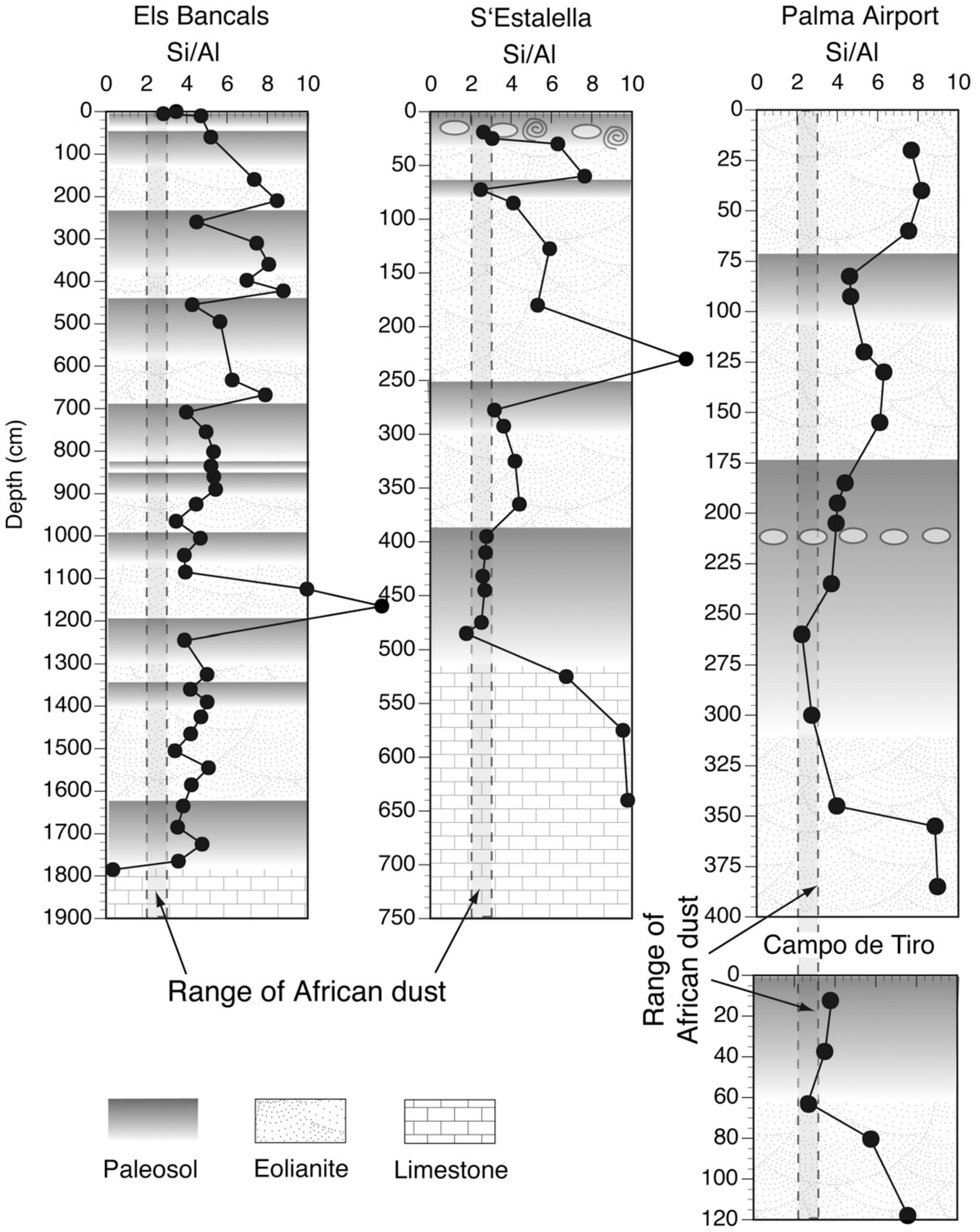

Fig. 21. Ratios of Si/Al at all three sections studied. Also shown for comparison is the range of Si/Al in African dust studied by Bergametti et al. (1989).

agree, however, that a colluvial component may be present in the paleosols. Evidence for colluvial additions takes the form of eolianite fragments and occasional fragments of cemented soil B horizon material. Nevertheless, morphological, mineralogical and chemical depth trends indicate to us that pedogenesis has been the dominant process in the formation of the red or reddish-brown zones between eolianite units. The paleosols, with their distinctive colors, contrast strongly in the field with the underlying eolianites and fall into the broad category of what were previously called terra rossa soils, Red Mediterranean soils, or Reddish Brown soils.

Paleosols developed on, or intercalated with high carbonate eolianites on Mallorca are characterized by lower carbonate mineral contents and higher silicate mineral contents. Thus, soils of this composition, developed on high carbonate substrates, require genesis either by a considerable amount of carbonate dissolution on a Quaternary timescale or additions of externally derived materials, whether local or far-traveled. Field evidence indicates that carbonate dissolution of eolianites is not likely a dominant process in the genesis of the paleosols on Mallorca. Although there is no question that karst processes have been and continue to be active on the island (Instituto Geológico y Minero de España, 1986;

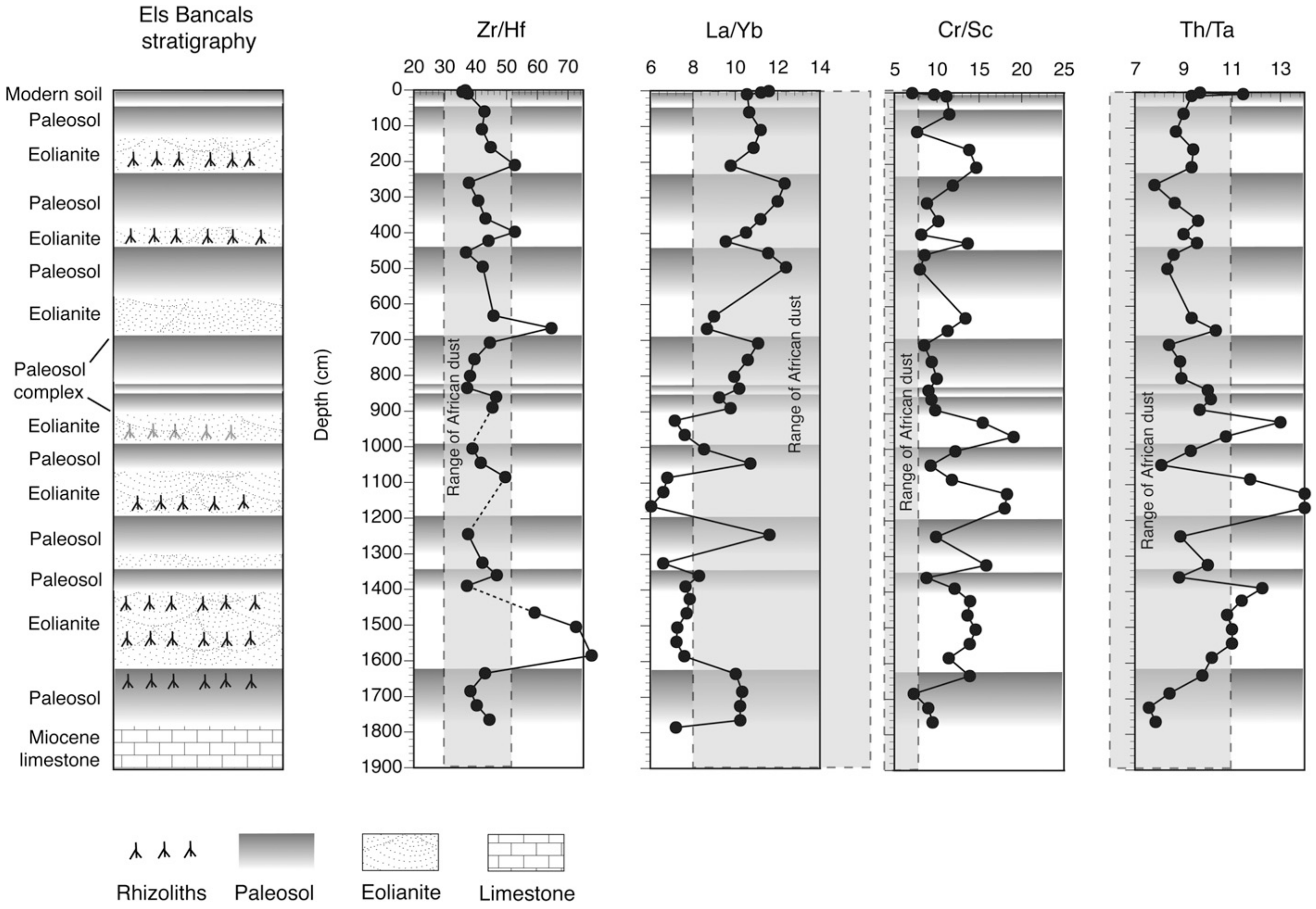

Fig. 22. Stratigraphy of the Els Bancals section and Zr/Hf, La/Yb, Cr/Sc, and Th/Ta values. Also shown is the range of these values in African dust (from data in Muhs et al., 2007a and this study).

Csoma et al., 2006; Tuccimei et al., 2006; Dorale et al., 2010), most of the soils and paleosols we examined show good horizontal continuity and follow the paleotopography defined by the underlying dune or sand sheet morphology (Fig. 7). Detailed horizontal cross sections at Els Bancals, illustrated by Nielsen et al. (2004), display this characteristic persuasively. If carbonate dissolution and accumulation of insoluble residues were the dominant processes in producing soils on Mallorca, we would expect to find field evidence of this. Such evidence might take the form of sinkholes filled with red or reddish soil materials, adjacent to eolianite surfaces that hosted thin soils or no soils at all. Such features are lacking in all sections we studied.

Additions of silicate-dominated parent materials to eolianite surfaces could, in principle, be locally derived from Mallorca, by alluvial, colluvial, or eolian processes. Nevertheless, there are limits as to how much silicate material could be added to soils on Mallorca from local sources. Carbonate lithologies, either limestones or dolomites of Tertiary or Mesozoic age, dominate the island bedrock of Mallorca (Gelabert et al., 1992, 2003; Pomar and Ward, 1995). Because these rocks have low silicate contents (see for example, data for S'Estalella in Figs. 16 and 17), it is unlikely that they would provide much non-carbonate parent material for soils developed on the Quaternary eolianites.

Ratios of immobile trace elements (Zr/Hf, La/Yb, Cr/Sc, and Th/Ta) provide the best basis for evaluating the origin of Mallorca's Quaternary soils and paleosols. Because all of these elements have minimal mobility in near-surface terrestrial environments, they long have been used as sediment provenance indicators. Even if past periods of pedogenesis occurred during paleoclimates more humid than Mallorca's present semiarid climate, loss of any of these elements by leaching is unlikely. As a consequence, ratios of the immobile elements should be faithfully retained in the paleosols and reflect the ratios of the same elements in the parent material.

Although Mallorcan eolianites have very high carbonate contents, there is a measurable silicate component. The silicate component of Mallorcan eolianites is likely derived from the small amount of non-carbonate minerals in the Mesozoic bedrock found in the mountainous portions of Mallorca. Such materials, along with a greater amount of detrital carbonate sediments, likely were eroded from the mountains, carried by fluvial systems offshore, and became a component of the shelf sands that ultimately became the source of Quaternary eolian sands. Fornós and Ahr (1997) report small amounts of quartz offshore Mallorca, which supports this scenario. The non-carbonate component of Mallorcan eolianites is high enough (∼4–∼12%) that abundances of Zr, Hf, La, Yb, Cr, Sc, Th, and Ta can be reliably measured by INAA methods. Ratios of Zr/Hf, La/Yb, Cr/Sc, and Th/Ta in the eolianites are consistently different from ratios of the same elements found in the soils and paleosols. Zr/Hf, Cr/Sc, and Th/Ta are always lower in paleosols than they are in eolianites, whereas La/Yb is always higher in paleosols than it is in eolianites. This pattern is apparent in all three sections examined, suggesting that it is not an artifact of locally derived additions of material. The observations suggest that the soils and paleosols are not derived primarily by

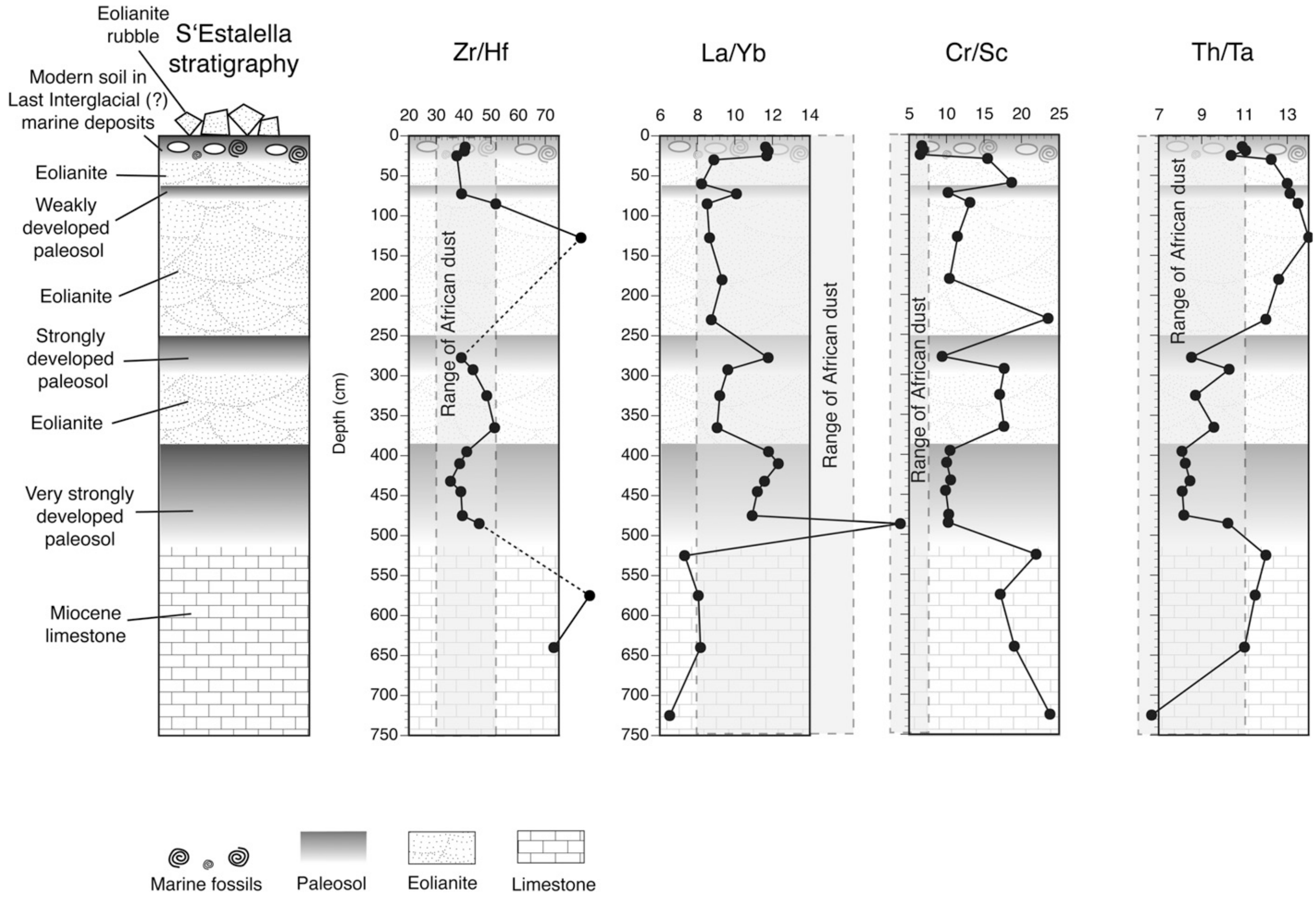

Fig. 23. Stratigraphy of the S'Estalella section and Zr/Hf, La/Yb, Cr/Sc, and Th/Ta values. Also shown is the range of these values in African dust (from data in Muhs et al., 2007a and this study).

concentration of insoluble residues in the eolianites and that additions of local sources are not involved to any great extent. From this, we conclude that a far-traveled, external eolian source must be involved.

One possible external source that must be considered is the fine-grained (<20 μm) component of loess from Europe. Because Mallorca is situated in the western Mediterranean Sea, the closest European source that could have affected the island would be the fine-grained component of that loess that was deposited in France. The recently published map of loess distribution in Europe shows that loess and "loess derivates" are widely distributed in northern France, along the southern margins of the English Channel, and eastern France, along the Rhone River valley (Haase et al., 2007). Although the geographic distribution of this loess suggests little transport beyond the local or regional sources, the mass accumulation rates calculated for the Last Glacial period are high enough (Frechen et al., 2003) that farther, long-distant transport of a fine-grained component of this loess must be considered a possibility. In addition, Antoine et al. (1999) show that loess in northwestern France contains a significant amount of clay (<2 μm) and fine silt (<20 μm), particle sizes that are capable of long-range eolian transport. Muhs et al. (2007a) considered the same possibility for a fine-grained Mississippi River loess component to soils on carbonate substrates in the Florida Keys and the Bahamas. Unfortunately, there are few geochemical data available for European loess to test this hypothesis. The only study of French loess of which we are aware that contains trace element geochemical data similar to ours is Gallet et al. (1998). Their data were derived from analyses of whole-sediment (i.e., all particle size fractions) and <160 μm fractions. Computation of La/Yb and Th/Ta values for these two fractions of French loess do not differ significantly from one another and all values fall into the same range that we observe for African dust. These observations permit the possibility that Mallorcan soils could contain a component of European-derived loess. Although this hypothesis requires more testing, preferably with just the <20 μm fraction, we do not favor a European origin for Mallorcan soils for two reasons. One is the lack of even thin loess in central and southern France, between the main loess bodies and Mallorca, suggesting that loess was mostly deposited locally, as pointed out by Frechen et al. (2003). The other reason we do not favor a European origin for Mallorcan soils is that deposition of loess in France (and Europe in general) occurred primarily during glacial or stadial periods (Frechen et al., 2003). In contrast, Mallorcan soils seemed to have formed primarily during interglacial or interstadial periods, as discussed earlier.

Although the trace element composition of soils and paleosols on Mallorca differs from that of the local eolianites, it is not significantly different from that of African dust. Zr/Hf, La/Yb, and Th/Ta values in the soils and paleosols are mostly all within the range of values for African dust at all three sections. Although Cr/Sc values in Mallorcan paleosols are slightly higher than those in African dust at Els Bancals and S'Estalella, they are much closer to African dust values than they are to Cr/Sc values in the local eolianite. From these observations, we conclude that Mallorcan

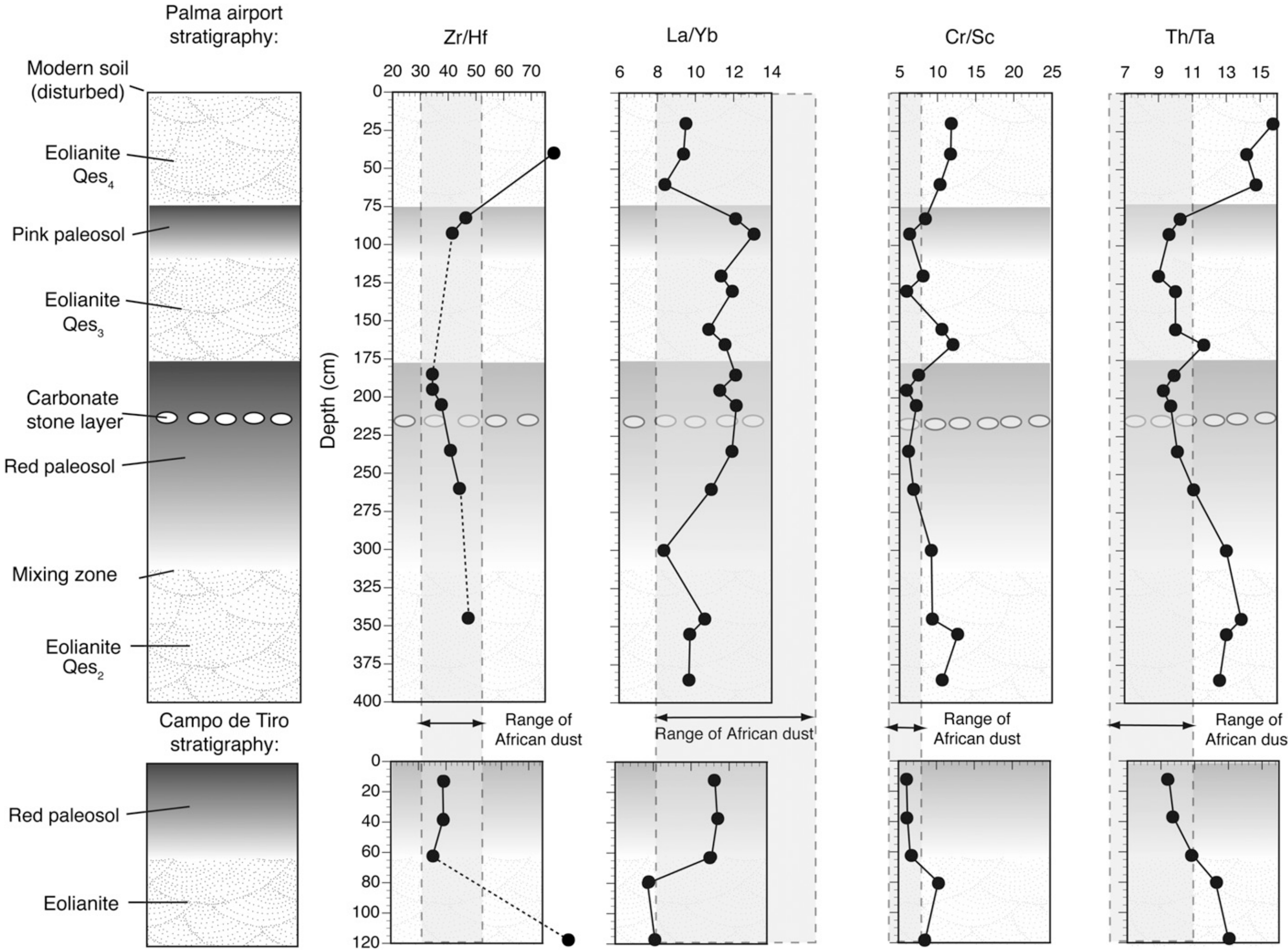

Fig. 24. Stratigraphy of the Campo de Tiro and Palma airport road cut sections and Zr/Hf, La/Yb, Cr/Sc, and Th/Ta values. Also shown is the range of these values in African dust (from data in Muhs et al., 2007a and this study).

paleosols may have a small component of silicate material derived from the local eolianite, but the majority of the particles in the paleosols are likely derived from African dust.

Nevertheless, a small component of locally derived silicate material in Mallorcan paleosols is supported by Si/Al values. Bergametti et al. (1989) collected African dust in 1985 and 1986 on the island of Corsica, not far from Mallorca (Fig. 1). These investigators conducted major element analyses of the dust they collected using wavelength-dispersive XRF. This is identical to the procedure used in the present study for the same major elements; thus, their data are directly comparable to ours. Mean values of Si/Al in African dust differ in the three regions identified as source areas by Bergametti et al. (1989). Sector 1 (Libya, Tunisia, and eastern Algeria) has a mean Si/Al value of 2.2; Sector 2 (western Algeria and Morocco) has a mean value of 2.67; and Sector 3 (western Africa south of ~30°N latitude) has a mean value of 2.92. Although the differences between the three regions are significant, it is noteworthy that all are low, with values less than 3.0. In principle, dust from broad regions such as those identified by Bergametti et al. (1989) should be well-mixed sediments derived from UCC. Nevertheless, Si/Al values from all three regions are significantly lower than the average UCC value of 3.83 (Taylor and McLennan, 1985). The much lower Si/Al values in the dust samples of Bergametti et al. (1989) likely reflect the higher content of phyllosilicate clay minerals, which are enriched in Al compared to average UCC. Because both Mallorca and Corsica are situated 700–1000 km from even the closest dust source regions in Africa, we hypothesize that African dust delivered to these islands is enriched in fine-grained phyllosilicate clay minerals, thus explaining the low Si/Al values.

If dust delivered to Mallorca from Africa has low Si/Al, similar to that of dust delivered to Corsica, then it follows that Mallorcan soils should have low Si/Al values (<3.0) if they are derived entirely from African dust. If Mallorcan soils have Si/Al >3.0 (Fig. 21), then they must contain a component of silicate material derived from a source other than African dust, most likely the non-carbonate component of the local eolianite. We recognize that, unlike the high-field-strength trace elements discussed below, Si is potentially mobile in soils under a wide range of conditions, although Al typically is not. Thus, if Mallorcan soils have low Si/Al values, within the range of African dust, it could reflect either derivation dominantly from that source or derivation from a local source, followed by Si loss from chemical weathering during pedogenesis. However, Si/Al values in Mallorcan soils that are higher than those seen in African dust would require contributions from a local, high Si/Al source, even with some Si loss due to weathering and pedogenesis.

All of the paleosols at S'Estalella have Si/Al values within the range of African dust, suggesting little or no contribution from the local eolianite. In contrast, with the exception of the modern soil,

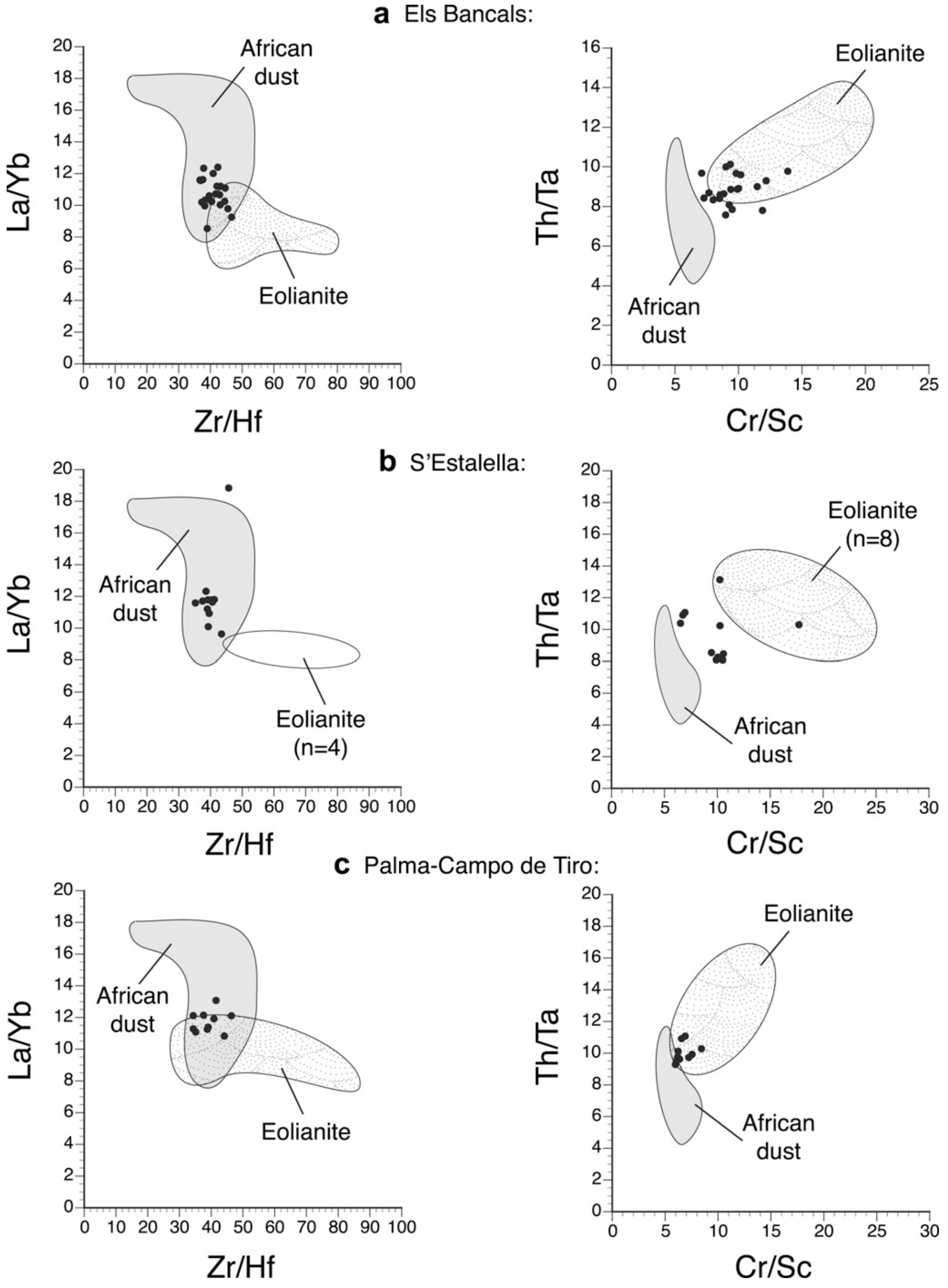

Fig. 25. Zr/Hf vs. La/Yb and Cr/Sc vs. Th/Ta as bivariate plots for African dust, eolianite and paleosols at (a) Els Bancals, (b) S'Estalella, and (c) Palma airport road cut/Campo de Tiro. Note scale difference on Cr/Sc vs. Th/Ta for (a). Not included are weakly developed paleosols; note that only 4 eolianite samples from S'Estalella had sufficiently high Zr contents for confident calculation of Zr/Hf values.

Si/Al in all the paleosols at Els Bancals are higher than even the highest values of African dust reported by Bergametti et al. (1989). The same is true, with two exceptions, for the horizons in paleosols at the Campo de Tiro/Palma airport road cut sections. We conclude from these observations that the paleosols at Els Bancals and Campo de Tiro/Palma airport sections must contain at least some component of locally derived silicate minerals, likely quartz. Nevertheless, even at these sections, Si/Al values in most paleosols are much closer to African dust values than to the local eolianite, suggesting that long-range-transported dust is the dominant soil parent material.

The identification of long-range-transported African dust as a significant parent material for soils on relatively pure carbonate substrates on Mallorca is supported by geochemical data collected elsewhere in the Mediterranean. Genova et al. (2001) studied soils on pre-Quaternary limestone, dolomite, and marl on the island of Sardinia, which sometimes receives dust from Africa at the same time as Mallorca (Fig. 1). From their data, we computed Zr/Hf, La/Yb, Cr/Sc, and Th/Ta for the soils and the carbonate rock substrates they studied. Results indicate that these values are similar among all the soils, regardless of substrate type or age, but soil values differ from those in the subjacent rocks. Furthermore, Zr/Hf, La/Yb, Cr/Sc, and

Th/Ta for the Sardinian soils are similar to the Mallorcan soils reported here. These observations suggest a common African dust source for soils on both islands. The broader implication of these results is that terra rossa soils (as well as other soils) in much of the Mediterranean region (Fig. 5) may be dust-derived, or at least have an important dust component. This supports the conclusions of many earlier workers, including Kubiëna (1953), Yaalon and Ganor (1973), MacLeod (1980), Duchaufour (1982), Jackson et al. (1982), Rapp (1984), Yaalon (1987, 1997), Pye (1992), Nihlén and Olsson (1995), Durn et al. (1999), and Delgado et al. (2003). The results also broadly support the model for varying African dust additions to European soils recently proposed by Stuut et al. (2009).

The results reported here also support the dust generation and flux model results reported by Mahowald et al. (2006) and shown in Fig. 3. In both glacial and interglacial periods, model results suggest that African dust flux to southwestern Europe, including Mallorca, is modest (5–10 $g/m^2/yr$). This magnitude of dust flux is perhaps enough to be detectable by detailed studies of the geologic record, but not likely of a sufficient magnitude to generate loess deposits that are apparent as such in the field. Pye (1992) reaches a similar conclusion after studying terra rossa soils and dust flux on the island of Crete. Nevertheless, Rose et al. (1999) report thin (~50 cm) loess deposits near Calo d'es Cans in northern Mallorca. This loess contains significant (~30%) amounts of sand, which suggests a possible local source. The origin of this loess could be investigated using some of the methods applied in the present study.

The paleosol-eolianite record on Mallorca suggests that African dust flux to the Mediterranean region may have been an important process for several interglacial–glacial cycles, over the past several hundred thousand years. The eolianite sections at S'Estalella and Campo de Tiro/Palma airport likely span all or most of the Late Pleistocene and part of the Middle Pleistocene. If the age estimates of Nielsen et al. (2004) are correct, the eolianite section at Els Bancals likely spans much of the later part of the Middle Pleistocene, perhaps back to 400,000 yr. Similarity of paleosol compositions in all these sections suggests that African dust flux has been an important component of the climatology and sedimentary cycles of the region for a long time. Because eolianites were likely deposited on Mallorca primarily during glacial periods, the paleosols probably represent interglacial or interstadial periods, when dust fluxes may have been somewhat lower (Fig. 3). During glacial periods of active dune or sand sheet deposition on Mallorca, fine-grained African dust was likely to stay in suspension or be diluted in thick carbonate sand deposits. Thus, records of higher dust flux, during glacial periods, could be sought in deep-sea sediment records of the western Mediterranean, as proposed by Rapp (1984).

11. Conclusions

(1) Eolianite-paleosol sequences on Mallorca span much of the middle and late Quaternary, thus providing a record of several glacial–interglacial cycles.
(2) Eolianites on Mallorca are of very high purity, which means that if they are to become a source for the development of red soils, then much carbonate dissolution must have taken place.
(3) Ratios of immobile trace elements (Zr/Hf, La/Yb, Cr/Sc, and Th/Ta) provide a means for "fingerprinting" sediment sources. Using these values, African dust and the non-carbonate component of eolianites on Mallorca are distinct from one another.
(4) Paleosols on Mallorcan eolianites have Zr/Hf, La/Yb, Cr/Sc, and Th/Ta that differ from the subjacent eolianites, but fall within, or close to the range of values for African dust, indicating that this far-traveled sediment is the most important soil parent material.
(5) Si/Al values in paleosols indicate that although African dust is the most important soil parent material, local eolianite has made a small contribution to the soils, probably mostly in the form of detrital quarz.
(6) Trace element ratios in Mallorcan soils and paleosols are similar to soils on carbonate substrates on the island of Sardinia, suggesting a common African dust source.
(7) African dust may explain the origin of many terra rossa soils on relatively pure carbonate hosts around much of the Mediterranean region.
(8) The evidence for African dust in Mallorcan soils, but lack of evidence for extensive thick loess deposits, agrees with global dust modeling estimates that suggest that African dust to the western Mediterranean region is measurable but modest.

Acknowledgements

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