

This manuscript has been submitted for publication in Earth Surface Processes and Landforms. This preprint has not undergone peer-review and subsequent versions of the manuscript may differ from this version. If accepted, the final version will be available via a DOI link on this page. Please contact the corresponding author by email with any queries – mstyllas@gmail.com Prepared for EarthArxiv on October 19th 2021.

The relative contributions of weathering and aeolian inputs to postglacial formation of Mediterranean alpine loess

MICHAEL STYLLAS^{1,2}, CHRISTOS PENNOS^{3,5}, MATHIEU GHILARDI⁴, AUREL PERSOIU^{5,6},
LAMBRINI PAPADOPOULOU⁷, NIKOLAOS KANTIRANIS⁷, ELINA AIDONA⁸,

¹ *Geoservice Ltd., Eirinis 14 Street, 55236, Thessaloniki, Greece (Email: mstyllas@gmail.com)*

² *Ecole Polytechnique Federal de Lausanne - EPFL, Lausanne, CH-1015, Switzerland*

³ *Department of Geography, University of Bergen, Fosswinckels Gate 6, 5007, Bergen, Norway*

⁴ *CEREGE - UMR 7330 CNRS Europôle de l'Arbois BP80 13545 Aix-en-Provence CEDEX 04 – France*

⁵ *Emil Racoviță Institute of Speleology, Romanian Academy, Cluj-Napoca, 400006, Romania*

⁶ *Romanian Institute of Science and Technology, Cluj-Napoca, 400022, Romania*

⁷ *Department of Mineralogy, Petrology and Economic Geology, School of Geology, Aristotle University of Thessaloniki, Thessaloniki, 541 24, Greece*

⁸ *Department of Geophysics, School of Geology, Aristotle University of Thessaloniki, Thessaloniki, 541 24, Greece*

1 **ABSTRACT**

2 Between the southern margin of the European loess belt and Sahara Desert, thin and irregularly
3 distributed loess deposits occur in Mediterranean mountains. During the most recent deglaciation, along
4 the Pleistocene-Holocene boundary, the deposition of glacial, periglacial and outwash sediments, was
5 the main local source of Mediterranean alpine loess, whereas proximal alluvial planes comprised a
6 secondary source. The mid-Holocene termination of African Humid Period and subsequent aridification
7 of Sahara Desert occurred simultaneously with a change of the regional climate from Atlantic to
8 Mediterranean-dominated, characterized by frequent episodes of southerly winds. This resulted to a
9 change of the loess source, as deflation of quartz rich silts enriched in Zr during intense episodes of
10 Sahara dust transport became more dominant. Here, a 32cm loess profile from the Plateau of Muses
11 (PM), below the summit of Mount Olympus, Greece, is investigated on the basis of grain size,
12 mineralogy, environmental magnetism and geochemistry. Comparisons of loess samples with glacial
13 and periglacial deposits, enables us to differentiate relative contributions of local sources and
14 allochthonous aeolian inputs. Calcite sand rich in feldspars makes up the glacial and periglacial clast
15 free matrix. In contrast, PM loess is composed by clay and fine silt fractions with minor calcite sand
16 contributions. The mineralogical matrix of loess contains quartz, phyllosilicates and mixed layer clays,
17 while its geochemical composition contains high amounts of detrital Fe-Ti oxides and aeolian
18 transported Al and Zr. Based on the multi-proxy approach applied here, the loess profile is partitioned
19 in three layers. Holocene average deposition rates (~2.5 cm/ka) broadly agree with modern Sahara dust
20 deposition (~2.0 cm/ka) and long-term postglacial Mediterranean mountain denudation rates (~0.5
21 cm/ka). Such low rates provided ample time for post depositional modifications, such as decalcification,
22 deferrification and removal of K, evident from the trends of chemical weathering proxies Ca/Sr, Fe/Ti
23 and K/Rb, respectively.

27

28 **Keywords** Loess, alpine plateau, glacial debris, Sahara dust, pedogenesis, chemical weathering, Mount

29 Olympus, Greece.

INTRODUCTION

The most recent deglaciation of the Mediterranean mountains between 12 and 9.5 ka BP resulted to deposition of large sequences of glacial, periglacial and outwash sediments that were mainly confined in the highest valleys of the massifs (Hughes and Woodward, 2016; Oliva *et al.*, 2018; Allard *et al.*, 2020). Antecedent to glacial retreat was the deposition of loess and subsequent formation of alpine soils on moraines, plateaus and outwash plains (e.g. Muhs 2007). Synergistic to the in-situ genesis of alpine soils, is the deposition windblown dust, which results to the formation of alpine loess soils (Muhs and Benedict, 2006; Küfmann, 2008; Lawrence *et al.*, 2013; Drewnik *et al.*, 2014; Yang *et al.*, 2016; Gild *et al.*, 2018). The multi-proxy analysis of postglacial alpine soils and loess deposits provides insights on the local climate - weathering relations (Egli *et al.*, 2010; Egli 2014; Yang *et al.*, 2016) and further determines the rate of geomorphic processes, such as landscape denudation. Furthermore, the study of deflated sediments within alpine soils and loess can provide insights on the local and regional atmospheric circulation patterns, reflected by the depositional dynamics of aeolian dust (e.g. Muhs *et al.*, 2007).

In the Mediterranean region, the formation of loess is influenced to a large extent by its proximity to Sahara Desert (Pye, 1995; Goudie and Middleton, 2001; Stuut *et al.*, 2009), but also to other local sources of deflated material such as glacial moraines and periglacial slopes, alluvial planes and sand dune fields (Amit *et al.*, 2020; Lehmkuhl *et al.*, 2021). Sahara dust comprises the parent material of red soils, formed on fluvial and coastal terraces (e.g. Durn, 2003), whereas on the Mediterranean mountains, thin drapes of loessic alpine soils form on karstic depressions, glacial moraines, outwash plains and glaciofluvial deposits (e.g. Rellini *et al.*, 2009). Similarly, alpine loess soils with significant Sahara dust subadditions are found past the margin of Mediterranean basin, as for example in Carpathian Basin (Varga *et al.*, 2016) and the Northern Calcareous Alps (Küffman *et al.*, 2008). In these locations, the formation of alpine soil loess sequences consists mainly of windblown silt rich in quartz and heavy minerals of North African crustal origin. In alpine settings with carbonate bedrock lithologies, the aeolian deposition is contemplated by the transport and deposition of carbonate weathering debris and

55 carbonate dissolution products, which form an insoluble residue incorporated in the pedogenic
56 sequences (Durn 2003; Varga *et al.*, 2016). Evidence from marine and lacustrine sediment archives from
57 the Mediterranean, show an intensification of Sahara dust transport after the termination of the African
58 Humid Period (AHP) around ~6 ka BP, due to increasing deflation of silt from desiccated areas in
59 northern Africa (e.g. Jimenez-Espejo *et al.*, 2014; Ehrmann *et al.*, 2017). Also, Holocene climatic
60 reconstructions from ice cave deposits in Romania, suggest that the end of the AHP coincided with a
61 shift in the winter climate of southeast Europe from Atlantic to Mediterranean dominated (Persoiu *et*
62 *al.*, 2017).

63
64 This study presents for the first time a precise characterization of Mount Olympus loess, by combing
65 grain size distributions, mineralogical and geochemical compositions, and magnetic properties. An
66 attempt to discern the relative contributions of local erosion, aeolian inputs and post depositional
67 weathering since the onset of the most recent deglaciation (~12 ka BP) is undertaken through a
68 multiproxy approach. Mount Olympus loess gives a unique opportunity to assess the contributions of
69 local and allochthonous sources and the paleoclimatic reconstruction potential of similar deposits, which
70 are widespread within modern Mediterranean paraglacial environments.

71 72 **SETTING**

73 **Climate**

74 Mount Olympus is the highest mountain of Greece, rising 2918 meters above the Aegean Sea (Fig. 1).
75 In the lower part of the mountain, Mediterranean type climate prevails with wet winters and generally
76 dry summers. Wet winters are linked to cyclogenesis in the Aegean Sea basin that results from enhanced
77 mid-latitude westerlies (Fig. 1 pattern B) and the influence of Atlantic climate (Xoplaki *et al.*, 2000).
78 This pattern was dominant during the first part of the Holocene (Peyron *et al.*, 2017). Dry winters are
79 associated with outbreaks of northerly continental cold and dry airflows (Fig. 1 pattern B) funneling
80 through the large fluvial valleys exiting on the Aegean Sea (Rohling *et al.*, 2002), which are connected
81 to the presence of high-pressure systems over the northern Balkans and/or Siberia (e.g. Xoplaki *et al.*,

2000; Bartzokas *et al.*, 2003). This pattern was persistent throughout the Holocene, when short periods of cold and dry winters linked to the intensification of Siberian High (e.g. Rohling *et al.*, 2002; Marino *et al.*, 2009) and resulted in Mediterranean rainfall minima associated Sahara dust transport episodes (Zielhofer *et al.*, 2017a). The transport of Sahara dust in the North Aegean occurs today under strong southerly (Sirocco) winds (Fig. 1 pattern C) during the winter and spring (Nastos, 2012), but there is lack of evidence of how the southerly winds outbreaks evolved during the Holocene. However, the study of Mediterranean alpine loess archives, where the Sahara dust signal is not blurred by erosion, reworking and pedogenesis, can provide valuable information on the tempo of southerly warm and moist wind outbreaks and their impacts on different ecosystems.

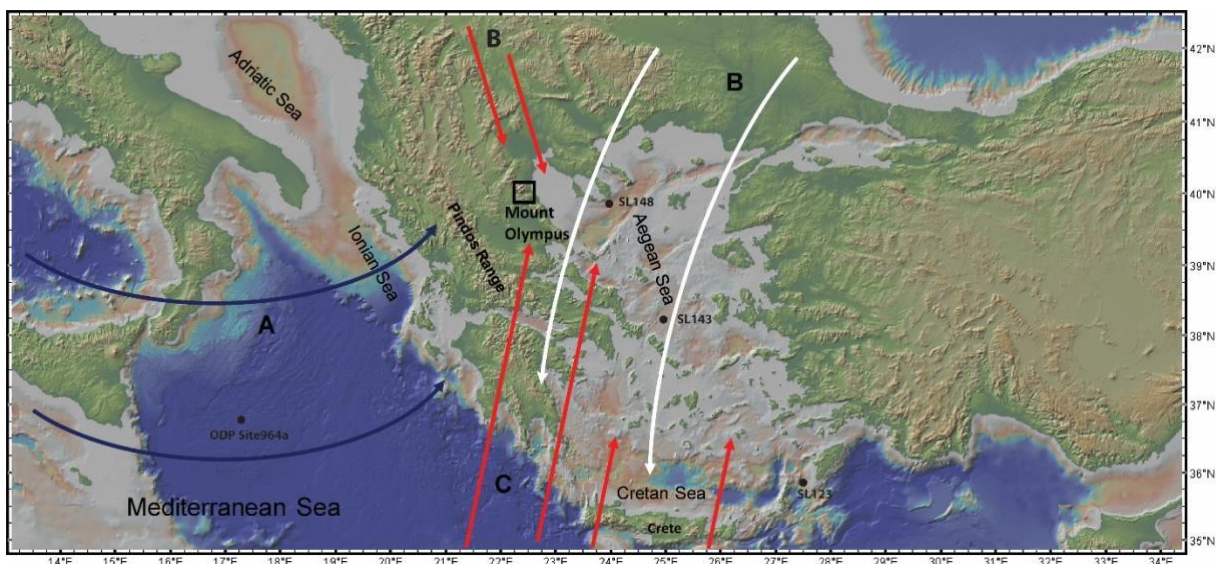


Fig. 1. General setting of Mount Olympus, in relation to the dominant atmospheric patterns. (A) Mid-latitude westerlies causing Mediterranean cyclogenesis and wet winters over the study area. (B) Cold and dry air masses of polar origin, resulting in low winter precipitation. (C) Southerly sirocco winds favoring intense transport Sahara dust and heat convection to the study area. Topographic and bathymetric background provided by Geomapapp (<http://www.geomapapp.org>).

Glacial erosion

The geologic structure of Mount Olympus involves a stratigraphic upwards sequence of Triassic, and

Lower Cretaceous to Eocene metacarbonates, uplifted since the late Miocene along a major NW – SE trending frontal fault (Fig. 2A) (Nance, 2010). During uplift, deposition of erosional products along the eastern (marine) and the western (continental) piedmonts occurred (Fig. 2A). Their Quaternary counterparts include thick sequences of glaciofluvial and alluvial fan deposits with intercalated soils, exposed along the main river valleys and the frontal fault scarp (Fig. 3 in Smith *et al.*, 2006). During the Last Glacial Maximum (LGM), between 28 and 24 ka BP (Allard *et al.*, 2020), an ice cap covered Mount Olympus’ highest cirques and upland plateaus extending to elevations of ~ 2000m (Kuhlemann *et al.*, 2008). The post LGM retreat was followed by a Late Glacial (LG) glacier expansion at ~15 ka BP that was confined in the highest cirques at elevations above 2200 m (Styllas *et al.*, 2018).

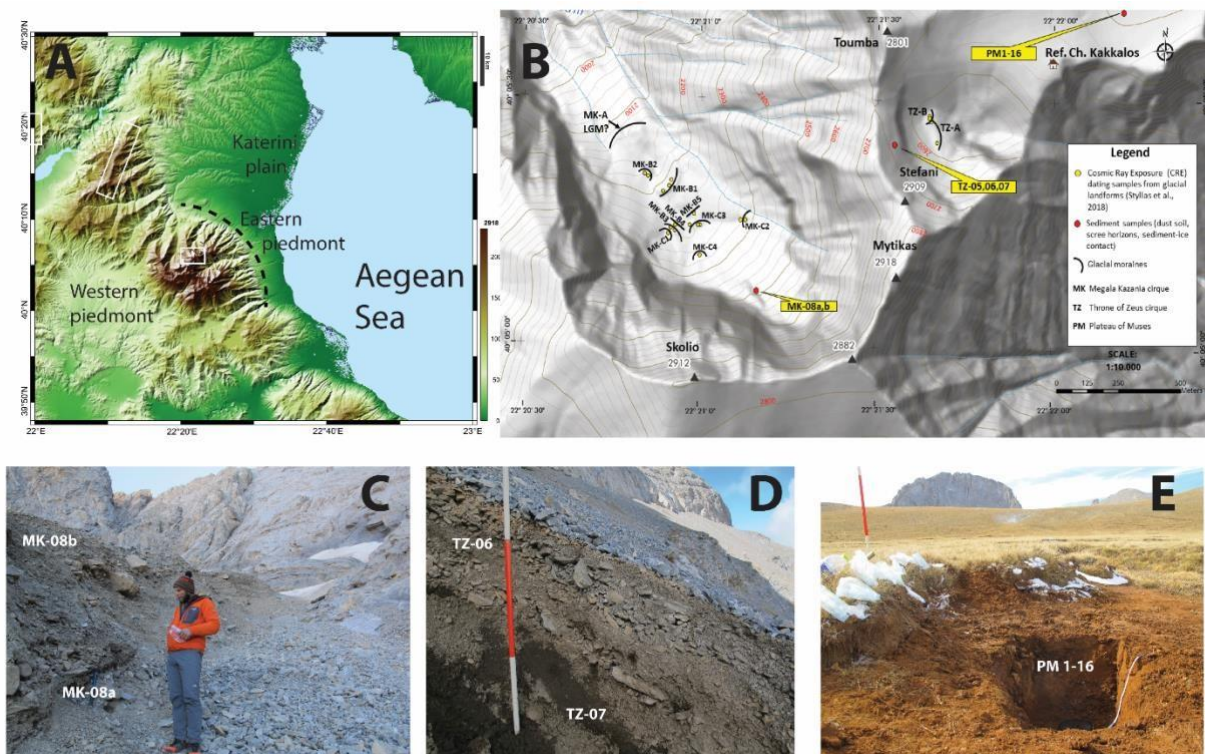


Fig. 2. Geomorphological properties of the study area. A. Setting of Mount Olympus (white box), with the two piedmonts along the marine and continental sides, the main frontal fault (dashed line) the mountaintop of adjacent Pieria Mountains (white box) and Katerini alluvial plane. B. The highest cirques of Mount Olympus with the respective locations of dated moraines (Styllas *et al.*, 2018) and the sampling locations considered in this study (yellow boxes). C. Upper left inset shows and the red box shows the geographical position of the map. (C) Sediment samples from clast free horizons of a lateral moraine in Megala Kazania (MK) cirque. (D) Stratified scree under

117 the rock face of Stefani (2909m) in the Throne of Zeus (TZ) cirque. (E) Postglacial / Holocene loess accumulation
118 in the Plateau of Muses (PM).

119
120 The most recent moraine stabilization phase common to the highest cirques of Megala Kazania (MK)
121 and Throne of Zeus (TZ) (Fig 2B), occurred at 12 ka BP and has been tentatively ascribed to Younger
122 Dryas (Styllas *et al.*, 2018), with thick accumulations of sediments below MK and TZ headwalls. These
123 occur either as stratified scree slopes, or as glacial till forming lateral moraines (Fig. 2B & C). Away
124 from the main depocenters of the most recent glacial activity, periglacial movements, and outwash
125 deposits along with the contribution of aeolian deflation of glacially grinded carbonate sands and silts
126 become dominant.

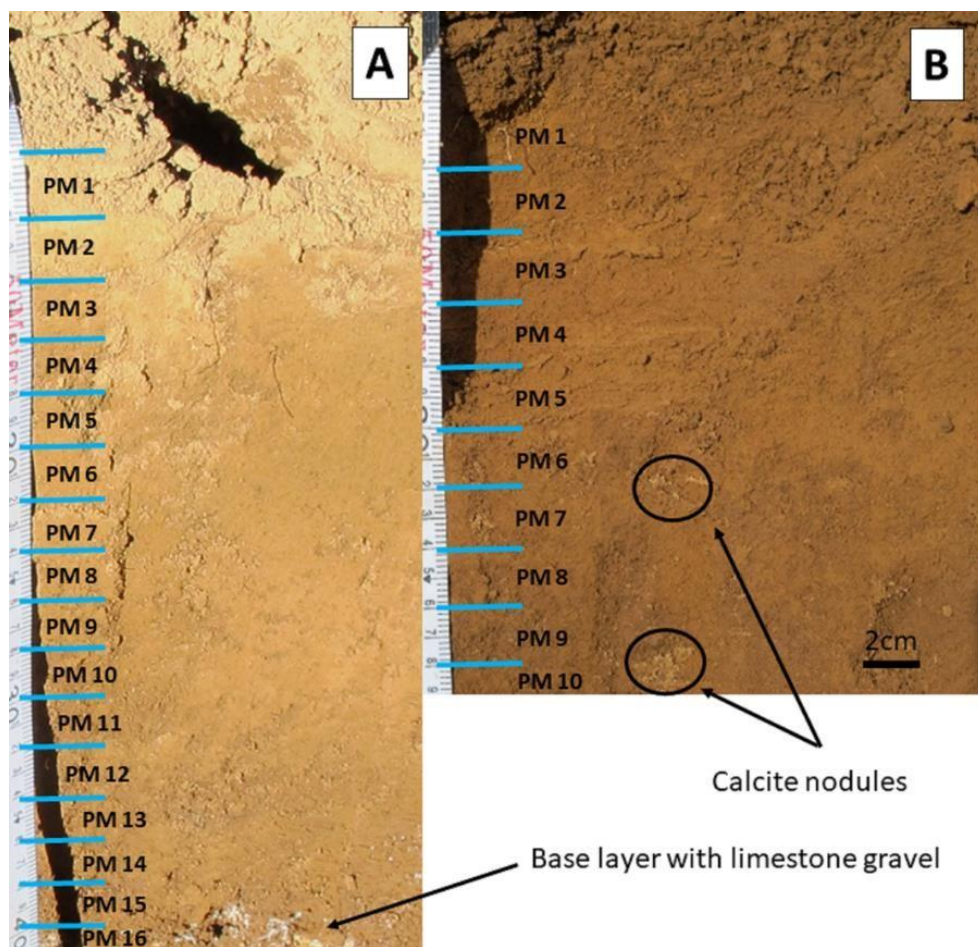
127 128 **Sources of aeolian dust**

129 The study of north Aegean core SL-148 marine sediments, suggests that the major sources of aeolian
130 dust transport in the North Aegean Sea (Fig. 1) is Sahara Desert (Hamann et al., 2008). Direct
131 observations from Mount Olympus over the past 15 years confirm the existence of severe episodes of
132 Sahara dust deposition during the winter and spring, where it forms distinct layers within the snowpack,
133 whereas measurements of Sahara dust deposition rates in Crete (Fig. 1), range between 0.66 and 2.14
134 cm/ka (Niéhlen et al., 1995). Additional dust sources are the eroded mountaintops of the ophiolitic Pieria
135 Mountains and the alluvial plain of Katerini (Fig. 2A). The sediments of Katerini alluvial plains have
136 mainly ophiolitic rock formations, as they drain Pieria Mountains (Krahtopoulou and Veropoulidou,
137 2017). Transport of deflated material from the alluvial plains can occur during periods of northern wind
138 outbreaks (Fig. 1, pattern B) and by summer convection, triggered by pronounced temperature gradients
139 between the coast and the upper mountain, which can reach 8 - 10° C/km (Styllas *et al.*, 2016).

140 **Plateau of Muses loess**

141 The Plateau of Muses (PM) is a planar depositional surface at 2600m, bounded by the TZ cirque lateral
142 moraine ridge to the south, and by several gentle sloping glacially eroded peaks along its northern, eastern

143 and western margins (Fig 2E). The Plateau of Muses extents 0.8 km² and is covered by unconsolidated
144 glacial and periglacial sediments. Periglacial features such as solifluction beds are present below the exposed
145 bedrock of the surrounding peaks, while patterned grounds exist along its topographically lower surface
146 (Styllas et al., 2018). These features are tentatively considered to have formed during cold intervals over the
147 last ~12 ka BP, following the deglaciation of TZ cirque, but may be still active today as the permafrost
148 elevation of the region is placed at 2700 m (Dobiński, 2005). The formation of PM is the result of the
149 combined action of glacial scouring and karstic dissolution. The low relief in combination to the elliptical
150 to circular plan shape of the plateau, points to a doline type karstic depression filled with glacial and
151 periglacial sediments with a thickness between 4 to 10m (unpublished data from geophysical survey). The
152 surface layer (> 35cm) of PM sedimentary sequence is composed by a red to yellow homogenous fine-
153 grained accumulation, with its basal part composed by glacial and/or snowmelt outwash limestone sand and
154 gravel, mixed with silty sediments (Fig. 2E, Fig. 3).



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156 **Fig. 3.** Pictures of the PM 32cm soil loess profile with the respective discrete samples taken every 2cm.

157 **HYPOTHESIS AND STUDY DESIGN**

158 Based on the considerations regarding: the onset of deglaciation Mediterranean mountains at ~12 ka BP
159 and the termination of the African Humid Period at ~6 ka BP, this study, based on a suite of analytical
160 methods applied to samples from Mount Olympus, Plateau of Muses loess, is challenging the hypothesis
161 that the evolution of Mediterranean alpine loess, occurred along three distinct phases:

- 162 i) The postglacial phase, between ~12 – 10 ka BP, when the alpine domain of Mediterranean
163 mountains entered a phase of postglacial adjustment with large volumes of glacial debris
164 available for deflations, transport and deposition under the influence of snow, water and
165 wind.
- 166 ii) The early to mid-Holocene phase between 10 and 6 ka BP when, under a warming and
167 seasonal Mediterranean climate (Peyron et al., 2017), the formation of loess was mainly
168 sourced by local glacial sediments and expanding alluvial planes in lower elevations. iii)
169 The mid to late Holocene from 6 to 0 ka BP, where following the termination of the African
170 Humid period and desiccation of Sahara Desert, along with a change of regional winter
171 climate from Atlantic to Mediterranean, increasing amounts of Sahara dust reached the
172 Mediterranean mountains during episodes of southerly advection.

174 **ANALYTICAL METHODS**

175 **Grain-size analyses**

176 Grain-size analyses were performed on 21 samples. Five samples were retrieved from distinct clast free
177 horizons of the MK and TZ stratified scree deposits and sixteen samples from the PM loess sequence
178 (Fig. 2B). Samples were wet-sieved through a 350 µm sieve and were then analyzed with a Mastersizer
179 3000 laser diffraction particle size analyzer (Department of Earth Science, University of Bergen,
180 Norway), with a sensitivity of 0.01 – 350 µm, to define the bulk grain-size distributions (GSD) of the
181 fine sand to clay fractions. GSD statistical analyses were performed with MATLAB Curve Fitting Lab

182 (CFLab), which performs curve fitting on sediment grain size distributions using Weibull Probability
183 Distribution Functions (Wu *et al.*, 2020).

185 **Chemical methods (XRF) and mineral analysis (XRD)**

186 All samples were analyzed for their bulk mineralogy and geochemistry. The relative elemental
187 composition was determined by X-ray fluorescence using an ITRAX core scanner in the Department of
188 Earth Science of the University of Bergen in Norway. One cubic centimeter of the finer (<350 μm)
189 fraction of the samples was air-dried, filled into sample cups and compacted by hand. Four units of 21
190 sample cups were mounted on sample holders and measurements with the ITRAX XRF core-scanner
191 were performed using a Mo-tube, which can detect a wide range of elements from Al to U (Croudace *et*
192 *al.*, 2006). Counting time was 10 s and power supply at 30 kV/55 mA. XRF spectra were translated into
193 element counts by mathematical peak fitting using Q-spec software (Croudace *et al.*, 2006). Major and
194 trace elements detected by the XRF analyses include Zr, Fe, Ti, Ca, Sn, Mn and Rb along with the main
195 elements.

196 X-ray diffraction analyses for the identification of mineral phases of the bulk samples from MK, TZ and
197 PM, were performed in the Centre de Recherché et d'Enseignement de Géosciences de l'Environnement
198 (CEREGE) in Aix en Provence, France using a Philips diffractometer (PW1800) with graphite
199 monochromator, Co radiation functioning in 40 kV, 40 mA. Two samples from the surface and base of
200 the PM sediment profile (PM1 and PM15), were additionally analyzed for the clay (<2 μm) mineralogy
201 by subjecting the samples to ethylene glycolation, followed by heating for 2 h at 550 °C. The PM bulk
202 samples were further processed for their semi-quantitative mineralogical composition, expressed in
203 weight percent (wt%) in the Department of Mineralogy, Petrology and Economic Geology, School of
204 Geology of the Aristotle University of Thessaloniki in Greece, by using a Philips diffractometer
205 (PW1710) with Ni-filtered CuK α radiation on randomly oriented samples. The corrections of the wt%
206 mineralogical compositions were performed using external standard mixtures of the main minerals
207 present scanning under the same conditions, while MAUD-Material Analysis software applied for full

208 pattern Rietveld refinement (Lutterotti *et al.* 2007). Applying these successive steps for the calculation
209 of mineralogical composition, precision of 1% was achieved (Kantiranis *et al.*, 2011).

211 **Petrographic methods (SEM)**

212 Petrographic analysis of the PM samples was conducted in the Scanning Microscope Laboratory imaging
213 (Department of Mineralogy, Petrology and Economic Geology, School of Geology of the Aristotle
214 University of Thessaloniki in Greece), focusing on the textural and fabric configuration of the PM
215 sedimentary sequence. The petrographic analyses were performed with a JEOL JSM-840A (Tokyo,
216 Japan) Scanning Electron Microscope (SEM) equipped with an Energy Dispersive Spectrometer - EDS
217 (INCA 250, Oxford) with 20 kV accelerating voltage and 0.4 mA probe current. Pure Co was used as
218 an optimization element. For the SEM observations, all samples were coated with carbon, with an
219 average thickness of 200 Å, using a vacuum evaporator JEOL-4X. Backscattered electron images (BSE)
220 were taken to detect areas of different chemical composition, since the brightness of the BSE image
221 tends to increase with the atomic number of an area. Matrix corrections were made following the
222 standard procedures using a combination of silicate, oxides, and pure metals (wollastonite for Ca and
223 Si, jadeite for Na, orthoclase for K, corundum for Al, periclase for Mg, metallic Fe and Ti for Fe and
224 Ti).

226 **Magnetic susceptibility (MS)**

227 The PM samples were subjected to magnetic susceptibility analyses. The samples were sieved to remove
228 all the impurities and packed in cylindrical plastic boxes (2 x 2 x 2 cm). The laboratory measurements
229 of the volume-specific magnetic susceptibility (κ , SI units) have been performed using the Barrington
230 MS2B sensor at low and high frequency (0.465 and 4.65 kHz respectively). The samples were weighted
231 before each measurement. Here the results are expressed as mass-specific magnetic susceptibility (χ ,
232 $10^{-8} \text{ m}^3/\text{kg}$). During the measuring procedure, every sample was measured at least 3 times and the average
233 value was assigned as a measurement. Two air measurements before and after the samples' measurements
234

were performed. Additionally, frequency dependent susceptibility ($\chi_{FD}\%$) was calculated according to Dearing et al. (1996) [$\chi_{FD}\% = 100(\chi_{LF}-\chi_{HF})/\chi_{LF}$].

RESULTS

The PM sediment profile (Fig. 2E) was excavated in the topographical lowest part of the plateau and displays different color and sediment texture characteristics compared to the MK glacial till and TZ scree profiles (Fig. 2 C & D). In the middle of PM profile, at depths between 18 and 16cm individual secondary carbonate nodules (calcretes) are present (Fig. 3).

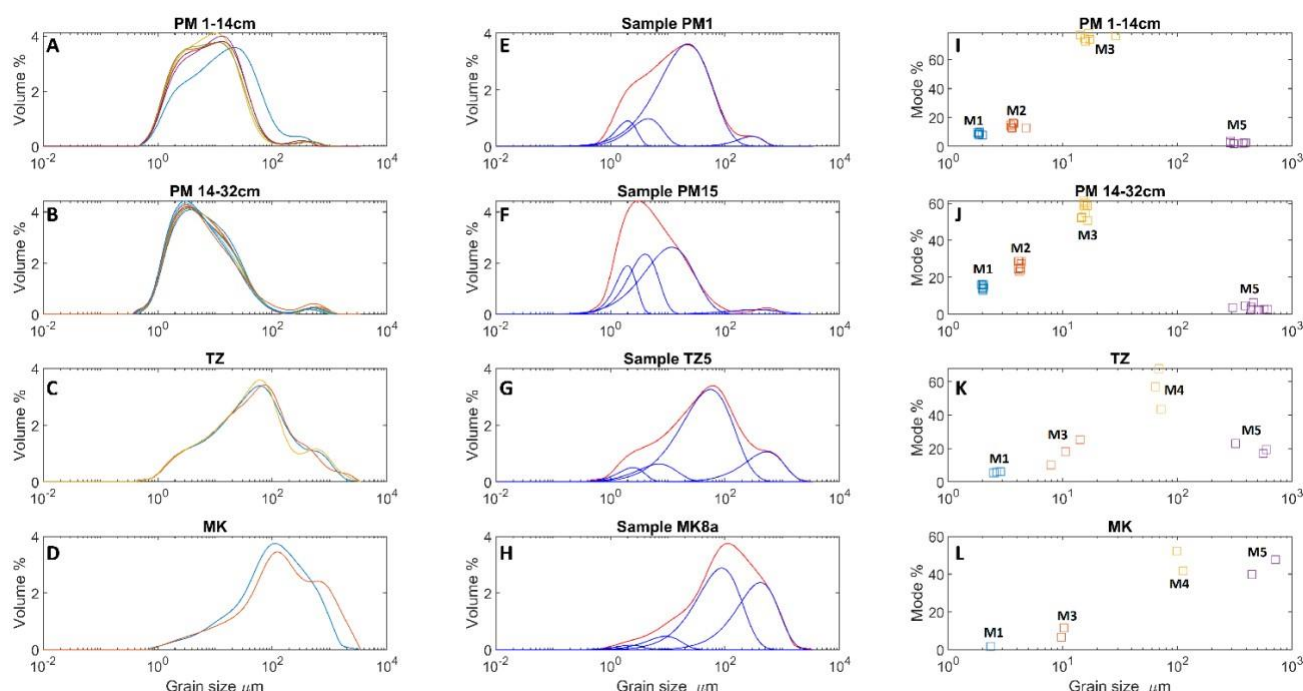
Grain-size analysis

The grain size distribution (GSD) curves of all samples are polymodal and demonstrate different shapes (Fig. 4A to D) between the different environments (PM, MK and TZ). In total, five grain size modes (M1 to M5) were mathematically derived through the application of CFLab curve fitting algorithm. In all cases, fitting degrees were >99% and fitting residuals were <0.1%, indicating excellent fitting of the raw GSD curves (Fig. 4 E to H). As shown in Fig. 4, the application of CFLab algorithm makes it easy to determine relatively separated modes. In the case of overlapped modes (for instance, M1 and M2), several trial-and-error fittings were undertaken, and a significant increase of fitting residuals was observed when the minor modes M1 or M5 were ignored. These exercises demonstrated the validity and accuracy of the Weibull probability distribution functions in fitting and partitioning the grain size components of polymodal sediments.

The clast free horizons from MK glacial till and TZ scree deposits are coarser compared to PM loess grain size distributions (Fig. 4 A to D). The GSD curves of PM loess contain four grain size modes (M1, M2, M3 and M5). The finer grain size modes, M1 and M2, are in range of clayey silt and appear either in very low concentrations (<5%) or are entirely absent from MK and TZ samples (Fig. 4I to L). M1 and M2 with respective grain size of ~2 and ~4 μm , account for 40% of the modal composition in the

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lower layer of PM loess. Their contribution is reduced to 25% in the upper layer, with minimum values near the surface (Figs 4A & B). M3 is composed of fine silt and has an average modal size of $\sim 15 \mu\text{m}$ (Fig. 4I to L). M3 is the most dominant mode of PM loess, with average concentrations of $\sim 55\%$ in the lower and $\sim 75\%$ in the upper layers (Fig. 5C). Substantially lower concentrations of M3 occur in TZ (18%) and MK (9%) samples (Fig. 4 K & L). Grain size mode M4 falls in the range of fine sand, with a modal size of $83.5 \mu\text{m}$ and high ($\sim 50\%$) concentrations under the TZ and MK headwalls, while it is entirely absent from PM loess. The coarsest grain-size mode M5 is present in all samples, with an average modal size of $440 \mu\text{m}$. The concentration of M5 is very low ($\sim 5\%$) in PM loess, while the opposite holds truth for TZ and MK samples, where M5 constitutes $\sim 20\%$ and $\sim 44\%$ of the GSD modal composition. The grain sizes of M4 and M5 can be directly linked to frost weathering of the carbonate bedrock. Such observation is in agreement with experimental results of quartz bedrock frost weathering between -12 and $+15^\circ\text{C}$ that yielded grain sizes between 500 and $63 \mu\text{m}$, with a mean grain size of $180 \mu\text{m}$ (e.g. Smith *et al.*, 2002).

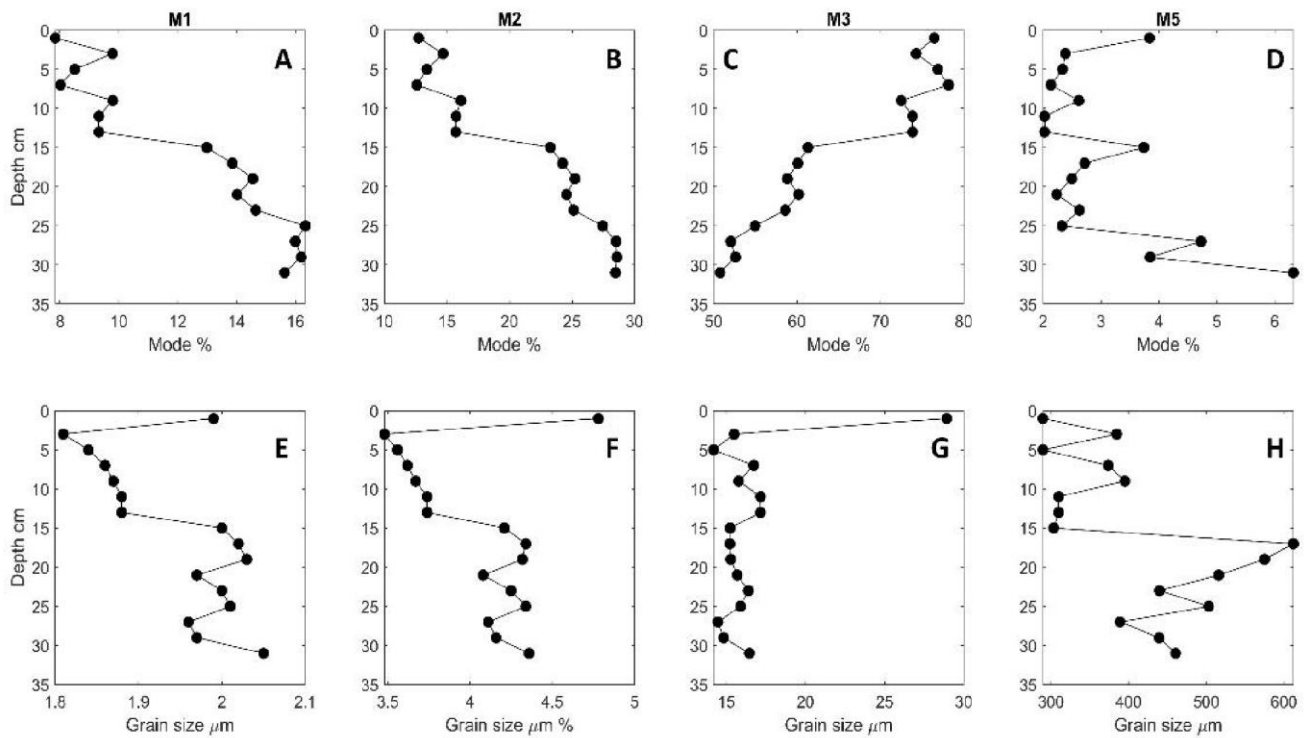


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Fig. 4. Results of the CFLab fitting algorithm with the respective grain size frequency distributions (GSD) and extracted grain size modes of selected samples from: (A) the upper (>14 cm profile depth), and (B) the lower (<14 cm profile depth) layers of the PM loess profile; (C) TZ and (D) MK clast free horizons. (E) Sample PM1 GSD;

278 279 (F) Sample PM15 GSD; (G) near surface samples of TZ and (H) MK clast free horizons. Respective modal percent
279 contributions of: (I) PM loess upper (>14 cm) layer; (J) PM loess lower (<14 cm) layer; (K) TZ and (L) MK samples,
280 respectively.

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282 The depth variations of PM loess fine fractions exhibit a contrasting pattern. M1 and M2 display an
283 upward decrease in their respective grain size and modal concentration (Fig. 5A, B, E & F). The upwards
284 fining of M1 and M2 along with the decrease of their concentrations, may result from either a decrease
285 in the transport capacity from the source, or from a decrease of their in-situ production. A fine mode
286 with a respective grain-size of $\sim 3 \mu\text{m}$, has been reported in almost all European loess representing either
287 regional, or supracontinental sources (Bosq *et al.*, 2020). This may be the case for either M1 and/or M2,
288 with their origin derived either from local sources such the glacial debris and/or the adjacent alluvial
289 plane of Katerini. However, the production of fine particles within the PM catchment from carbonate
290 dissolution and pedogenic processes and their transport to the lower topographic points should not be
291 excluded. The combination of pressure solution in the limestone replacement zone that produces red
292 clays, and of chemical dissolution of limestone bedrock to form coarse sandy debris (e.g. Merino and
293 Banerjee, 2008), are effective mechanisms that can contribute significant amounts of fine particles (i.e.
294 M1 and M2) to PM loess sequence. In contrast to M1 and M2, the modal contribution of M3 is increasing
295 upwards (Fig. 5C), with no changes in its grain-size along the entire profile (Fig. 5G). M3 percent
296 concentration shows a gradual increase in the lower layer, followed by an abrupt (15%) increase between
297 14 and 16 cm of profile depth (Fig. 5C). This most likely reflects an increase of the available material,
298 and/or enhanced transport dynamics from the source area(s). Sandy mode M5 exhibits a coarsening
299 upward trend along the lower layer, intercepted by an abrupt change along the boundary between upper
300 and lower layers, at 16-14 cm depth (Figs 5D & H), with an overall decrease in its modal concentration.

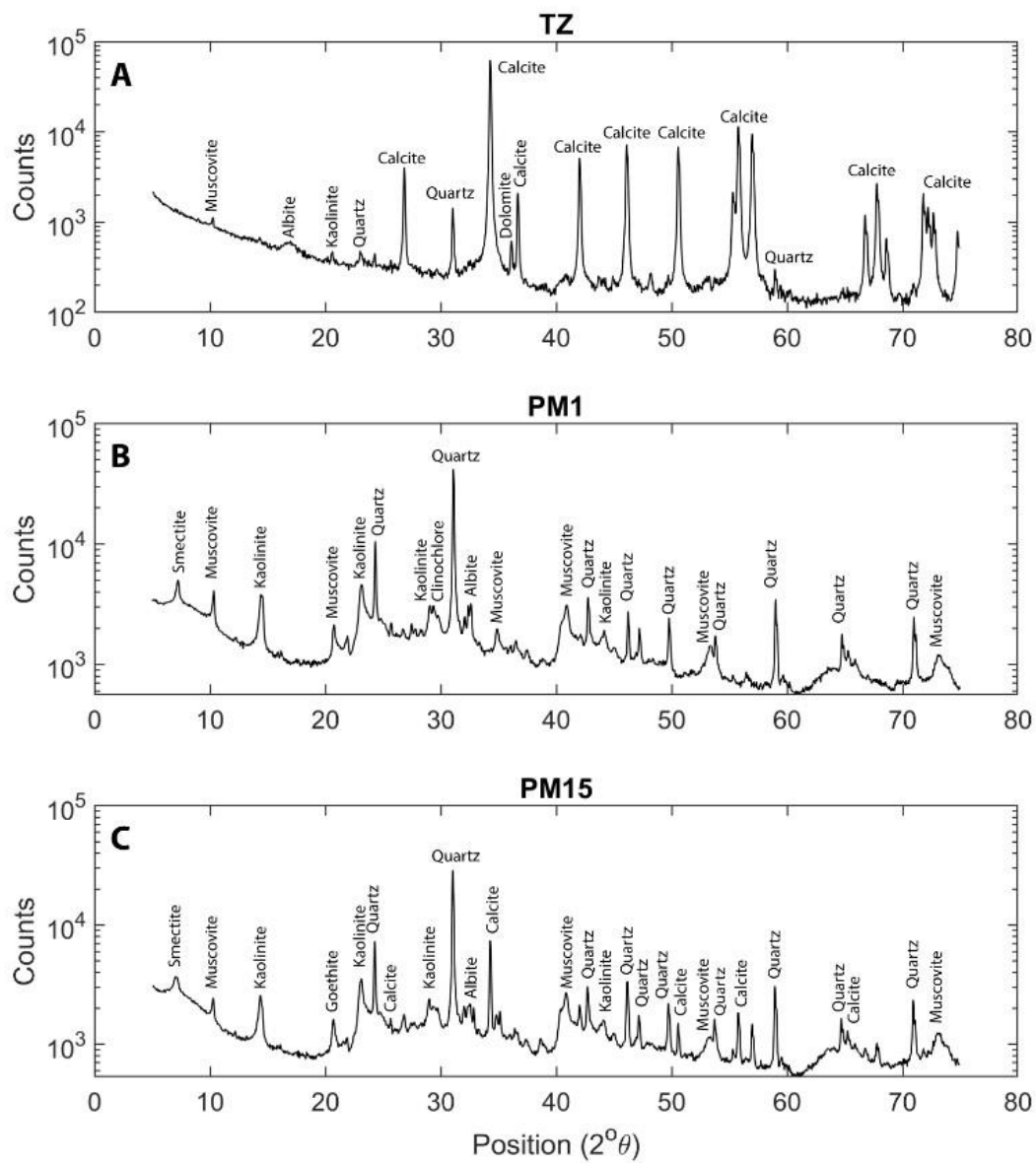


303 **Fig. 5.** PM sediment profile depth variations in the contributions of grain-size modes M1, M2, M3 and M5 (A to 304
 D), and depth variations of the grain-size of modes M1, M2, M3 and M5 (E to H).

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306 Mineralogy

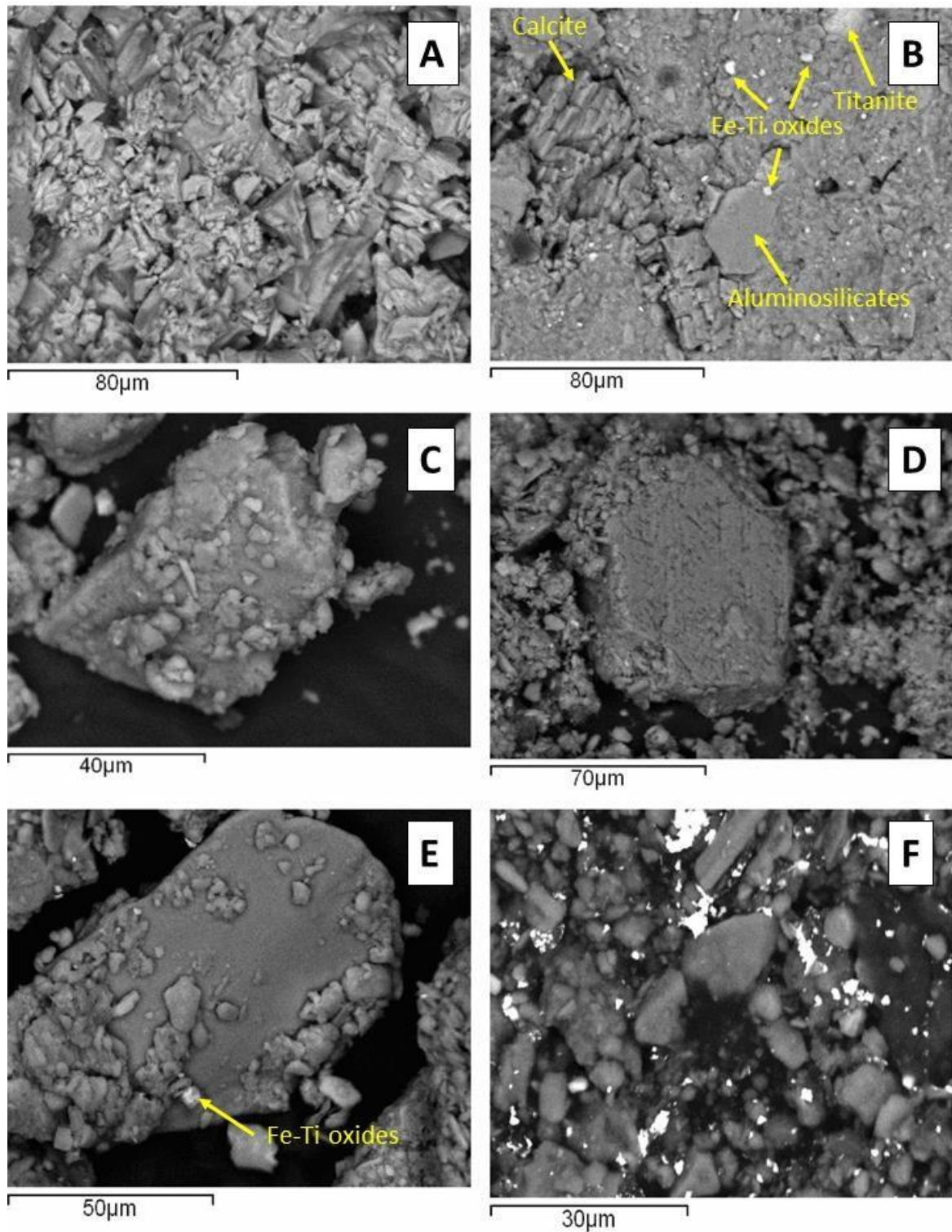
307 The X-ray diffraction analyses of the bulk samples identified a mineralogy that differs between MK, TZ
 308 and PM sediments. The most dominant mineral phases in the clast free material of MK and TZ deposits
 309 are calcite and dolomite (Fig. 6A). Other identified minerals are plagioclase (albite), K-feldspars and
 310 clays. On the contrary, the bulk mineralogical composition of PM loess exhibits a richer suite of mineral
 311 phases (Fig. 6 B and C) that include mainly quartz and K-feldspars, plagioclase, iron oxides, micas,
 312 amphiboles and a mixed layer of clays. Calcite is dominant (~50%) only in basal samples PM15 and 16,
 313 along with mixed aluminosilicate faces and Fe-Ti oxides. Quartz and mixed layer clays are the most
 314 dominant mineral phases of samples PM1 to PM14 with average wt% concentration of ~80%, with
 315 plagioclase, feldspars and mica represent the remaining 20%.



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317 **Fig. 6.** Contrasting patterns of X-ray diffraction results for the sediments of (A) TZ and MK stratified scree clast

318 free horizons and (B) and (C) from PM surficial and basal samples PM1 and PM15.



318

319 **Fig. 7.** SEM images from selected samples of PM loess profile. (A) Calcite from basal sample PM 16. (B) Mixed
 320 phase of aluminosilicates with calcite and Fe-Ti oxides and titanite from basal sample PM 16. (C) K-feldspar. (D)
 321 Albite. (E) Surface sample PM1 aggregate of aluminosilicates and Fe-Ti oxides. (F) Quartz grains from sample
 322 PM3.

323 SEM image analyses of PM loess samples, illustrate the dominance of mixed phases, composed of
324 aluminosilicates, clay minerals and Fe-Ti oxides. These mixtures form aggregates present along the
325 entire profile (Figs 7A, B, E). Fe and Ti are low mobility elements especially in oxidizing environments
326 with pH values above 4.5, so they are not affected by element translocation during weathering (Buggle
327 *et al.*, 2008). Their presence throughout PM loess profile suggests a detrital origin from bedrock
328 formations. Highly magnetic Fe–Ti oxide minerals, such as magnetite and titanomagnetite, commonly
329 intergrown with hematite, ilmenite and ilmenorutile, form originally in igneous rocks during initial
330 cooling (e.g. Reynolds *et al.*, 2001) and here must have been a compartment of Mount Olympus
331 metacarbonates, during the initial stages of their formation. Fe-Ti oxides are incorporated with
332 aluminosilicates and mixed layer clays within aggregates of variable grain sizes (Fig. 7B), that likely
333 form during post depositional processes, but are not represented in the grain size distributions, since they
334 were chemically dissolved during samples preparation. SEM images also suggest that plagioclase
335 (albite) and K-feldspar are also derived from carbonate bedrock weathering, as their grain size (~80 μm)
336 falls close to the grain size of modal class M4 (Figs 7C & D). On the contrary, the ubiquitous presence
337 of quartz in PM loess samples is confined within the fine fractions and suggests a non-detrital origin.
338 Quartz grains display a rounded shape and variable grain sizes smaller than 15 μm (Fig. 7F), which imply
339 their presence within M1, M2 and M3 grain size modes, respectively. The rounded shape of quartz grains
340 depicted from the SEM images, likely is a product of long-range aeolian transport.

341

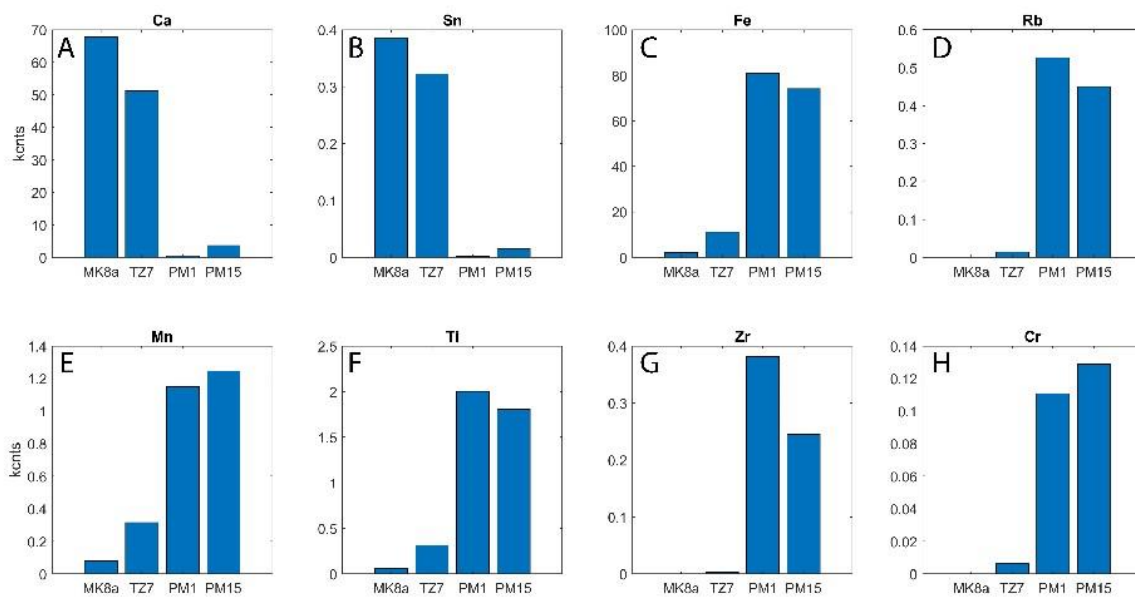
342 Clay mineralogy analyses of surficial (PM1) and basal (PM15) samples identified the dominance of
343 smectite and kaolinite, with lesser contributions of chlorite and illite. In sample PM1 smectite (45%)
344 and kaolinite (35%) are dominant, with lesser contributions of illite and chlorite (10%). Basal sample
345 PM15 has higher smectite contents (65%), with reduced concentrations of kaolinite (25%) and illite and
346 chlorite (5%), compared to the surface. Kaolinite is also traced in MK and TZ samples and can either be
347 of detrital origin or formed from the alteration of other bedrock minerals such as plagioclase (albite), a
348 process that is common in glacial and periglacial environments (e.g. Anderson, 2005). Similarly, high
349 amounts of smectite have been found in alpine soils within proglacial fields and result from the alteration

350 of detrital or aeolian micas (e.g. Egli et al., 2014). XRD analyses suggest the presence of micas in PM,
351 MK and TZ samples, so the alteration of detrital muscovite is likely an ongoing process on Mount
352 Olympus periglacial environments, but the analyses undertaken here cannot provide further conclusions
353 on secondary mineral formation.

354

355 **Elemental composition**

356 The elemental composition of the sediments considered here is expected to reflect their mineralogy and
357 provides an additional tool to discriminate the relative sources of MK, TZ and PM depositional
358 environments. Ca is the dominant element in the MK and TZ samples, while Fe is the most abundant
359 element in the PM samples. Along with Ca, Sn is also present in MK and TZ samples, but barely
360 detectable in the PM loess (Fig. 8B). Sn bearing sulfides exist in metacarbonates in southern Greece and
361 are formed by carbonate replacement during exhumation (e.g. Voudouris *et al.*, 2008). Sn presence in
362 the metamorphosed Cretaceous limestones of Mount Olympus may thus reflect its uplift and exhumation
363 history. The minimal Sn counts in PM samples (Fig. 8B) are explained by the slow mobility of Sn during
364 weathering (Kabata-Pendias, 2001), whereas low Ca counts likely result from decalcification of the loess
365 profile, a process common in other loess deposits (e.g. Bosq *et al.*, 2020). Mn and Rb are also present in
366 MK and TZ samples (Fig. 8 D and E) and appear enriched in PM loess. Mn has been found in manganese
367 rich shale bands of the North Calcareous Alps (e.g. Rantitsch *et al.*, 2003), which may be the case for
368 Mount Olympus, as interbedded shale bands are visually observed within the cirques' headwall
369 Cretaceous limestones. Rb shows similar behavior with K, but is less mobile (Bosq *et al.*, 2020) and is
370 generally hosted K-feldspars, micas and clays (Nesbitt *et al.*, 1980), which constitute integral
371 compartments of Mount Olympus bedrock. Thus, it is safe to consider that the Mn and Rb enrichment
372 of PM loess (Fig. 8D & E) results from sorting during catchment erosion. Besides Fe, Ti, Mn and Rb,
373 PM loess is additionally enriched in Zr and Cr, which appear in negligible quantities in MK and TZ
374 samples (Fig. 8E to H), so their origin from mechanical weathering of bedrock, is implausible. An
375 alternative mechanism for their transport and deposition in PM loess is deflation from distal or local
376 sources, such as the Sahara Desert, the proximal ophiolitic Pieria Mountains and Katerini alluvial plane.



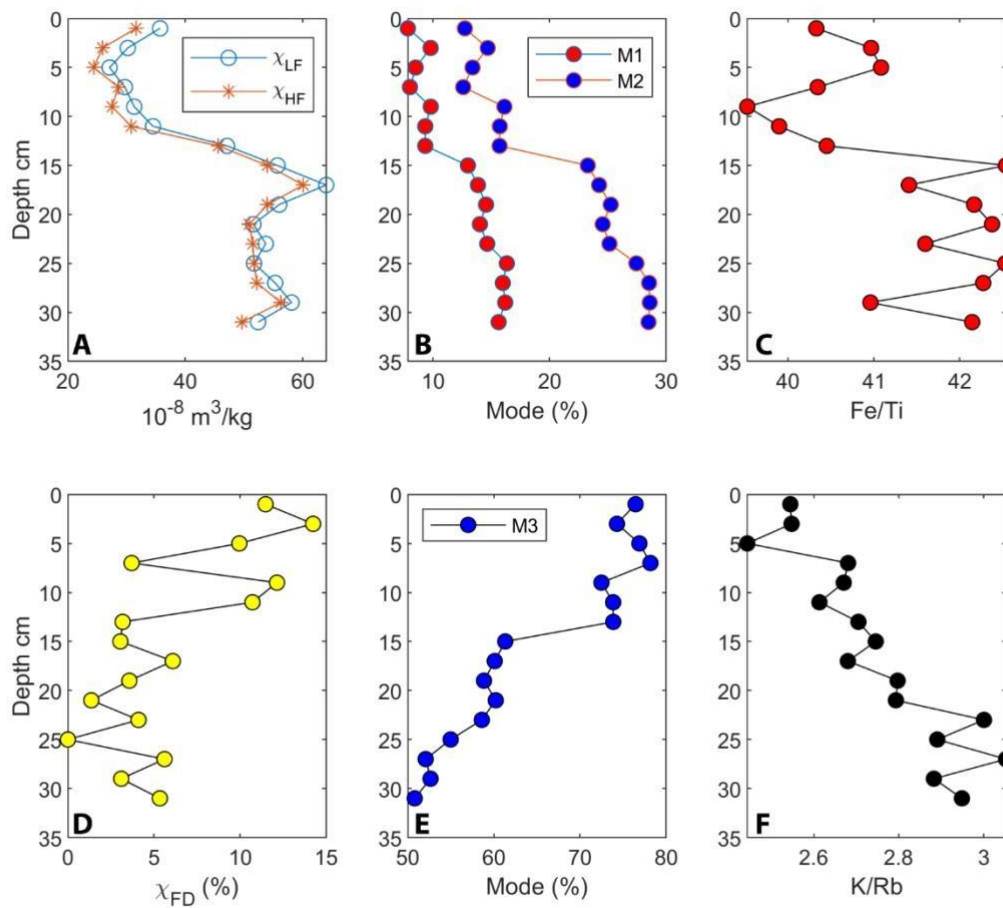
378
 379 **Fig. 8.** Elemental differences between selected samples representing the contrasting sedimentary environments of PM,
 380 TZ and MK.

381

382 **Magnetic susceptibility**

383 The mass-specific magnetic susceptibility profile of PM loess reflects changes in grain size and
 384 mineralogy between stages of loess deposition and when climatic conditions favored pedogenesis. The
 385 lower layer of PM loess (32-16cm) is characterized by higher low frequency magnetic susceptibility
 386 (χ_{lf}) with a mean value of $55 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$. On the contrary, lower χ_{lf} values are found in upper layer,
 387 with a mean value of $36 \times 10^{-8} \text{ m}^3 \text{ kg}^{-1}$ (Fig. 9A) and occur concurrently with a reduction in M1 and M2
 388 concentrations and with a decrease of the Fe/Ti ratio (Fig. 9B & C). Higher χ_{lf} values in the PM loess
 389 lower layer can be attributed to increasing abundance of stable single domain (SD), pseudo single
 390 domain (PSD), and multi domain (MD) grains (e.g. Költringer et al., 2020). The frequency dependence
 391 of magnetic susceptibility (χ_{fd} given in %), has respective mean values of 3.6% for the lower and 8.6%
 392 for the upper layers, respectively (Fig. 9D). Due to its sensitivity to super paramagnetic (SP) particles,
 393 χ_{fd} is often used to identify ultrafine-grained iron oxide formation e.g., magnetite, maghemite, and
 394 hematite during pedogenesis (e.g. Maher, 2011). Dearing et al., (1996), suggests that samples dominated
 395 by multi domain grains (MD), show relatively high χ_{lf} but χ_{fd} close to zero. Values of $\chi_{fd} < 5\%$ are

396 inferred to be typical for samples containing stable single domain (SD) grains, or a very fine (<0.005
 397 μm) SP fraction. Samples dominated by SP grains, show χ_{fd} of 10% and more. In this context, the χ_{fd}
 398 values of the lower layer of PM loess point towards the presence of MD and stable SD grains, while the
 399 upper layer appears to be enriched in SP grains, implying that during deposition of PM loess upper layer
 400 conditions on Mount Olympus were favorable to pedogenesis. The upwards increase of χ_{fd} coincides
 401 with an increase in M3 concentration (Fig. 9 D & E) and with a decrease of the K/Rb ratio (Fig. 9F),
 402 which is a proxy for post depositional weathering of K (Bugge et al., 2011) and likely results from post
 403 depositional weathering of PM loess.



404
 405 **Fig. 9.** (A) Depth variations of low and high frequency magnetic susceptibility, along with M1 and M2
 406 concentrations (B), Fe/Ti ratio (C), frequency depended magnetic susceptibility (D), M3 concentration and
 407 chemical weathering proxy K/Rb (F).

408 **DISCUSSION**

409 **Local weathering**

410 The low correlation between M5 grain size and the Ca XRF counts among all samples ($r = 0.45$, $p <$
411 0.05 , $n = 21$), contrasts the notion that the coarse rich sands, are only produced by physical weathering
412 of bedrock carbonate formations. The low correlation can be attributed to dissolution kinetics and
413 leaching of Ca during disintegration of carbonate bedrock to gravel and sand. Within PM loess sequence,
414 the positive correlation between M1 and M2 concentrations with M5 grain-size ($r = 0.67$, $p < 0.05$),
415 suggests that the production of coarser sandy debris is associated with higher concentrations of fine
416 particles. A physical mechanism that can explain this statistical relationship is the isovolumetric
417 replacement of Ca-rich sand to clay, as proposed by Merino and Banerjee (2008). Thus, the formation
418 of M1 and M2 depends, at least partially, on the availability of carbonate sands of environmental
419 conditions that either favor frost cracking and mechanical disintegration of bedrock, and/or post
420 depositional weathering of the eroded grains.

421

422 On the other hand, between M1 and M2 concentrations, low (χ_{lf}) and high (χ_{hf}) magnetic susceptibility
423 and Fe/Ti ratio, a proxy for iron rich clay variations in alpine soils (Muhs *et al.*, 2001), show high positive
424 correlations ($r > 0.7$, $p < 0.001$) and display higher values in PM loess lower layer. In general, high values
425 of magnetic susceptibility in loess sequences result from enrichment of ferromagnetic minerals during
426 pedogenesis (Zhou *et al.*, 1990; Maher and Thompson, 1992; Heller and Evans, 1995), but the high
427 values (>10) of frequency dependent magnetic susceptibility (χ_{fd}), in the upper layer of PM loess (Fig.
428 9D), contrast this notion. In addition, similar Fe/Ti ratio values between PM loess (42) with MK (36)
429 and TZ (35) samples confirm the presence of Fe-Ti oxides in carbonate bedrock formations and supports
430 their detrital and not pedogenic origin. Thus, the high correlations between χ_{lf} , M1 and M2 with Fe/Ti
431 ratio imply that weathering of bedrock and subsequent release of Fe-Ti oxides, such as titanomagnetite,
432 ilmenite and hematite, control the magnetic signal of PM loess. The formation of coarser aggregates is
433 attributed to post depositional processes, whereas the exhaustion of glacial carbonate debris evident from
434 the reduced concentration and fining of the coarsest grain size mode M5
435 (Fig. 5D & H), resulted, to an unknown extend, in the reduction of M1 and M2 and hence to the lower
436 χ_{lf} and χ_{hf} values. Because Fe and Ti vary together ($r = 0.95$, $p < 0.001$), their ratio also permits to

437 evaluate the possible loss of Fe to Ti via dissolution. The decrease in Fe/Ti ratio in the upper layer (Fig.
438 9C), may result from the post-depositional dissolution of ferromagnetic oxides, mainly magnetite and
439 hematite. Overall, the reduction in the grain size and concentration of modes M1 and M2 (Fig. 5), is
440 linked to the exhaustion of carbonate debris supply and occurred during conditions favorable to
441 pedogenesis, as indicated by the upper layer increasing ($> 5\%$) χ_{fd} values that favored the dissolution of
442 Fe-Ti oxides and subsequent removal of Fe from PM loess. Similar patterns have been found in Holocene
443 loess-paleosol sequences along the NW Chinese loess plateau, where the contrasting behaviour between
444 χ_{lf} and χ_{fd} have are attributed to the formation of nonferromagnetic minerals associated with
445 waterlogging (Chen et al., 2012). If this has been the case for PM loess, it is argued that extended
446 snowcover and/or wet summer conditions during the deposition of PM loess upper layer, likely induced
447 waterlogged conditions which forced the observed pedogenic depletion of Fe (deferrification).

448

449 **Aeolian dust sources**

450 The local dust sources of Mount Olympus are the exposed glacial and periglacial landforms, the Pieria
451 mountaintops, which with an elevation of 2200m were likely affected by the late glacial erosion and the
452 alluvial plain of Katerini (Fig. 2A). Mount Olympus glacial and periglacial landforms are rich in
453 carbonate sand, whereas ultramafic sediments eroded from Pieria Mountains ophiolitic formations are
454 deposited in the alluvial plane of Katerini. On the contrary, Sahara dust mineral composition is rich in
455 quartz, phyllosilicates and clays (Brooks *et al.*, 2005), but also in crustal heavy minerals such as Ti and
456 Zr (Moreno *et al.*, 2006; Aarons *et al.*, 2013; Scheuven *et al.*, 2013). Zr and Ti usually vary together in
457 sediments because they are chemically immobile and in alpine soils have been used as proxies of detrital
458 heavy minerals (Muhs *et al.*, 1990). However, the low correlation between Ti and Zr ($r = 0.55, p < 0.03$),
459 implies that the origin of Zr and Ti in PM loess is likely different. Ti results from the mechanical
460 weathering of carbonate bedrock and is deposited in the PM loess sequence in the form of Fe-Ti oxides
461 occurring within mixed aggregates, while Zr is mainly transported in Mediterranean alpine systems
462 during episodes of Sahara dust transport (e.g. Jimenez-Espejo *et al.*, 2014). The nearly zero

463 concentrations of Zr in MK and TZ samples, rules out its detrital origin and supports the mechanism of
464 Zr aeolian enrichment in PM loess.

465

466 Within PM loess profile, the weight percent (wt%) concentration of mica and Zr XRF counts, display
467 high correlations ($r > 0.70$, $p < 0.003$) with M3 concentration. This relation argues that additionally to
468 mica (muscovite) presence in bedrock formations (TZ and MK samples) depicted from XRD spectra,
469 micaceous silt grains are also transported during Sahara dust episodes. The range of the M3 mean grain
470 size ranges between 14 and 28 μm and is similar to modern values Sahara dust modal and median grain
471 sizes from Crete (Fig. 1), which range between 8 and 30 μm (Goudie and Middleton, 2001) and 4 to 16
472 μm (Mattson and Niéhlen, 1996), respectively. Thus, it is reasonable to support that M3 can be
473 considered a representative grain size mode of Sahara dust contribution to PM loess.

474

475 However, rounded quartz grains occur in a variety of grain sizes from 2 to 15 μm (Fig. 7F), which is
476 also supported by the correlation between the sum of M1, M2 and M3 modal concentrations with quartz
477 wt% ($r = 0.74$, $p < 0.001$). This suggests that transport of Sahara dust to Mount Olympus includes finer
478 particles in clayey silt range, assuming that all aeolian quartz comes from Sahara region. Since quartz is
479 traced in minor quantities in MK and TZ samples (Fig. 5A), the conclusion that the finer modes M1 and
480 M2 contain aeolian components, either from Sahara, or from local sources (Pieria Mountains and
481 Katerini alluvial plane), is valid, but the exact origin of quartz cannot be defined from the existing
482 analyses. Therefore, synergistic to the weathering of Mount Olympus carbonates and deposition of
483 detrital components with subsequent post depositional production of fine particles and aggregates rich
484 in Fe-Ti oxides, is the deposition of fine dust incorporated into M1 and M2. Background dust with grain
485 size similar to M1 and M2 ($\sim 3\mu\text{m}$), is found in many European loess sequences and represents local,
486 regional, or supracontinental sources (Bosq et al., 2020). Deflation and deposition of fine particles from
487 Pieria Mountains and Katerini alluvial plane on Mount Olympus and their contribution to M1 and M2
488 composition, are supported by the strong ($r > 0.85$) correlations between M1 and M2 concentrations
489 with Cr and Ni, which are elements representative of ultramafic rocks. The source of Cr and Ni can be
490 deflated ophiolitic sediments either deposited by periglacial activity on the mountaintops of Pieria

491 Mountains or transported by fluvial action to the proximal Katerini alluvial plain. The respective
492 correlations between M1 and M2 concentrations with Cr and Ni, are high only along the lower (>16 cm)
493 layer of PM loess. The reduction of Ni and Cr along the upper layer is concomitant with the reduction
494 of M1 and M2 concentrations, but the partitioning of deflation sources between, background aeolian
495 dust, Mount Olympus glacial landforms, and the reworked sediments from Pieria Mountains and
496 Katerini alluvial plane, to the geochemical contribution M1 and M2 is difficult to discern.

497

498 **Post depositional alteration**

499 It widely accepted that in proglacial fields and alpine soils secondary clay mineral formation through
500 alteration of mica to smectite (Egli *et al.* 2003) and plagioclase to kaolinite (Anderson *et al.*, 2000)
501 occurs in rates similar to non-glacierized catchments (Anderson, 2005). Given the significant (>30%)
502 contributions of smectite and kaolinite in the clay (< 2 μ m) fraction of samples PM1 and PM15, the
503 possibility of post depositional alteration of mica to smectite and of plagioclase to the formation of
504 kaolinite, cannot be excluded for PM loess. Higher concentrations of chemical weathering products
505 (smectite and kaolinite) in the base of PM profile (sample PM15), compared to the surface sample
506 (PM1), enhance the view of enhanced mineral alteration towards the base of the profile. However,
507 significant concentrations (~12%) of kaolinite in the sediments of north Aegean Sea core SL148 and its
508 crystallinity characteristics, have characterized kaolinite as a proxy for Sahara dust input (Fig. 10B),
509 (Ehrmann *et al.*, 2007). So, the exact origin of kaolinite within PM loess clay fraction, either detrital,
510 aeolian, or formed through plagioclase weathering, cannot be accurately defined here.

511

512 In terms of both proglacial sediments and loess weathering dynamics, the early stage of Ca removal, is
513 followed by the intermediate stage of K removal and by the final stage of Si removal (e.g. Anderson *et*
514 *al.*, 2000; Hošek *et al.*, 2015). In Aquitaine, southwest France, decalcification of loess has been attributed
515 to syn or post-depositional Ca removal and is described by the Ca/Sr ratio (Bosq *et al.*, 2020). Ca/Sr
516 decreases with increasing intensity of Ca weathering (Bokhorst *et al.*, 2009) and within PM loess it
517 ranges from 8 to 2 from the base towards the surface of the PM profile. These values are considerably
518 lower compared to the respective values of the TZ (Ca/Sr = 89) and MK (Ca/Sr = 189) sediments. Ca/Sr

519 is significantly positively correlated with the finer modes M1 and M2 ($r > 0.6$), which could be explained
520 by the fact that decalcification of PM loess, largely occurs within the finer fractions, with subsequent
521 replacement of calcite to the formation of clay particles and mixed aggregates found in SEM images
522 (Fig. 7).

523 The observed Rb enrichment in PM, compared to MK and TZ samples (Fig. 6D), results from the
524 weathering of K-bearing minerals, such as mica (e.g. Anderson *et al.*, 2000; Hošek *et al.*, 2015; Zech *et*
525 *al.*, 2008). In the previous section, it is argued that in addition to its detrital origin, mica is an inherent
526 component of Sahara dust transport to Mount Olympus and is identified in small concentrations (~6%)
527 in PM loess. The loss of mica to smectite cannot be quantified, but it appears that after its initial
528 deposition, mica is subjected to post depositional weathering with removal of K (Buggle *et al.*, 2011,
529 Bosq *et al.*, 2020). This is supported by the low values of K/Rb elemental ratio (Fig. 9F), used on many
530 occasions to describe the weathering intensity and removal of K from loess deposits (Profe *et al.*, 2016).
531 Similar to Ca/Sr ratio, K/Rb values in PM loess are considerably lower (2.7) compared to TZ (45) and
532 MK (228) sediments and show high correlations ($r > 0.85$) with M1 and M2 concentrations, suggesting
533 that intense weathering and production of fine particles occurs within PM loess. This likely explains the
534 low concentration of fine modes within the MK and TZ clast free horizons since the chemical weathering
535 conditions under the cirque headwalls are low due to extensive snow cover, slope steepness, aspect, and
536 high production rates of coarse carbonate debris that enhances percolation of snowmelt. Translocation
537 of clay particles in the coarse matrix of glacial till and stratified scree deposits may also be responsible
538 for the minor contents of fine particles, but the assessment of these factors is beyond the scope of this
539 study.

540

541 **Relative chronology of PM loess**

542 The main step in establishing the relative chronology of PM loess deposition is to constrain the transition
543 period between the upper and lower layers from 14 to 16 cm of profile depth that partition several
544 sedimentological and geochemical changes. The 15% increase of M3 concentration along the transition
545 layer (Fig. 4C), suggests a growth in Sahara dust availability that can be associated with the
546 midHolocene termination of the African Humid Period (AHP; 10–6 ka BP) and the regional climatic

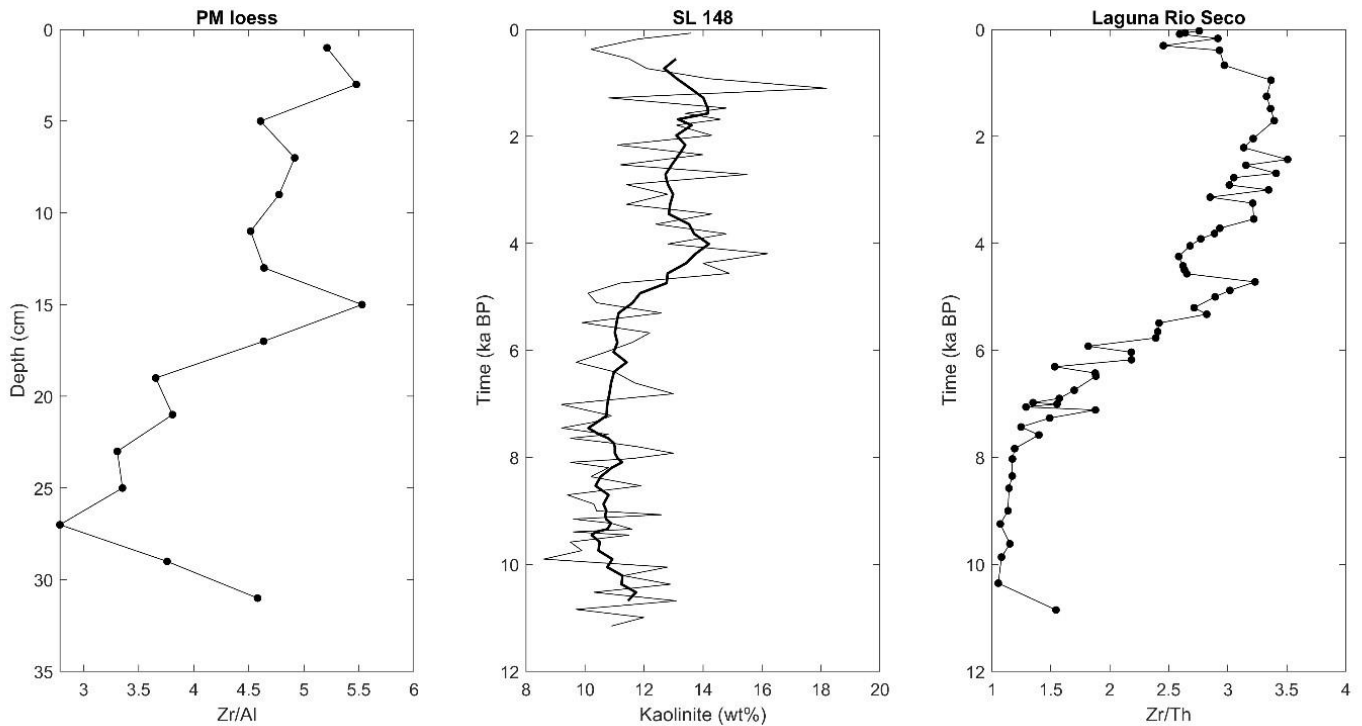
547 shift from Atlantic- to Mediterranean dominated with enhancement of southerly air flows. Several
548 studies
549 (e.g., Magny *et al.*, 2013, Perçoiu *et al.*, 2017, Zielhofer *et al.*, 2017b) have suggested a major
550 reorganization of winter atmospheric circulation patterns across Europe during the mid-Holocene
551 (coincident with the termination of the AHP), with the negative phase of the North Atlantic Oscillation
552 (NAO) dominating before ~6 to 5 ka BP and the positive phase, after ~6 to 5 ka BP. The NAO controls
553 the delivery of moisture from the Atlantic Ocean to mainland Europe (Hurrell *et al.*, 2013) in winter.
554 During NAO- periods, the low pressure difference between the Icelandic Low and the Azores High result
555 in southward deflected westerlies across Europe and associated high values of winter precipitation, while
556 during the NAO+ phase, the high difference between the two pressure systems deflects the westerlies
557 northwards, leaving southern Europe dry. The stronger than usual Azores High active during NAO+
558 periods further lead to increased advection of air and dust from Sahara towards southern Europe. D-
559 excess values in cave ice in East-Central Europe (ECE) indicate the intensification of Mediterranean
560 winds after ~6 ka BP, further supporting strengthened southerly advection of air over SE Europe. The
561 above, tentatively place the boundary between PM loess upper and lower layers at ~6 ka BP. Throughout
562 the literature, Zr concentrations expressed by the Zr/Th, or the equivalent Zr/Al ratios, have been used
563 as a proxy of Sahara dust input in the Mediterranean region (Jimenez-Espejo *et al.*, 2014 and references
564 therein). In Central Mediterranean marine core ODP 964 (Fig. 1), the Zr/Al ratio along with the wt%
565 concentrations of kaolinite and palygorskite, have been also used as a Sahara dust proxy, with generally
566 lower values during past AHP (e.g. Gallego-Torres *et al.*, 2007). Kaolinite content is also used as a proxy
567 of Sahara dust influx in the north Aegean Sea core SL-148 (Ehrmann *et al.*, 2007). Further chronological
568 constrains on the PM loess depositional history are achieved through the visual correlation of Zr/Al ratio
569 of PM loess with the respective Zr/Th ratio from Laguna De Rio Seco in the Spanish Pyrenees (Jimenez-
570 Espejo *et al.*, 2014) and with the kaolinite concentrations of north Aegean marine core SL 148 (Ehrmann
571 *et al.*, 2007).

572
573 The curve similarity of the three profiles shown in Fig. 10, tentatively confirms the previous
574 consideration that the transition period between the lower and upper layers of PM loess broadly coincides

575 with the termination of African Humid Period at ~6 ka BP. A subsequent peak in Sahara dust transport
576 around 4.5 ka BP marks the upper boundary of this transition layer. Of particular interest is the temporal
577 constrain of the profile base with the relative date of sample PM3 placed ~10 ka BP. This implies that
578 the calcite rich samples PM1 and PM2 were deposited during the initial stages of Mount Olympus
579 deglaciation phase, between 12 and 10 ka BP, in agreement with the stabilization of moraines in TZ
580 cirque (Fig. 2A). During this phase, influx of meltwater from the retreating cirque glaciers provided
581 aggressive solutions that were reacting with the carbonate bedrock dissolving it in high rate.

582 A further implication that can be derived from chronological constrain of PM loess, concerns the
583 pedogenetic conditions between 6 and 0 ka BP. During this period, lower concentrations of the finer
584 modes M1 and M2, lower magnetic signal and decreased values of Fe/Ti, suggest that pedogenesis was
585 induced by wetter than present summer conditions, which are confirmed from pollen based climatic
586 reconstructions validated by a regional climatic model (Peyron et al., 2017). Broadly speaking, an
587 additional feature that can be deduced from the relative chronology of PM loess profile concerns its
588 average sedimentation rate. Niéhlen *et al.* (1995), conclude an average Holocene deposition rate of
589 Sahara dust of ~2 cm/ka, which is close to ~2.5 cm/ka derived from for the upper layer of PM loess. The
590 remaining ~0.5 cm/ka should represent the contribution of local weathering, as supported by a recent
591 study, which is based on ³⁶Cl-inferred denudation rates from western Mediterranean mountains and
592 suggests average weathering rates of 0.3-0.6 cm/ka during the last glacial / interglacial cycle (5-40 ka
593 BP, Thomas et al., 2018).

594



596

597 **Fig. 10.** Establishment of relative chronology from comparison with respective proxies of Sahara dust aeolian
 598 transport from eastern and western Mediterranean, during the Holocene. Zr/Al ratio of PM loess, kaolinite wt%
 599 concentrations of marine core SL 148 (Ehrmann *et al.*, 2007), with thick black line representing the 7-point running
 600 mean and Zr/Th ratios from laguna Rio Seco in Spanish Pyrenees (Espejo *et al.*, 2014).

601

602 COCLUSIONS

603 The formation factors and aeolian dust sources of a Holocene loess sequence deposited in the highest
 604 plateau of Greece are assessed for the first time through a multi proxy analysis. The Plateau of Muses
 605 (PM) on Mount Olympus, located at 2600 m is formed by the combined action of karst dissolution and
 606 glacial erosion and constitutes a natural trap of locally eroded and aeolian sediments. Collectively, the
 607 sedimentological, chemical, mineralogical and magnetic data, along with a relative chronological
 608 framework, confirm the hypothesis that deposition of PM loess started ~10 ka BP, following the most
 609 recent deglaciation phase on Mount Olympus. Grain size analyses depicted 5 dominant grain size modes
 610 in all samples. Calcite rich sandy populations dominated by Ca and Sn make up to 95% of the clast free
 sediment horizons of glacial and periglacial sediments, collected from the adjacent glacial cirques,

611 contrasting the aeolian quartz rich (60%) silts and clays that dominate (95%) PM loess sedimentary and
612 mineralogical composition. Glacially derived coarse carbonate sand constitutes a minor proportion (5%)
613 of its grain size modal contribution, whereas similar Fe/Ti values among the contrasting depositional
614 environments considered, supplemented by SEM imagery, suggest the release of Fe-Ti oxides from
615 bedrock formations and their subsequent enrichment in PM loess in the form of mixed aggregates.

616

617 The most dominant dust source of PM loess is Sahara Desert, as the study area is located within the
618 boundaries of intense Sahara dust fallout zone. Local dust sources include the deglaciated slopes of
619 Mount Olympus and the adjacent Pieria Mountains and Katerini plain dominated by ophiolitic alluvial
620 sediments. The aeolian silt fraction of PM loess, expressed by grain size mode M3, shows small
621 variations in its grain size (15 μm) and high correlations with Zr/Al and mica wt%, properties
622 representative of modern Sahara dust grain size, mineralogy and geochemical composition. Reduced
623 concentrations of M3 between 10 and 6 ka BP are attributed to the reduced deflation of quartz rich silts
624 during the most recent AHP and by the dominance of an Atlantic type of climate with enhanced
625 westerlies. A mid-Holocene swift in the regional climate from Atlantic to Mediterranean type with drier
626 conditions and more frequent periods of Scirocco winds coincided with the termination of AHP and
627 increased deflation of Sahara dust grains from the desiccated areas. This regional climatic shift resulted
628 in prominent increases in the aeolian silt deposition (increase of M3 concentrations) and Zr/Al ratio
629 between 6 and 4.5 ka BP, with a concomitant decrease in the pedogenic modification of the deposited
630 dust and decreasing clay particle formation (decrease of M1 and M2 concentrations). Contrary to the
631 enhancement of Sahara dust transport on Mount Olympus since 6 ka BP, is the decrease of local dust
632 from the Pieria mountaintops and Katerini plain, as shown by the correlation of clay and fine silt with
633 Cr and Ni. The associated decrease of clay concentration with the heavy elements, can result either from
634 decreases either in summer convection and/or to northern winds outbreaks.

635

636 PM loess is decalcified and subjected to secondary syn or post depositional chemical weathering, which
637 include removal of Ca and K respectively. The upwards decreasing trends of Ca/Sr and K/Rb imply that
638 the elemental modification of PM loess has been gradual and independent of the aeolian deposition and

639 regional climatic dynamics. The secondary mineralogical modification may be responsible for the high
640 amounts of smectite and kaolinite observed in the clay fraction, through weathering of mica to smectite
641 and plagioclase to kaolinite, but further conclusions on these processes cannot be achieved through the
642 analyses presented here. In addition, during deposition of the upper PM loess layer (6 – 0 ka BP), wetter
643 than present summer conditions likely resulted in waterlogging and subsequent dissolution of Fe from
644 the Fe-Ti oxides (deferrification) and to pedogenic depletion of the magnetic signal.

645

646 Overall, the mechanisms responsible for the formation of PM loess are complex and involve several
647 convoluting processes, such as mechanical weathering of the glacial carbonate debris, chemical
648 dissolution of the weathered products, syn and post depositional alteration and formation of aggregates,
649 pedogenetic modification and aeolian dust deposition from local and regional sources. In the absence of
650 continuous reconstructions from Mediterranean alpine settings, future analyses of alpine loess deposits
651 in the sub cm scale can provide a powerful tool to study the local weathering dynamics and regional
652 atmospheric circulation patterns, focusing on periods of Sahara dust events and enhanced Sirocco winds
653 throughout the Holocene.

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