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Timing and mechanisms of sediment accumulation and pedogenesis: Insights from the Po Plain (northern Italy)

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Abstract

Paleosol stratigraphy in the Po Plain, reconstructed through sedimentological analysis and correlation of ca. 170 core data chronologically constrained by 376 radiocarbon dates, revealed strict relationships between pedogenetic and fluvial processes in alluvial and paralic environments. Vertically stacked, weakly developed paleosols within Upper Pleistocene and Holocene mud-prone strata testify to intermittent pedogenesis, periodically interrupted by overbank sedimentation. Individual paleosols are laterally traceable for tens of km and exhibit A-Bk-Bw, A-Bk or A-Bw profiles. Stratigraphically ordered ^{14}C calibrated ages from A horizons testify to slow aggradation during 4-6 thousand years-long exposure periods. Burial ages, with an error of few centuries, are provided by plant debris at the top of A horizons.

Millennial-scale climate oscillations and glacio-eustasy are the main drivers of the pedo-sedimentary evolution of the area during the last 50 kyr. Pleistocene (P) paleosols developed in well-drained floodplain environments, dissected by fluvial incision, during relatively warm periods. Overbank sedimentation and paleosol burial occurred during colder phases. High-sediment-supply during the Last Glacial Maximum hindered pedogenesis and led to the accumulation of 3-10 m-thick overbank strata. Widespread soil development (paleosol PH) occurred at the end of Last Glacial Maximum, following glaciers retreat and afforestation of drainage basins. At distal locations, paleosol PH was progressively buried under estuarine sediments during the Holocene phases of post-glacial sea-level rise. Beyond the area of marine influence, burial ages of paleosol PH change from a place to another without specific spatial trends and reflect upstream fluvial sedimentation dominated by avulsions and deposition of spatially restricted alluvial units. Holocene (H) paleosols show a poor correlation potential and laterally variable degree of maturity that reflect avulsive

49 sedimentation patterns and crevassing. Deforestation and land reclamation affected pedo-
50 sedimentary processes during the last 6 kyr.

51

52

53 **Keywords**

54 Paleosol stratigraphy, pedo-sedimentary evolution, radiocarbon dates, climate change, eustasy,
55 Late Quaternary

56

57

58 **1. INTRO**

59

60 In alluvial settings, the development and burial of soil horizons are intimately related to fluvial
61 activity (Kraus and Bown, 1993) and governed by a variety of factors. High sediment supply and
62 fluvial aggradation inhibit pedogenesis (Demko et al., 2004), whereas low sediment supply and
63 fluvial incision promote soil development in interfluvial areas (Blum and Törnqvist, 2000). Climate
64 influences the amount of sediment delivered to the basins, modulating the magnitude and type of
65 precipitations, the vegetation cover of drainage areas and the river's transport capacity
66 (Vandenberghe, 2003). Alluvial sedimentation can be locally hindered or enhanced by tectonic
67 movements (Holbrook and Schumm, 1999; Viseras et al., 2003; Benvenuti et al., 2008; Pati et al.,
68 2019), differential subsidence (Cohen et al., 2003) or by avulsions and other processes related to
69 the intrinsic dynamics of the fluvial apparatus (Kraus, 1999; Cleveland et al., 2007; Hajek and
70 Wolinsky, 2012). In coastal settings, fluvial dynamics are strongly influenced by sea-level oscillations
71 (Atchley et al., 2004; Blum et al., 2013). Incised valleys (and associated interfluvial paleosols) related

72 to glacio-eustasy are a widely recognized trait in the sedimentary record (Shanley and McCabe,
73 1994; Aitken and Flint, 1996; McCarthy and Plint, 1998; Plint et al., 2001; Gibling et al., 2011).

74 The causal relationships between paleosol formation and alluvial sedimentation have been
75 reconstructed in ancient alluvial successions, where the geometric relationships between paleosols
76 and associated fluvial sediment bodies are superbly exposed (Gibling and Bird, 1994; McCarthy and
77 Plint, 2003; Dubiel and Hasiotis, 2011; Soares et al., 2020). However, the role of distinct controlling
78 factors on paleosol development and burial can hardly be assessed (Wright and Marriott, 1993;
79 Atchley et al., 2004), because of the temporal resolution of pre-Quaternary dating methods,
80 insufficient to describe pedo-sedimentary processes which typically act at millennial time scales
81 (Rasbury et al., 1997; Hofmann et al., 2017). Furthermore, diagenesis may have substantially
82 modified soil properties after burial, hindering comparison with modern soils and the reconstruction
83 of the soil-forming environment (Trendell et al., 2012).

84 Improved techniques of subsurface investigation have recently prompted the study of buried
85 upper Quaternary alluvial successions (Choi, 2005; Tsatskin et al., 2015; Amorosi et al., 2017a,b). In
86 this relatively narrow time window, drainage areas, ecosystems and soil-forming environment did
87 not change significantly. Moreover, the high temporal resolution of Quaternary dating methods
88 permits to link unequivocally pedogenetic processes to a well-known climate and eustatic history.

89 Radiocarbon is probably the most reliable dating method for the last 50 kyr (Reimer et al., 2020).
90 However, numerous uncertainties remain in the interpretation of ^{14}C dates from paleosols. Organic
91 carbon is continuously incorporated in soils, and subjected to complex, and not totally understood,
92 biological and physical transformations (Larionova et al., 2017; Meingast et al., 2020). These
93 processes can act at different velocity and magnitude, depending on climate, morphology of the
94 exposed surface, parent material, and vegetation (Alexandrovskiy and Chichagova, 1998; Kleber,
95 2010).

This study is based on refined pedostratigraphic reconstructions and on a vast ^{14}C dataset which permitted to chronologically constrain the major architectural and facies changes. The primary aim of this research is to understand the causes and mechanisms of soil development and burial in an alluvial-deltaic setting and its relation to fluvial and coastal dynamics. To this purpose, we focused on Upper Pleistocene-Holocene paleosols and on their stratigraphic relationship with older, coeval and younger pedo-sedimentary units. Secondary objectives are to discuss: (i) the relative role of global and local controlling factors on soil development and burial; (ii) the potential use (and major limitations) of radiocarbon to assess the exposure age of paleosols.

2. GEOLOGICAL SETTING

2.1. Structural setting and basin-scale stratigraphy

The Po sedimentary basin is enclosed in a natural amphitheatre bordered by the Alps, to west and to the north, and by the Apennines, to the south. The eastward prolongation of the Po Plain is the Adriatic epicontinental sea (Fig. 1). The most external thrusts of the Southern Alps and of Apennines, showing opposite vergence, locally face each other beneath the Po Plain (Fantoni et al., 2004). South-Alpine buried thrusts are arranged in a single wide arc running from Milan to the Garda Lake (Vannoli et al., 2015). The Apennine front is composed of three tectonically active arcuate thrust systems: from west to east, the Monferrato, Emilia and Ferrara-Romagna Arcs (Ghielmi et al., 2013). Thrusts of the Emilia and Ferrara Arcs are sealed beneath a pile of sediments with extremely variable thicknesses (from a few hundred metres to ca. 8 km; Amadori et al., 2019). These lateral variations in thickness are controlled by lateral changes in subsidence rates (Campo et al., 2020),

120 ranging between zero and 2 mm/y (Bruno et al., 2020a). The northern margin of the Apennines,
121 which corresponds to an out-of-sequence system of N-NE verging thrusts (Maestrelli et al., 2018),
122 has been involved in a generalized uplift since the Early Pleistocene (Argnani et al., 2003).

123 The Po Basin fill shows an overall regressive trend from deep-marine to continental and coastal
124 units (Ghielmi et al., 2013). Widespread alluvial sedimentation started in the Po Plain around 870
125 kyr B.P. (Muttoni et al., 2003, 2011; Gunderson et al., 2014). The Po River, flowing in W-E direction,
126 divides the alluvial plain in two sectors (Fig. 1A). In the northern sector, Alpine tributaries flow
127 entrenched in Pleistocene sandy units (Ravazzi et al., 2012; Fontana et al., 2014; Bruno et al., 2018).
128 To the south, paleosol-bearing Pleistocene muds are sealed by a thick (up to 30 m) Holocene alluvial
129 and paralic succession (Amorosi et al., 2017a, b). Early Holocene estuarine deposits typically onlap
130 onto Upper Pleistocene alluvial strata, testifying to the progressive drowning of the alluvial plain
131 after 11.5 cal kyr B.P. (Bruno et al., 2017). Following stabilization of sea-level, the Middle-Late
132 Holocene records the progressive filling of the Po estuary and the successive delta progradation
133 (Amorosi et al., 2017b, 2019).

134

135 **2.2. Paleosols of the Po Basin**

136

137 Highly weathered rubified soils have been reported from the Po Basin margin (Cremaschi, 1987;
138 Garzanti et al., 2011) and from isolated hills in the alluvial plain, generated by South-Alpine (Zerboni
139 et al., 2015) and Apennine blind thrusts (Zuffetti et al., 2018). In depocentral areas, mature paleosols
140 are replaced by sequences of aggradationally stacked, weakly developed paleosols laterally
141 traceable for tens of km (Amorosi et al., 2017a). A series of closely spaced paleosols, marking the
142 MIS 3/MIS 2 transition, has been described as the equivalent of the Interfluve Sequence Boundary
143 (McCarty and Plint, 2003) in a high accommodation setting, due to its peculiar geometric

relationships with genetically related fluvial-channel bodies (Amorosi et al., 2017a). The most widespread paleosol reported in the Po Plain marks the Pleistocene-Holocene boundary and responds to several local names: 'Caranto' in the Venetian Plain (Mozzi et al., 2003), 'Ringhiera paleosol' in the Romagna area (Ravazzi et al., 2006), 'YD paleosol' in the Po Delta Plain (Amorosi et al., 2017b; Morelli et al., 2017), 'PH paleosol' in the Bologna urban area (Amorosi et al., 2014; Bruno et al., 2020b; this name will be used in this paper). Although a wide literature reported the existence of this pedogenized horizon, the mechanisms and causes of its formation and burial are not yet fully understood. More discontinuous paleosols have been described from the Holocene succession (Bruno et al., 2015, 2013).

153

154

155 **3. METHODS**

156

This work is based on the sedimentological and stratigraphic analysis of core material. Sixty-nine cores, 30-50 m deep (red circles in Fig. 2), were analysed and sampled for radiocarbon dating. For each core lithology, grainsize, colour and accessory materials (vegetal and shell macrofossils, carbonate concretions, redoximorphic features) were reported. The reaction to HCl was tested as a qualitative indicator of the presence of calcium carbonate. Resistance was measured on fine-grained sediments with a Pocket Penetrometer (PP). One hundred and seventy-two wood, peat, vegetal-remains and bulk-soil samples were dated at the KIGAM AMS laboratory (Daejeon, Republic of Korea) and at the Ion Beam Physics laboratory of the ETH (Zurich, Switzerland, Synal et al., 2007). Samples were collected fresh from the innermost part of the core and desiccated in a 40° oven. The humin fraction was separated through acid-alkali-acid (AAA) pre-treatment and AMS dated. Humic

166

167 acids, extracted from 9 bulk soil samples were also dated. Conventional ^{14}C ages were calibrated
168 using OxCal 4.4, with the IntCal 20 curve (Reimer et al., 2020).

169 More than 100 core descriptions from previous works and from the database of the Geological,
170 Seismic and Soil survey of Regione Emilia-Romagna (yellow circles in Fig. 2) were reinterpreted
171 following calibration with new cores. These descriptions provide information on lithology, colour,
172 consistency, reaction to 10% HCl, pocket penetrometer measurement and radiocarbon dates.
173 Additional 204 radiocarbon dates derive from previously published works (Geological Map of Italy
174 to scale 1:50.000; Ravazzi et. al., 2006; Cacciari et al., 2017; Marabini and Vai, 2020).

175

176

177 **4. RESULTS**

178

179 **4.1. Sedimentary facies**

180

181 The sedimentological analysis of the studied cores led to the identification of several facies
182 associations which belong to alluvial, deltaic, estuarine and shallow-marine depositional
183 environments. To the purposes of this paper, we will describe in detail four main facies associations,
184 which are part of the alluvial, upper delta-plain and inner estuary depositional systems. For other
185 facies associations, the reader is referred to Amorosi et al. (2017b, 2019) and Bruno et al. (2017).

186

187 **4.1.1. Fluvial channel-related facies association (FCR)**

188

189 This facies association is composed of cross-stratified or laminated coarse-to-fine sand with local
190 mud intercalations (Fig. 3). The occasional presence of floated woods or plant debris is recorded.

191 Marine or lagoonal macrofossils are absent, whereas shell fragments of freshwater gastropods
192 are seldom encountered.

193 Based on lithology, sedimentary structures and on the lack of marine and lagoonal macrofossil,
194 these coarse-grained deposits are interpreted as related to fluvial activity (in-channel deposition,
195 bar migration, crevassing, proximal overbank; Miall, 1985; Toonen et al., 2012; Aslan, 2013).

196

197 4.1.2. Freshwater swamp and marsh (SM)

198

199 This facies association is dominated by fine-grained material, with abundant wood, vegetal
200 remains and peat (Fig. 3). Peat horizons, 5-30 cm thick, are composed of poorly decomposed woody
201 material and are separated by grey (2.5 YR 7.1, 7.5 YR 8/1), peat-free clays. Freshwater fossils were
202 locally encountered. Consistency is very weak and PP values are $< 1 \text{ kg/cm}^2$. Carbonate concretions
203 and redoximorphic features are absent. This facies association shows poor lateral extent in the
204 Upper Pleistocene succession. On the contrary, in Holocene distal deposits it is volumetrically
205 dominant and only locally interrupted by channel-related sands.

206 The grain size of this facies group denotes a low-energy depositional environment. The
207 abundance of poorly decomposed organic material and the lack of oxidation suggest the setting of
208 reducing conditions in submerged environments. The latter were likely isolated ponds during the
209 Late Pleistocene, and swamp-marshes part of wider delta-plain and bayhead delta systems during
210 the Holocene (Amorosi et al., 2017b; Bruno et al., 2017; Cacciari et al., 2020). Peat horizons
211 separated by peat-free clays testify to intermittent sedimentation, with peat accumulation during
212 sedimentation breaks (Hijma and Cohen, 2011; Ishii et al., 2016; Bruno et al., 2019).

213

214 4.1.3. Poorly drained floodplain (PDFP)

215

216 This facies association is composed of grey (7.5 YR 6/1) silty clays with faint lamination given by
217 millimetre-scale silt intercalations. Soft and isolated carbonate concretions (CC in Fig. 4) and sparse
218 plant debris (UOM in Fig. 4) were seldom encountered. Redoximorphic features are lacking.
219 Freshwater gasteropods were seldom encountered. Consistency is weak to firm. Pocket
220 penetrometer values are in the range of 1-2 kg/cm².

221 Based on grainsize and fossil content, this facies association is interpreted as deposited in a distal
222 fluvial environment, occasionally subjected to river flooding. Lack of redoximorphic features
223 suggests a groundwater table fluctuating close to the topographic surface (poorly drained
224 floodplain). The amount, distribution and type of secondary calcite are consistent with this
225 interpretation.

226

227 4.1.4. Well-drained floodplain (WDFP)

228

229 This facies association is composed of firm-to-hard clayey silts and silty clays. Sedimentary
230 structures are absent. By contrast, polygonal structures, desiccation cracks and rootlets are
231 commonly observed. A well-expressed horizonation is given by colour and CaCO₃ distribution (Fig.
232 3). Dark horizons, showing weak or no reaction to HCl alternate with lighter horizons showing strong
233 reaction to HCl. Carbonate concretions, in the form of coalescent nodules, coatings, filaments and
234 sub-horizontal bands are observed in lighter horizons. Redoximorphic features, in the form of
235 orange mottles or bands (RF in Fig. 6), are commonly observed at the mesoscale. Plant debris and
236 freshwater gasteropods are seldom encountered. PP values are > 2 kg/cm².

237 As for facies PDFP, grainsize and fossil content point to a distal fluvial environment, occasionally
238 inundated by river waters (Collinson, 1996). The alternation of dark and light horizons is interpreted

239 as the result of multiple phases of soil development interrupted by overbank deposition and
240 paleosol burial. Dark colour reflects the accumulation and decomposition of organic matter in the
241 topsoil (Bruno et al., 2020b). Carbonate distribution, with carbonate-free horizons overlying CaCO_3
242 enriched horizons, is interpreted as the result of carbonate leaching from the topsoil (A horizon)
243 and illuviation to the underlying Bk horizon (Boul et al., 2011). Carbonate mobilization requires the
244 percolation of meteoric waters in a vadose zone. We, thus, argue that the groundwater was located
245 a few metres below the topographic surface during subaerial exposure (well-drained floodplain).

246

247

248 **4.2. Paleosol stratigraphy**

249

250 Paleosols were identified at three stratigraphic intervals in the Upper Pleistocene-Holocene
251 succession of the Po Plain.

252 The lower interval is composed of a series of closely-spaced paleosols (paleosols P in Figs. 4, 5
253 and 6), whose cumulative thickness is in general < 5 m. Calibrated ages span from the ^{14}C detection
254 limit (~ 50 kyr B.P.) to ~ 23 kyr B.P. (Figs. 5 and 6). These paleosols show A-Bk-Bw or A-Bk profiles. A
255 horizons, 30-50 cm thick, are decalcified (Bruno et al., 2020b; Amorosi et al., this volume) and
256 present diffuse organic matter (DOM in Fig. 4) and slight bands of Fe oxides and/or hydroxides (Fig.
257 3). Undecomposed organic matter was not observed and carbonate concretions have locally been
258 reported from the base and the top of these horizons. Bk horizons are typified by high CaCO_3 content
259 (30-40% of the total volume, Bruno et al., 2020b). Carbonate concretions are abundant as
260 coalescent nodules or whitish sub-horizontal bands (evolutionary stage II of Gile et al., 1981;
261 Machette, 1985). Transition to overlying and underlying horizons is in general gradational. Bw
262 horizons show moderate CaCO_3 content (Bruno et al., 2020b; Amorosi et al., this volume) and faint

263 mottles of Fe oxides and-or hydroxides. Evidences of clays mobilization (Bt horizons) were not
264 observed. At distal locations, these paleosols may show less clear horizonation (A-Bw profile) or may
265 be replaced by coeval peat-bearing swamp deposits (Fig. 5).

266 In the second interval, a single paleosol (paleosol PH, Figs. 4, 5 and 6) is encountered at the top
267 of a 3-10 m thick sedimentary unit composed of overbank muds and subordinated channel-related
268 sands (Last Glacial Maximum Sedimentary Unit, LGM SU in Figs. 4, 5 and 6). Textural characters of
269 paleosol PH vary from proximal to distal locations. At inland locations (core EM15, Fig. 3), paleosol
270 PH shares many characteristics with older P paleosols: (i) a carbonate-free, brown (10YR 4/4, 3/3) A
271 horizon, up to 100 cm thick, with DOM and RF; (ii) a lighter Bk horizon with abundant nodules or
272 bands of secondary calcite (evolutionary stage II); (iii) a Bw horizon showing evidence of incipient
273 alteration. At more distal locations (cores EM1 and EM3, Fig. 3), paleosol PH exhibit a grey (5Y 4/1,
274 2.5Y 6/1) decalcified A horizon, 30-50 cm thick, with DOM. Plant debris, absent in the lower part,
275 become increasingly abundant upsection and, at places, grade into the overlying peats (see core B3,
276 Fig. 4). The underlying Bk horizon, when present, is relatively thin (a few dm), with secondary calcite
277 in the form of nodules, coatings or filaments (evolutionary stages I and II). Redoximorphic features
278 were not observed at the mesoscale along the whole soil profile.

279 In the third interval, dated to the Holocene, paleosols (H1 and H2 in Figs. 4, 5 and 6) have been
280 observed only at inland locations. At distal locations, they are replaced by coeval estuarine and
281 deltaic deposits. H paleosols have poor correlation potential and their textural features vary
282 dramatically from a place to another, without specific spatial trends. In few localities, they show
283 characteristics similar to those of their Pleistocene counterparts. At places, they are barely
284 identifiable due to a poorly developed horizonation. Archeological artifacts are increasingly
285 abundant in younger paleosols.

286 Paleosols P and PH bound nearly tabular sediment packages and are locally replaced by coeval
287 incisions filled with FCR or SM deposits (see cores CNT, RN3 and EM2, Fig. 5). On the contrary,
288 Holocene paleosols overlie or are overlain by CR facies (see cores MW and 15, Figs. 5 and 6). Alluvial
289 units delimited by these soil horizons are poorly extended and typically wedge out laterally or
290 downdip (Figs. 5 and 6).

291

292

293 **4.3. Radiocarbon dating of paleosols**

294

295 Upper Quaternary successions provide the great opportunity to link pedo-sedimentary events to
296 the detailed climatic and eustatic history described in a vast literature (eg. Clark and Huybers, 2009;
297 Lambeck et al., 2011; Rasmussen et al., 2014). Assessing the timing of soil development and burial
298 is crucial to disentangle the relative influence of global and local factors. This chapter summarizes
299 observations deriving from the analysis of a chronological database composed of 376 radiocarbon
300 dates (160 from paleosols).

301 Along single cores, multiple dating of A horizons yielded stratigraphically ordered ages, spanning
302 3-5 kyr (see cores B3, RW28 and 13, Figs. 4, 5C and 6). Only in core SPR (Fig. 5C) two samples from
303 the base and the top of the same A horizon provided similar ages. In distal cores, plant debris from
304 topmost A horizons yielded ages that are considerably younger (up to 5 kyr) than those from bulk-
305 soil samples and just a few centuries older than those from overlying SM peats (see cores EM1, EM3
306 and B3, Figs. 3, 4 and 5). Dates from A horizons indicate aggradation at extremely low rates (0,05-
307 0,23 mm yr⁻¹) during soil development. Accumulation rates above or between paleosols are at least
308 one order of magnitude greater (> 1 mm/yr). Distinct ages were also obtained from different

309 fractions extracted from the same sample. Particularly, humic acids yielded, in 9 cases out of 10,
310 ages older than those obtained from the humin fraction (Table 1).

311 Dates from bulk-soil samples change also laterally, along the same paleosol, spanning in a range
312 of 4-6 kyr (Fig. 3). No specific trend was observed from proximal to distal locations. Ages also change
313 over short distances, as observed in the Bologna area. In the Two Tower Medieval district, where
314 distance between cores is just 10-20 m, six samples from paleosol P3 yielded ages in the range of 3-
315 4 kyr (Fig. 6).

316 The frequency distribution of radiocarbon dates from paleosols (grey rectangles in Fig. 7) shows
317 a major peak between 14 and 10 kyr B.P., related to the PH paleosol. Samples from the P paleosols
318 yielded ages clustered around three minor peaks (P1, P2 and P3, Fig. 7), at 46-41, 38-32 and 29-25
319 cal kyr B.P., respectively. The overall decay with time in the number of ^{14}C dates across the
320 Pleistocene simply reflects the fact that older sediments were penetrated by a progressively lower
321 number of cores. A decline in the number of ^{14}C dates from soil samples is also recorded towards
322 younger ages, together with a sharp increase in dates from SM and PDFP deposits. Ages from
323 Holocene soil samples are clustered around two peaks. The older peak reflects, in part, the youngest
324 ages from paleosol PH and in part dates from a distinct paleosol (H1 in Figs 4, 5 and 6) observed at
325 several inland locations. In the Bubano Quarry (BQ in Fig. 5C), 5 m-thick overbank strata separate
326 H1 from PH. In the Bologna area (core 16, Figs. 4 and 6), paleosol H1 bears Neolithic and Eneolithic
327 artefacts (Bruno et al., 2013). The second peak (H2, Fig. 7) corresponds to another archeological
328 level, rich in Late Bronze Age, Iron-Age and Roman rests (IR paleosol of Bruno et al., 2013), which
329 was buried in large part of the Po Plain during the Middle Ages (see core CRV, Fig. 5A). Locally, this
330 paleosol is still exposed (Fig. 5). Interestingly, the maximum number of ^{14}C dates from facies FCR is
331 recorded between H2 and H1.

332

5. DISCUSSION

5.1 Use and limitations of radiocarbon for the estimation of the exposure age of paleosols

The use and reliability of radiocarbon dates from paleosols have been widely debated because, unlike plant macrofossils, which preserve the particular ^{14}C content of the moment of grow, soils are open systems where organic carbon is continuously incorporated, humified and-or oxidized to CO_2 before burial (Orlova and Panychev, 1993; Martin and Johnson, 1995; Gaudinski et al., 2000). Therefore, ^{14}C ages from paleosols reflect the mean residence time of the organic carbon plus the time of burial (Matthews, 1980; Geyh et al., 1983). The amount of organic carbon added to a soil and the velocity of its transformation processes change from place to place as a function of climate, vegetation and parent material (Reimer et al., 2020). Moreover, turnover and breakdown of organic matter may lead to the redistribution of ^{14}C ratios in different soil fractions. As a result, apparently conflicting ages can be obtained from the same paleosol (Figs. 5 and 6) and from different soil fractions (Table 1). Because of this inherent variability, many authors concluded that radiocarbon dates from soils are unreliable and the concept of “age” cannot be strictly applied to paleosols (Gilet-Blein et al., 1980; Becker-Heidmann et al., 2002). However, if not subjected to post-burial contamination (e.g. rootlets), paleosols should yield ages scattered within its formation period. The estimation of the length of this period, which can attain several millennia, is an intriguing goal for Quaternary paleosol stratigraphers.

The lack of evidence of clay mobilization, the evolutionary stage of secondary calcite and the limited rubefaction suggest that Po Plain paleosols evolved over relatively short periods. Radiocarbon ages from deposits below and above paleosols constrain their exposure period within

10-12 thousand years (Figs. 5 and 6). Dates from paleosols are within the narrower range of 4-6 millennia and provide a robust constrain for stratigraphic correlations over long distances (Amorosi et al., 2017a). More complex is the precise assessment of the ages that mark the onset and end of pedogenesis for each paleosol, even if radiocarbon dates are available from the base and top of A horizons. Previous work has demonstrated that, although some organic carbon can remain in soils for millennia (Feng, 2009), the oldest organic fraction accumulated at the onset of pedogenesis may have been completely replaced by younger organic carbon (Scharpenseel, 1971; Alexandrovskiy and Chichagova, 1998) introduced by roots in the deepest soil horizons. Thereby, radiocarbon dates from organic-sediment samples collected from the base of the A horizon may not be representative of the onset of soil formation (Gilet-Blein et al., 1980; Alexandrovskiy and Chichagova, 1998). Several factors influence carbon turnover times in soils (Kleber, 2010; Chen et al., 2013; Pandey et al., 2014). Particularly, carbon renewal is slower in colder climates and in deep soil horizons (Alexandrovskiy and Chichagova, 1998; Gaudinski et al., 2000; Fontaine et al., 2007). Therefore, for soils that developed in cold climatic conditions, such as paleosols P3, which formed at the onset of Last Glacial Maximum, dates from the base of the A horizon should better approximate the onset of pedogenesis. This assumption is corroborated by dates from core B3 (Figs. 3, 4 and 5), where the oldest age from paleosols P3 partially overlaps the age from underlying paleosol P2. For paleosols developed in warmer periods (PH and H paleosols), ages from the base of A horizons may post-date the onset of pedogenesis.

Sediment samples collected from the top of A horizons may be older than burial time, because a certain amount of time is required for litter humification and mineralization (Chichagova and Cherkinsky, 1993). At inland locations of our study area, where O horizons are generally not preserved, burial ages cannot be assessed precisely. Available dates permit to assert, for example, that paleosol PH was buried after 10.3 kyr B.P. in core 13 (Figs. 4 and 6) and between 14.3 and 8.6

381 kyr B.P. in core 201S4 (Fig. 5). Conversely, at distal locations, plant macrofossils preserved in the
382 topsoil provides ages that approximate the paleosol burial age with an error of a few centuries. For
383 example, in EM1, where estuarine sedimentation was already established around 8.1 kyr B.P., burial
384 of paleosol PH occurred around 8.5 kyr B.P. (Figs. 3, 4 and 5).

385 Age/depth gradients, like those observed in the Po Plain paleosols, have been reported from
386 several soil profiles (Matthews, 1981; Matthews and Dresser, 1983; Alexandrovskiy and Chichagova,
387 1998; Becker-Heidmann et al., 2002) and can possibly be explained by the fact that the amount of
388 carbon added to the topsoil through litter accumulation is in general order of magnitudes greater
389 than the one introduced in deeper soil horizons via roots exudation (Fontaine et al., 2007). The
390 measured accumulation rates, incompatible with alluvial sedimentation, suggest that litter
391 accumulation accounted for soil aggradation during exposure, with a possible contribution of wind-
392 transported dust. Therefore, paleosols from the Po Plain represent condensation horizons, rather
393 than depositional hiatuses. On the contrary, sedimentation between pedogenetic phases seems a
394 quasi-instantaneous process (see ages from P2 and P3 in core B3, Figs 4 and 5). Paleosols
395 aggradation is also testified by the presence of carbonate concretions in lower A horizons, likely
396 related to the upward migration of the eluvial-illuvial (A-B) boundary, and by vertically stacked sub-
397 horizontal bands of secondary calcite in Bk horizons (Fig. 3). Carbonate concretions in upper A
398 horizons, on the other hand, reflect post-burial illuviation from overlying paleosols. For this reason,
399 we avoided radiocarbon dating of carbonates and removed secondary calcite in acid steps of the
400 AAA sample preparation, before AMS analysis. Humic acids are also considered as a mobile
401 compound, which may yield anomalously young ages due to contamination from overlying strata
402 (Matthews, 1980). In our case, the oldest ages are provided by humic acids (Table 1). We thus
403 excluded contamination from overlying strata during each phase of pedogenesis.

404

5.2. Pedo-sedimentary evolution of the Po Plain and controlling factors

With the chronological constraint of 376 radiocarbon dates, the alluvial succession of the southern Po Plain provides an unprecedented record of pedo-sedimentary events across the last 50 kyr.

Aggradationally stacked paleosols 'P' developed during the Late Pleistocene, when the Adriatic coastline migrated in response to the stepwise sea-level fall toward its lowstand position, ~300 km away from the modern one (Pellegrini et al., 2018). Among P paleosols, only P3 developed during a phase of sea-level drop (~30–50m between 30 and 24 cal ka BP, Peltier and Fairbanks, 2006; Fig. 7B). Conversely, P1 and P2 developed in a period characterized by low-amplitude sea-level oscillations (Chappell, 2002; Fig. 7B). Far from the influence of sea-level change, pedo-alluvial processes are controlled by the balance between sediment supply and water discharge (Blum and Törnqvist, 2000). Low sediment supply-discharge ratios result in channel incision and soil development in the adjacent interfluvies, whereas high sediment supply-discharge ratios promote fluvial aggradation and paleosol burial (Demko et al., 2004). Bruno et al. (2020b) documented that soil development and burial during the Late Pleistocene was mainly controlled by changes in vegetation driven by climatic oscillations at the scale of Bond cycles (Bond et al., 1993). Soils started to form at the onset of Bond cycles, when warm climate conditions favoured the spread of dense forests (Magri, 1999; Fletcher et al., 2010), which protected the Apennine drainage basins from erosion (low sediment supply). Soil development was systematically interrupted during the subsequent cold phases, at the culmination of Bond cycles, corresponding to North Atlantic Heinrich events (light blue vertical lines in Fig. 7B). These periods record generalized forest retreats (Fletcher et al., 2010; Helmens, 2014), which enhanced erosion in the drainage basins and sediment transfer to the alluvial plains. Increased sediment supply promoted the filling of fluvial incisions and the

430 burial of formerly exposed interfluvies by overbank sedimentation. Thin sediment packages were
431 deposited between two successive phases of subaerial exposure (Figs. 3 to 6). These deposits (and,
432 partly, the underlying paleosols) were modified during the successive pedogenetic phase. A similar
433 response to millennial-scale climate oscillations has been observed in loess paleosol-bearing
434 successions (Rousseau et al., 2011, 2007; Moine et al., 2017; Zeeden et al., 2018).

435 The Last Glacial Maximum was characterized by deposition of thick and widespread fluvial-
436 channel gravel-sand bodies in the Po Plain (Ravazzi et al., 2012; Campo et al., 2016) and in other
437 alluvial systems worldwide (Lewis et al., 2001; Briant et al., 2005). Alpine glaciers, which extended
438 down to the Po Plain margin, supplied huge amounts of sediments to the related fluvio-glacial
439 systems (Fontana et al. 2014). In the Apennine domain, high sediment supply, favoured by a
440 predominantly herbaceous vegetation cover of drainage basins (Vescovi et al., 2010), resulted in the
441 deposition of the LGM SU, which bears poor evidence of pedogenesis (Figs. 4, 5 and 6).

442 Above the LGM SU, PH is the most widespread paleosol in the Po Plain. This surface yielded
443 radiocarbon dates spanning between ~17 and 6 kyr B.P. (Fig. 8), but mostly clustered around 14 and
444 10 kyr B.P. (Fig.7). The precise assessment of the age of PH formation is hampered by paucity of
445 dateable material from the underlying LGM SU. However, available dates indicate that the onset of
446 post-LGM pedogenesis was diachronous across the Po Plain, without a specific spatial trend (Fig. 8).
447 Whereas in the Bologna area PH started to develop at least 16.8 kyr B.P. (see core RW28, Figs 6 and
448 8), in the Romagna plain (Bubano Quarry - BQ, in Figs 5C and 8) pedogenesis started after 14.4 kyr
449 B.P. (Ravazzi et al., 2006). We argue that post-LGM pedogenesis was favoured by decreased
450 sediment supply related to the progressive afforestation of the area (Pini et al., 2010; Vescovi et al.,
451 2010; Campo et al., 2020), with diachroneity possibly related to particular local conditions (e.g.
452 distance from active channels). North of the Po River, in the Alpine tributary system, fluvial incision

453 occurred soon after glacial retreat (~17 kyr B.P., Monegato et al., 2017), due to sediment trapping
454 in lakes which formed in place of abandoned glacial amphitheatres (Fontana et al., 2014).

455 Interestingly most of the ^{14}C dates from paleosol PH post-date the Oldest Dryas (OD in Fig. 7).
456 The clustering of dates around the Younger Dryas in the Po delta plain (Figs. 7A and 8), suggested a
457 possible control of this cold event on paleosol formation (Amorosi et al., 2017b, Morelli et al., 2017).
458 The wider dataset shown in this work highlights that pedogenesis started in many sites of the Po
459 Plain at least during the Bølling and Allerød warm phases (BA in Fig. 7A). Unlike cold phases at the
460 culmination of pre-LGM Bond cycles, the Younger Dryas cold spell did not lead to paleosol burial.
461 This event was probably too short-lived to promote the filling of fluvial incisions and overbank
462 sedimentation. Paleosol development recorded after LGM in other alluvial (Mol, 1997; Choi, 2005;
463 van Balen et al., 2010; Janssens et al., 2012) and eolian (Kaiser et al., 2009; Hošek et al., 2017)
464 successions, indicates a likely climate control at a supra-regional spatial scale. Progressively warmer
465 conditions lead to widespread afforestation and reduced sediment supply (Fletcher et al., 2010;
466 Helmens, 2014).

467 Whereas Alpine rivers still flow entrenched in Pleistocene deposits, Apennine incisions were filled
468 during the Early Holocene (see cores RN3 and EM2, Fig. 3C) and paleosol PH was buried soon after.
469 In the coastal plain, where paleosol PH is encountered at greater depths beneath estuarine deposits,
470 burial ages show an overall inverse power law with elevation (Fig. 9) that reflects the progressive
471 drowning of the area during the early Holocene sea-level rise (Bruno et al., 2017). Slight dispersion
472 of data may reflect post-burial deformation of paleosol PH by differential subsidence and
473 neotectonics (Amorosi et al., 2021). In the area under marine influence (shades of blue in Fig. 10),
474 burial ages decrease landwards from ~11.5 to 5.4 kyr B.P.. This overall spatial trend is not recorded
475 in two isolated areas, where paleosol PH and older paleosols are upbended (see also Fig. 5C). In
476 cross-sections, the progressive drowning of paleosol PH is testified by onlap terminations of

477 Holocene flooding surfaces (Fig. 5A). These horizons, associated to prominent facies backstepping,
478 are coeval to the main Holocene melt water pulses (MWP-1b, 1c, 1d; Fig. 7B), which forced the
479 landward migration of the Po estuary across the study area. The effects of MWP-1A are not recorded
480 in the study area, which was too far from the coastline at that time (Storms et al., 2008).

481 Pedo-sedimentary features of paleosol PH reflect this evolutionary history. Carbonate
482 redistribution along the soil profile and relatively low amounts of organic carbon in the A horizons
483 (Amorosi et al., this volume) are indicative of a low groundwater table (Kalisz et al., 2010), enhanced
484 by fluvial incision during the early phases of soil development (Tebbens et al., 1999). Preserved plant
485 debris in the topsoil, radiocarbon dated to the early Holocene (Figs. 4 and 5), reflects the progressive
486 rise in groundwater table (Bohacs and Suter, 1997) up to the setting of waterlogged conditions and
487 to the accumulation of peat beds (Diessel, 1992; Richardson and Vepraskas, 2001; Stolt and
488 Rabenhorst, 2011). The rise of the groundwater table, forced by sea-level rise (Kosters and Suter,
489 1993; Törnqvist, 1993; Wadsworth et al., 2003), led to the progressive filling of Apennine fluvial
490 incisions by fine-grained and organic-matter-rich estuarine strata (facies SM, cores RN3 and EM2 in
491 Fig. 5C). The lack of fluvial sands (facies FCR) at the base of fluvial incisions, suggests that valley
492 filling was promoted by eustasy rather than by an upstream increase in sediment supply (Kvale and
493 Archer, 2007).

494 Beyond the area of marine influence, at elevations > -3 m, the burial age of paleosol PH is
495 substantially unpredictable (Figs. 9 and 10). The dispersion of burial dates in this upstream sector
496 reflects the dynamics of Holocene sedimentation, dominated by avulsion processes and by the
497 deposition of poorly extended alluvial units (Figs. 5 and 6). This aspect is particularly evident in the
498 Bologna area (Fig. 6): in core 16, for example, paleosol PH was already buried around 8.2 kyr B.P.,
499 when paleosol H1 was still developing (see also Fig. 4); in the nearby core 15, at a distance of just
500 224 m, the burial of paleosol PH occurred only around 7 kyr B.P.. The transition to an avulsive

501 sedimentation pattern at the onset of the Holocene, recorded in several worldwide fluvial systems
502 (Phillips, 2011; Stouthamer et al., 2011), is an indirect consequence of the landward migration of
503 the coastline, which dramatically reduced the area available for sediment storing and lowered rivers
504 longitudinal gradients. Decreased stream power, and capability to erode banks and create space for
505 sediment storage, led to river aggradation, crevassing and avulsions (Blum et al., 2013).

506 The discontinuity of Holocene paleosols (Figs. 5 and 6) reflects the limited areal extent of the
507 sedimentary units on which they developed. Avulsive sedimentation patterns and crevassing also
508 determined laterally variable maturity of Holocene paleosols. Indeed, soil development was
509 interrupted only in restricted areas affected by alluvial deposition. In sediment-starved areas, soils
510 continued to evolve. Lateral changes in soil maturity are observable in modern soils, which started
511 to develop during the Late Bronze age (areas where H2 is still exposed – Fig. 5), and after the Middle
512 Ages (areas where H2 is buried - Fig. 5).

513 The peak in radiocarbon dates from fluvial channel-related deposits, centred around 5-6 kyr B.P.
514 (Fig. 7), corresponds to a generalized re-organization of the river network at the onset of
515 Neoglaciation (Piovan et al., 2012; Bruno et al., 2015; Amorosi et al., 2017b), which led to the burial
516 of paleosol H1. Slope instability and enhanced sediment production, recorded in the Northern
517 Apennines since 5.5 kyr B.P. (Soldati et al., 2006; Picotti and Pazzaglia, 2008), likely reflect
518 widespread deforestation of the Apennine drainage basins during the early Eneolithic (Cremaschi
519 and Nicosia, 2012). The progressive return to stable floodplain conditions was paralleled by
520 improved techniques of land reclamation and management from the late Bronze age to late
521 Antiquity (Bruno et al., 2013).

522

523

524 6. CONCLUSIONS

525

526 Based on the sedimentological analysis and correlation of more than 170 core data, with the
527 chronological constraint of 376 radiocarbon dates, this study provided new data on: (i) the complex
528 relationships between pedogenetic, fluvial and costal processes in alluvial and paralic environments;
529 (ii) the relative influence of global (i.e. climate and eustasy) and local (e.g. river avulsions) factors on
530 soil development and burial; (ii) the use and limitations of radiocarbon to assess the exposure age
531 of paleosols.

532 In the Upper Pleistocene-Holocene mud-prone succession of the Po Basin, vertically stacked,
533 weakly developed paleosols testify to intermittent pedogenesis, periodically interrupted by
534 overbank sedimentation. Radiocarbon analyses permitted to define the duration of the main phases
535 of pedogenesis and to compare them to climate and eustatic curves.

536 Radiocarbon dates from paleosols (bulk-soil and plant-debris samples) and from underlying and
537 overlying sedimentary units indicate exposure periods encompassing 4-6 millennia. This estimate is
538 consistent with pedological features (e.g. type of secondary calcite, lack of illuvial clay, limited
539 rubefaction), which denote incipient pedogenesis. Stratigraphically ordered ¹⁴C calibrated ages
540 within A horizons revealed that Po Plain paleosols are condensation horizons, which experienced
541 slow aggradation during exposure (0,05-0,23 mm/yr) due to accumulation of plant litter, with a likely
542 contribution of wind-transported dust.

543 Millennial-scale climate oscillations and glacio-eustasy concurred in driving the pedo-
544 sedimentary evolution of the Po Plain during the Late Pleistocene. In particular, during the MIS 3,
545 pedogenetic events were mainly controlled by changes in vegetation driven by climatic oscillations
546 at the scale of Bond cycles. Paleosols developed in well-drained floodplain environments, dissected
547 by fluvial incision, during relatively warm periods. Overbank sedimentation and paleosol burial

548 occurred during colder phases. During the Last Glacial Maximum, high-sediment-supply hindered
549 pedogenesis and led to the accumulation of 3-10 m-thick overbank strata.

550 Widespread soil development (paleosol PH) occurred at the end of the Last Glacial Maximum,
551 following glaciers retreat and afforestation of drainage basins. Burial of paleosol PH was
552 diachronous as indicated by radiocarbon dates from vegetal remains preserved at the topsoil. At
553 distal locations, paleosol PH was progressively drowned and buried under estuarine sediments
554 during the Holocene phases of post-glacial sea-level rise. Beyond the area of marine influence, burial
555 ages of paleosol PH change from a place to another without specific spatial trends, reflecting
556 upstream fluvial sedimentation dominated by avulsions.

557 Holocene (H) paleosols, observed at inland locations, grade seaward into coeval estuarine and
558 deltaic deposits. These pedogenized horizons show a poor correlation potential and laterally
559 variable degree of maturity that reflect avulsive sedimentation patterns and widespread crevassing.
560 Human activities (e.g. deforestation and land reclamation) became an increasingly important factor
561 affecting pedo-sedimentary processes during the last 6 kyr.

562 This study highlights the potential for paleoenvironmental reconstructions of upper Quaternary
563 sedimentary successions. The pedo-stratigraphic approach adopted here, relying upon a robust
564 chronological framework, permitted to assess the temporal and spatial scale of pedogenetic events
565 and to discern the relative contribution of distinct controlling factors that typically act
566 contemporaneously.

567

568

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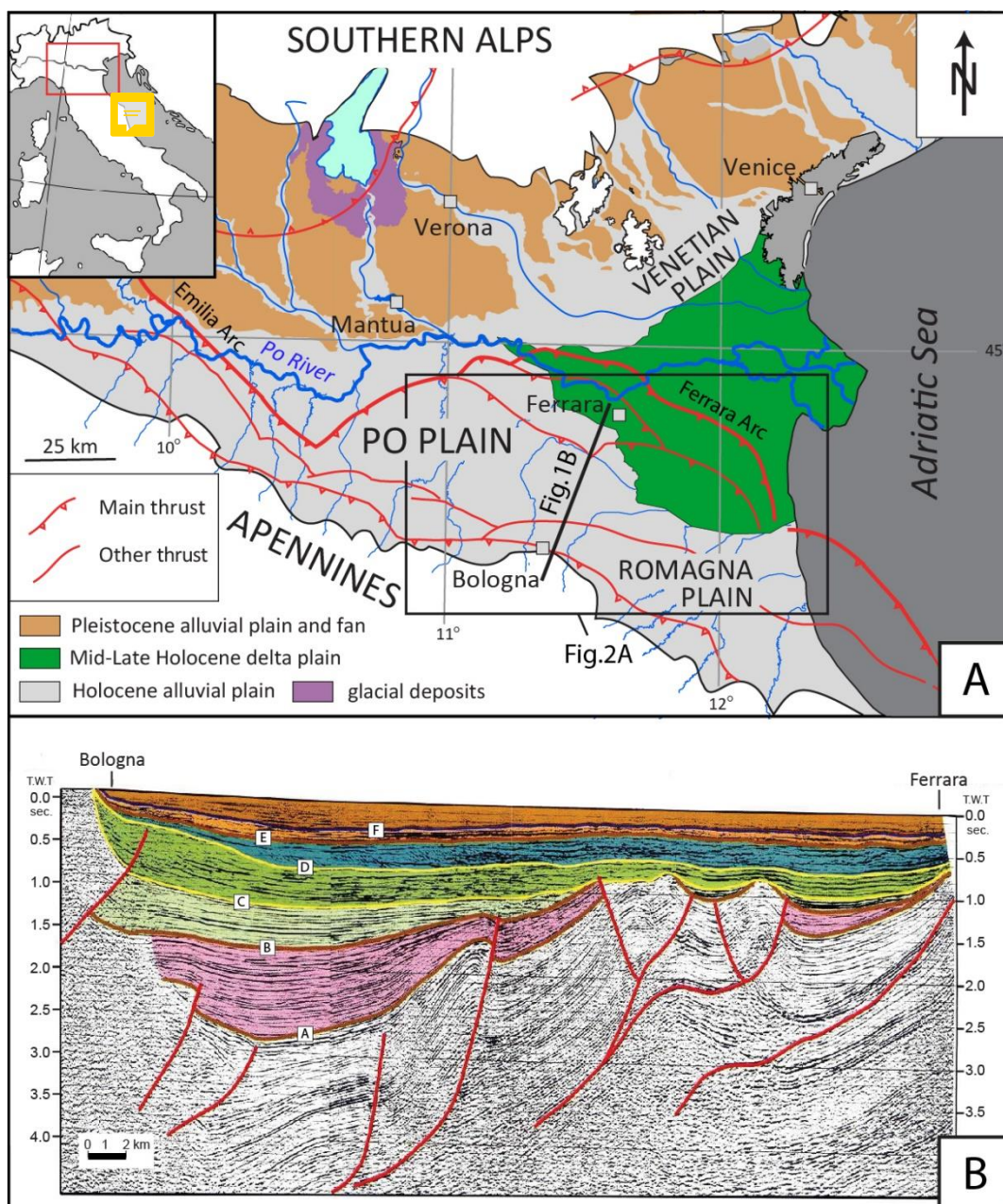
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Figures and captions

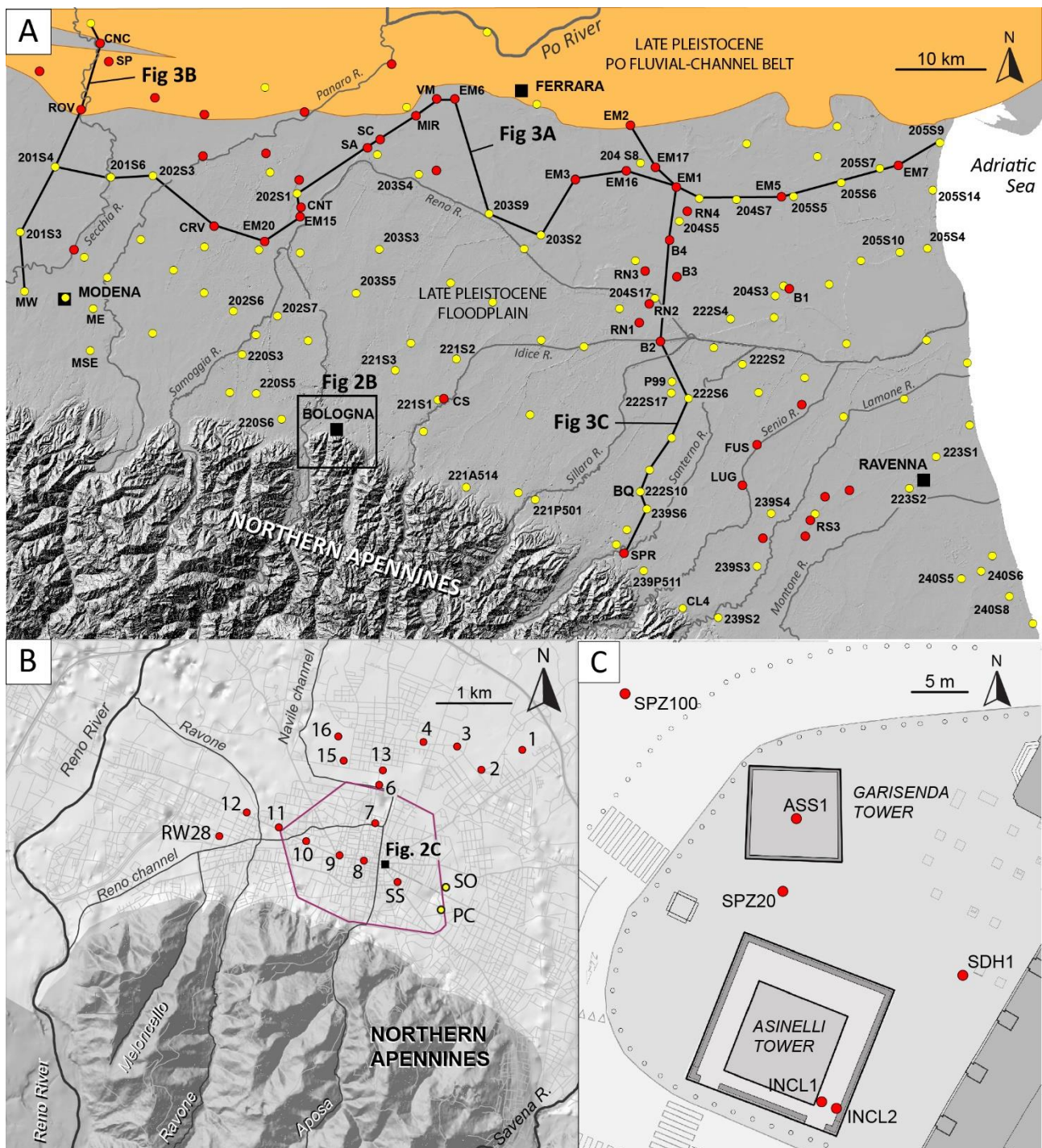
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1013 Fig. 1 – A. Geological map of the Po Plain, with location of the study area and of the seismic profile
1014 of Fig. 1B (modified after Bruno et al., 2019). The extent of Pleistocene alluvial deposits is from
1015 Fontana et al. (2014). Buried structures are modified after Burrato et al., (2003). B. Interpreted deep
1016 seismic profile (from Regione Emilia-Romagna & Eni-Agip, 1998, location in Figure 1A), with
1017 identification of the major blind thrusts (red lines) and main stratigraphic unconformities (lines from
1018 A–F) within the Po Basin fill (coloured area).

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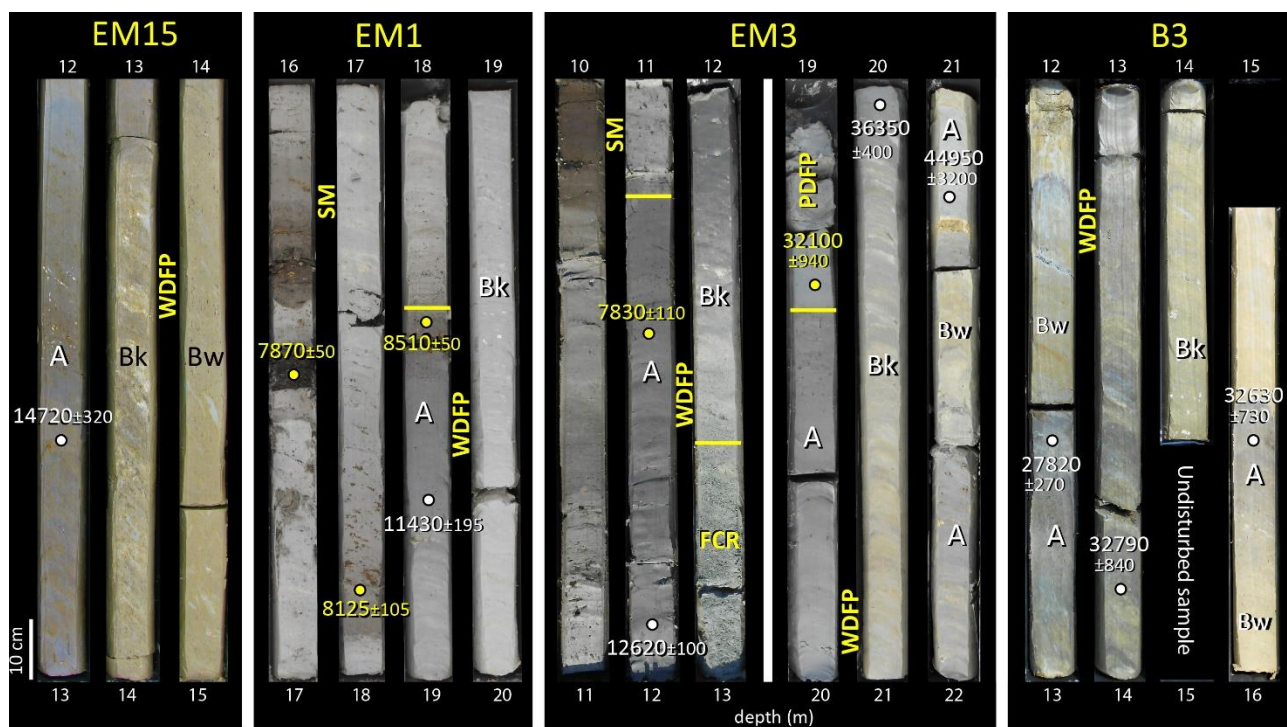
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Fig. 2 – A. Study area with location of cores (red circles), core descriptions (yellow circles) and cross-sections of Fig. 5. B. Close-up on the Bologna area, with location of cores shown in Fig. 6. C. Location of cores drilled in the Two-Towers Medieval district (modified after Bruno et al., 2020b).



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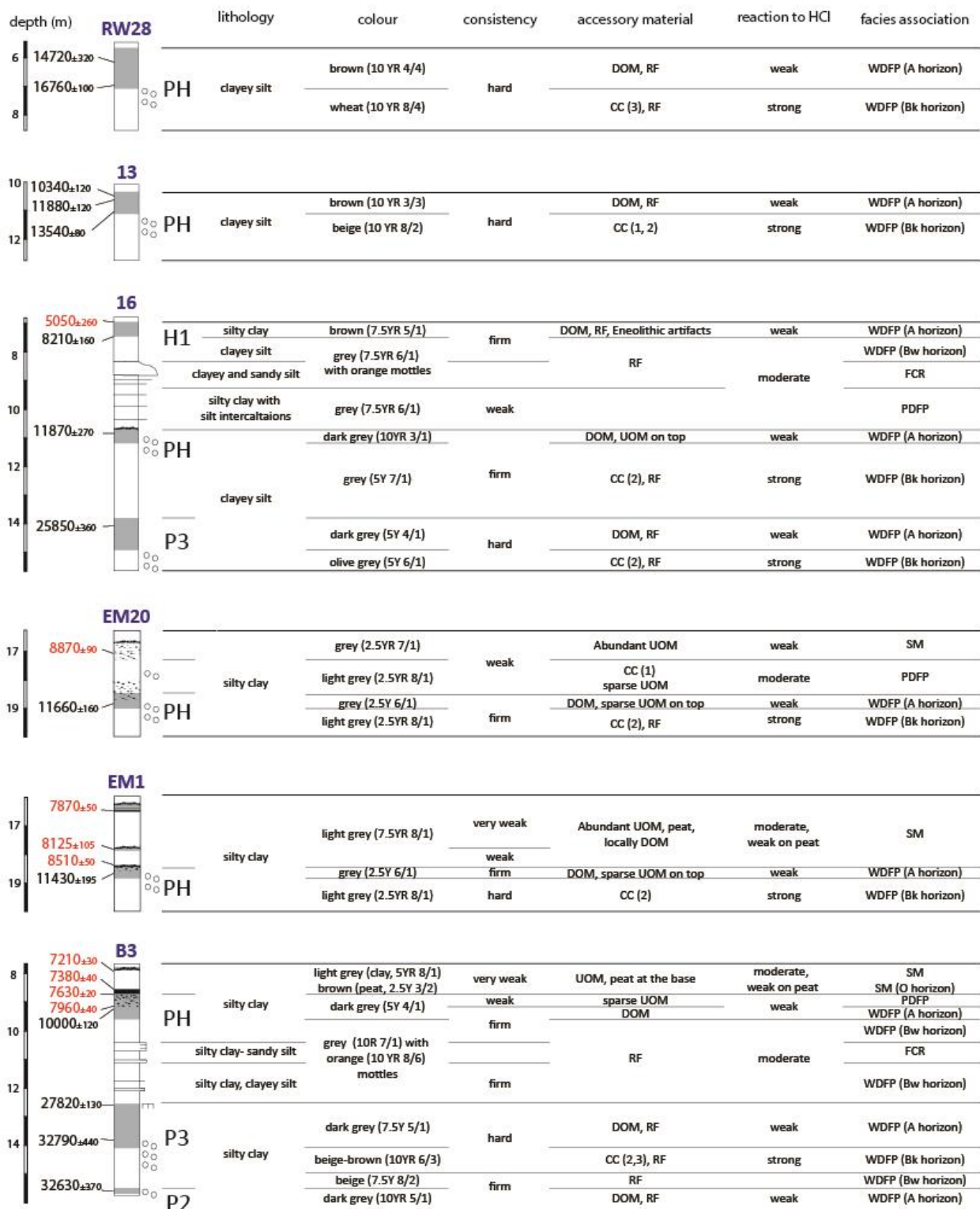
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Fig. 3 – Representative photographs of paleosols from cores EM1, EM3 and B3 (location in Fig. 2).

WDFP: well-drained floodplain; PDEF: poorly-drained floodplain; SW: swamp; FCR: fluvial-channel-related deposits. For pedological horizons A, Bk and Bw, see text. The white dates (cal kyr B.P.) are from bulk-sediment samples, whereas the yellow ones are from plant macrofossils.



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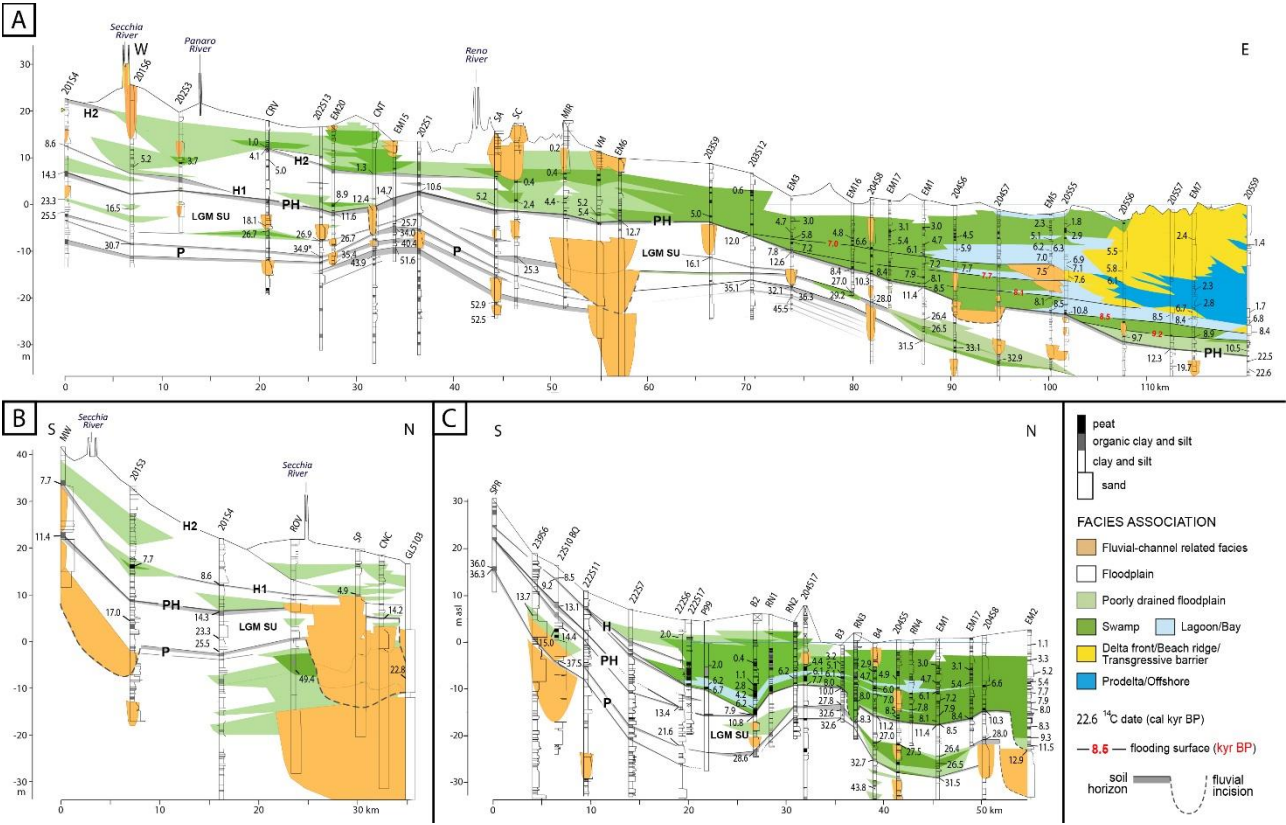
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Fig. 4 – Sedimentological characters and radiocarbon ages of paleosols P2, P3, PH and H1 from cores RW28, 16, 17, EM1, EM20 and B3 (for location see Fig. 2). DOM: dissolved organic matter; UOM: undecomposed organic matter; RF: redoximorphic features; CC carbonate concretions (the number indicates the evolutionary stage, from Gile et al., 1981; Machette et al., 1985). WDFP: well-drained

1036 floodplain; PDFP: poorly-drained floodplain; SW: swamp; FCR: fluvial-channel-related deposits. The
1037 black dates (cal kyr B.P.) are from bulk-sediment samples, whereas the red ones are from plant
1038 macrofossils.

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1041 Fig. 5 – Upper Pleistocene and Holocene stratigraphy of the Po Plain depicted in cross-sections with
1042 W-E (A) and S-N (B and C) orientations (location in Fig. 2A). LGM SU: Last Glacial Maximum
1043 Stratigraphic Unit.

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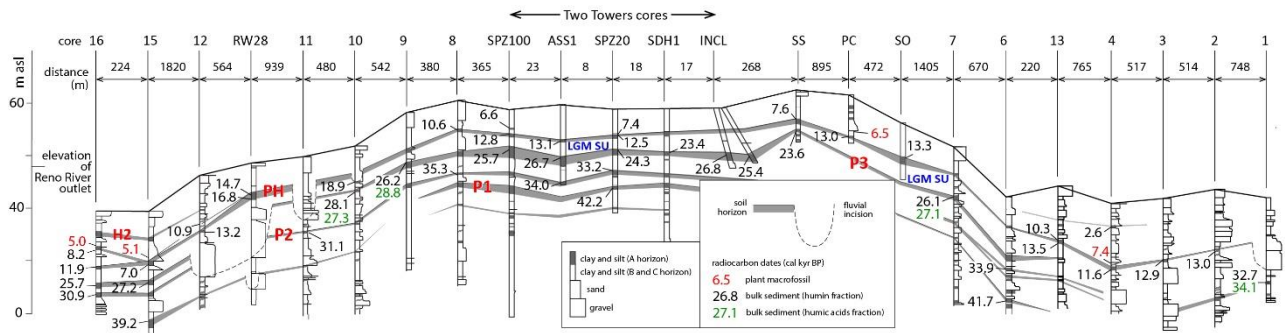
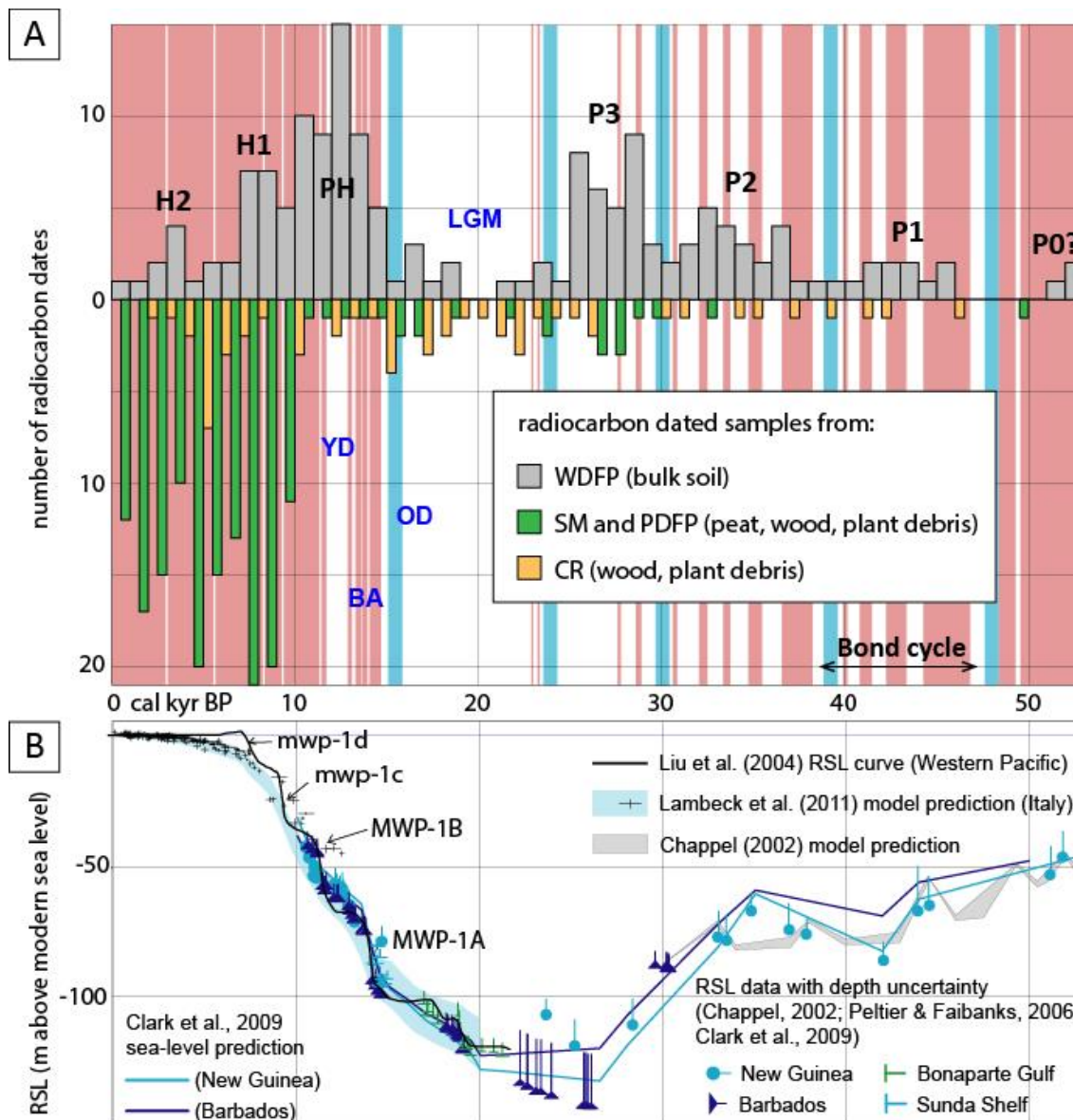


Fig. 6 – Paleosol-based correlations in the Bologna Area. LGM SU: Last Glacial Maximum Stratigraphic Unit. Location of cores in Fig 2.



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1053 Fig. 7 – A. Distribution of radiocarbon dates from bulk-soil and plant-macrofossil samples. A major

1054 peak in dates from bulk-soil samples corresponds to paleosol PH; five minor peaks are related to

1055 paleosols P1, P2, P3, H1 and H2. Notable is the lack of radiocarbon dated soils around 20 cal ky B.P.

1056 (LGM). The sharp increase in radiocarbon dates from plant-macrofossils after 10 ky B.P., paralleled

1057 by a decline in the number of dates from bulk-soil samples, reflects the onset of estuarine-deltaic

1058 sedimentation in the distal Po Plain and the burial of paleosol PH. Red areas correspond to

1059 interstadials, according to Bond et al., (1997) and Rasmussen et al. (2014). OD: Oldest Dryas; YD:

1060 Younger Dryas. Light blue vertical bars indicate Henrich events (Bond et al., 1992). B. Sea-level

1061 oscillations during the last 50 ky.

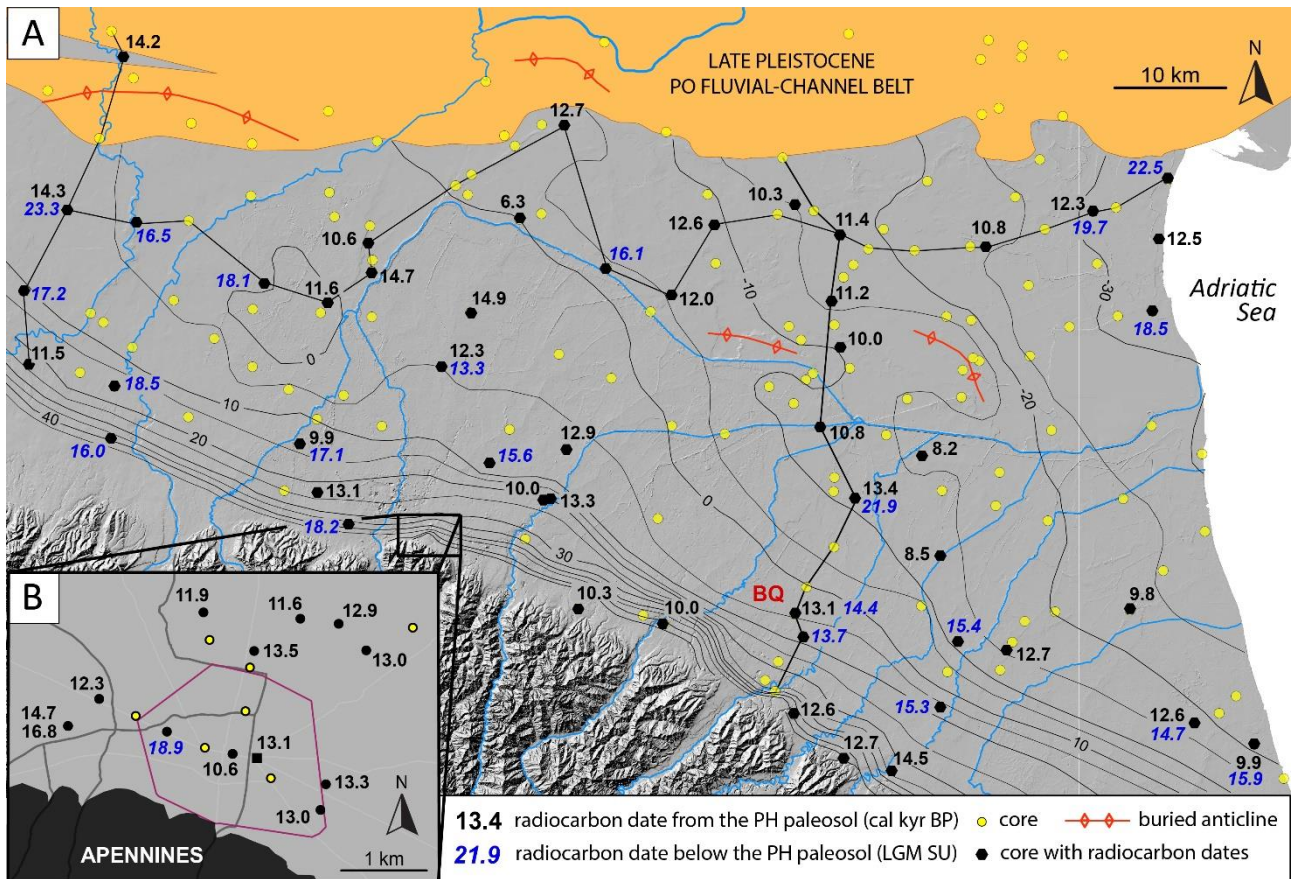
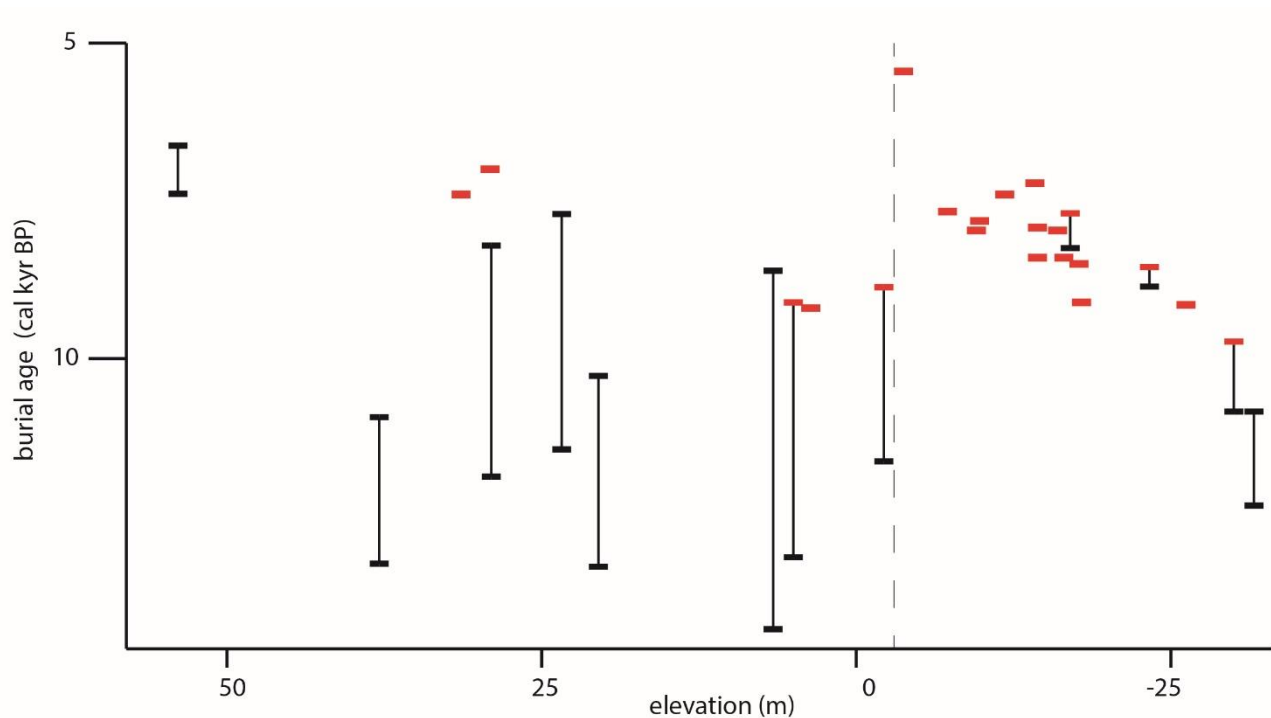


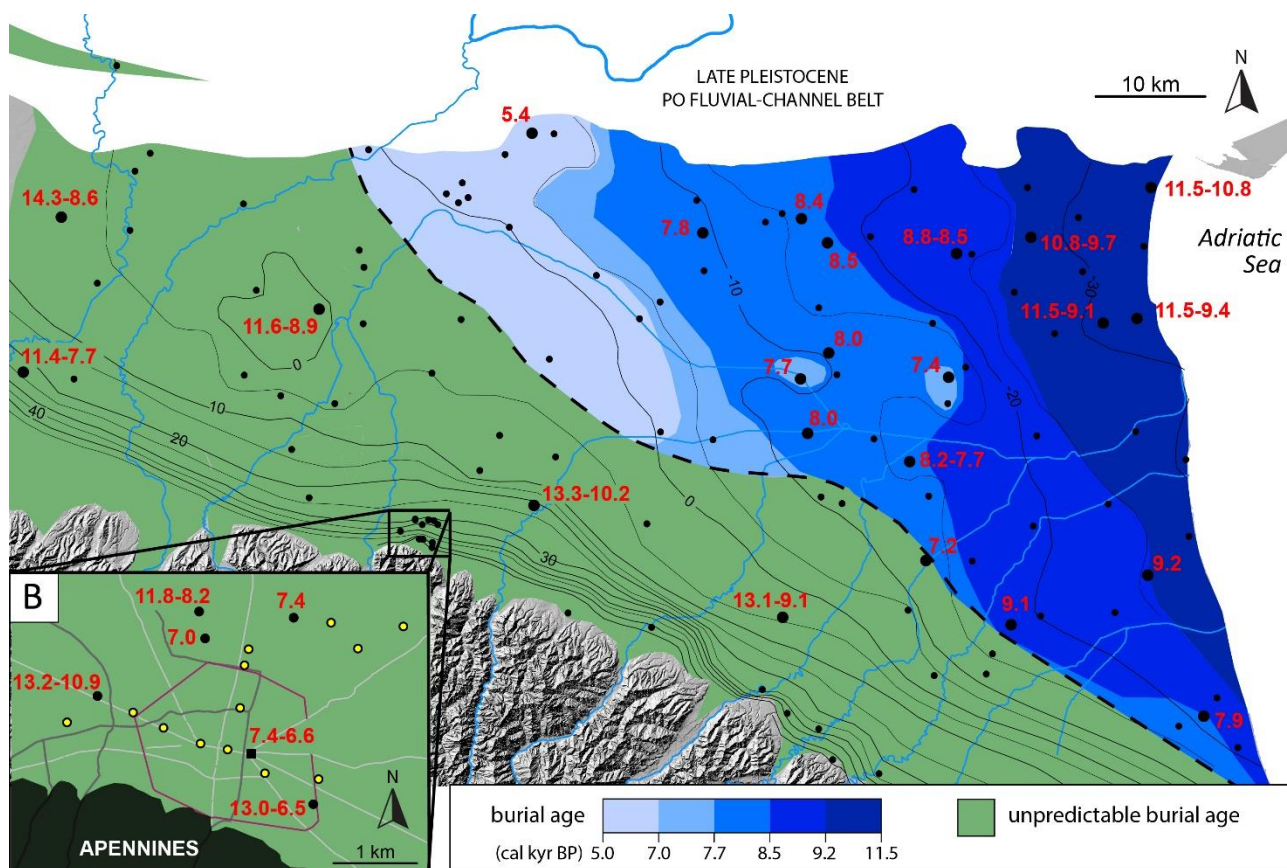
Fig. 8 – Radiocarbon dates (black numbers, cal ky B.P.) from paleosol PH (bulk soil samples) in the Po Plain (A) and in the Bologna urban area (B). Blue numbers are the youngest ages from the underlying LGM stratigraphic unit. The present-day elevation (metres above modern sea-level) of paleosol PH is depicted through 5 m-spaced isolines. BQ: Bubano Quarry.



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1069 Fig. 9 – Burial ages of paleosol PH plotted against its present-day elevation (metres above modern
 1070 sea-level). Black rectangles are radiocarbon dates from bulk soil samples. Red rectangles are
 1071 radiocarbon dates from plant macrofossils. Burial ages from samples at elevation < - 3 m (dashed
 1072 line) show an overall inverse proportionality with elevation. On the contrary, no specific trends are
 1073 observable in burial-age distribution at elevation > - 3 m.

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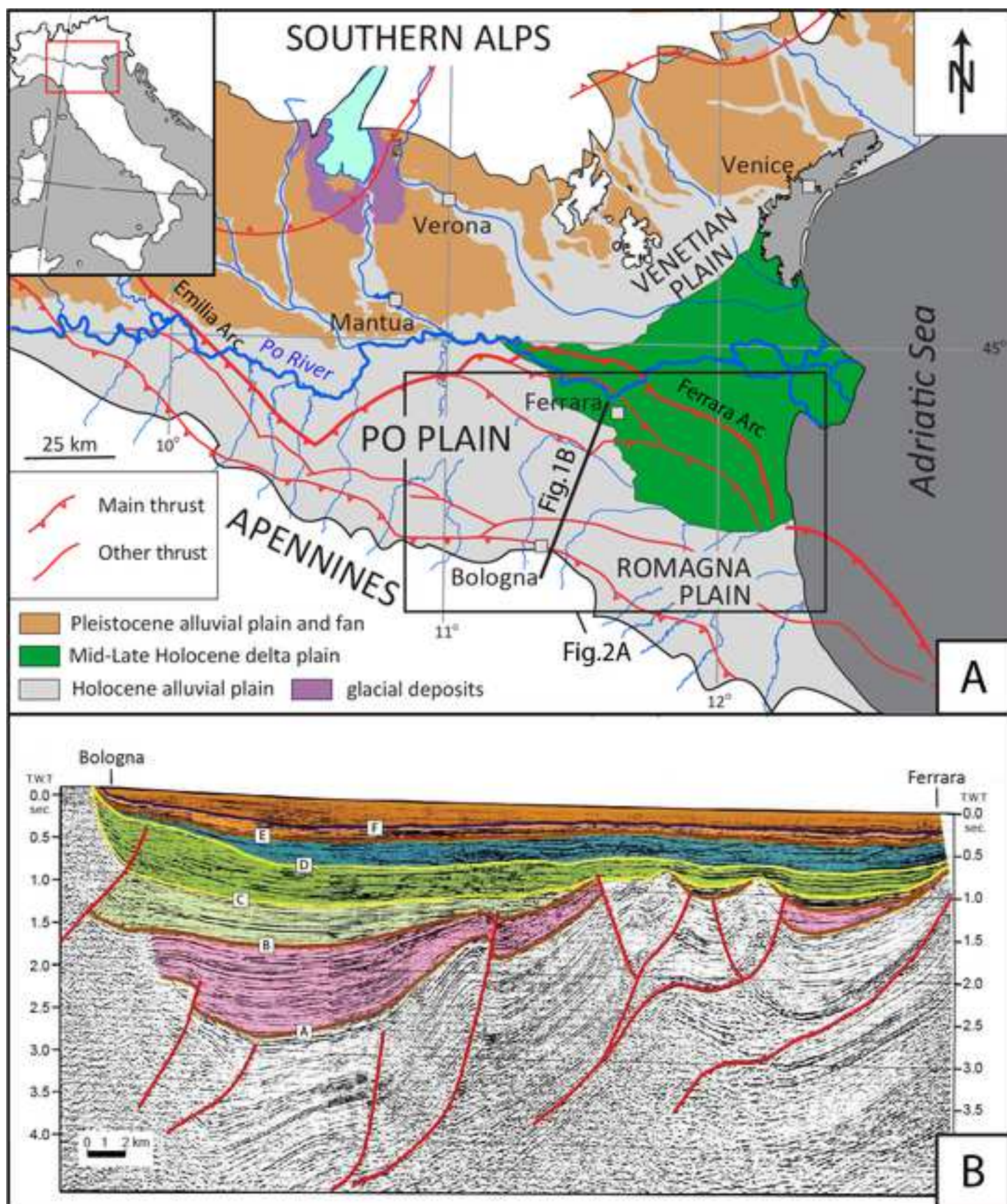
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Fig. 10 – Map showing burial ages (cal kyr B.P.) of paleosol PH in the Po Plain (A) and in the Bologna urban area (B). The shades of blue depict progressively younger burial ages upland. In the green area burial ages do not show specific spatial trends. Red numbers represent burial ages or intervals measured along single cores (black circles). The map also depicts the present-day elevation (metres above modern sea-level) of paleosol PH through 5 m-spaces isolines. Black dots are the countered data points. The dashed line represents the maximum landward extent of Holocene estuarine and delta plain deposits.

Figure 1



A

Fig 3B

Fig 3A

Fig 2B

Fig 3C

Fig. 2C

Fig. 2A

Fig. 2B

Fig. 2C

Fig. 2D

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Fig. 2AB

Fig. 2AC

Fig. 2AD

Fig. 2AE

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Figure 1 displays four panels showing vertical profiles of soil properties (EM15, EM1, EM3, B3) versus depth (m). Each panel displays three vertical columns of data. The top column shows soil color (A, Bk, Bw) and the bottom column shows soil texture (SM, WDFP, FCR). The middle column shows soil moisture (SM) and soil texture (WDFP). The rightmost column shows soil texture (WDFP) and soil moisture (SM). The depth scale is in meters (m).

Panel	Depth (m)	Soil Color	Soil Texture	Soil Moisture (SM)	Soil Texture (WDFP)	Soil Moisture (SM)	Soil Texture (WDFP)
EM15	12	A					
	13	Bk					
	14	Bw					
EM1	16						
	17						
	18						
	19						
EM3	10						
	11						
	12						
	13						
B3	12						
	13						
	14						
	15						

Figure 4

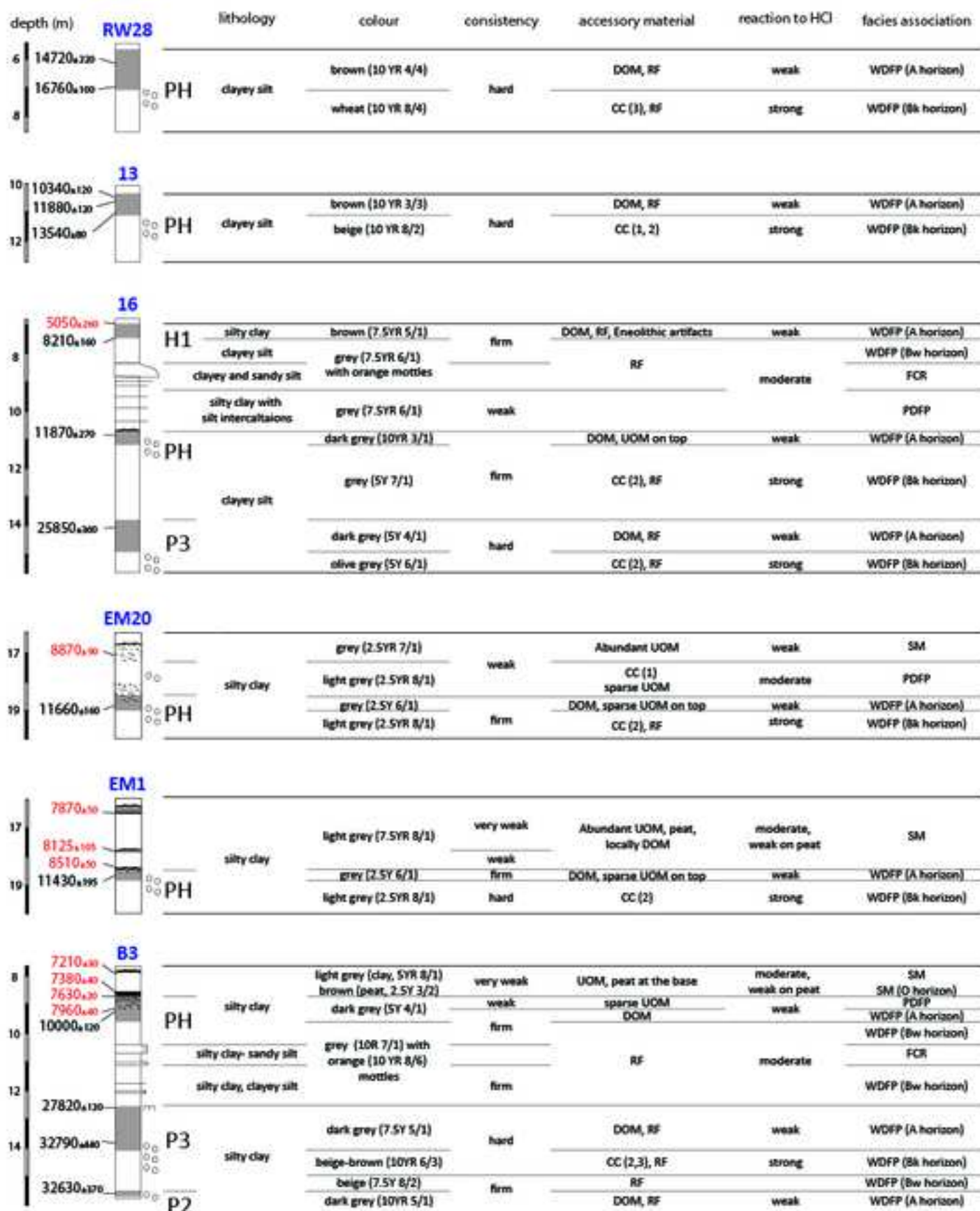


Figure 6

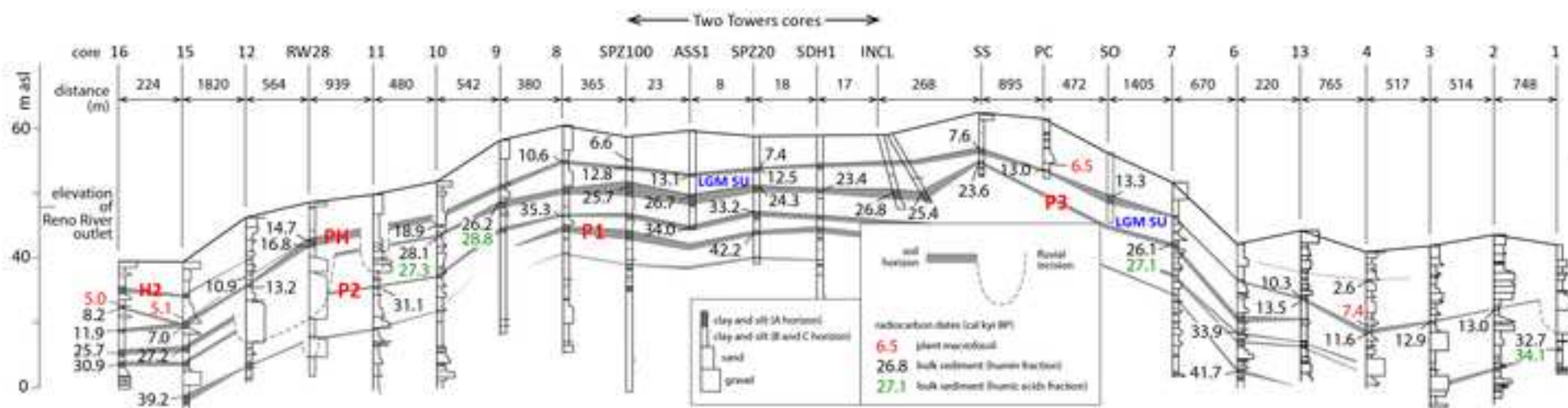


Figure 7

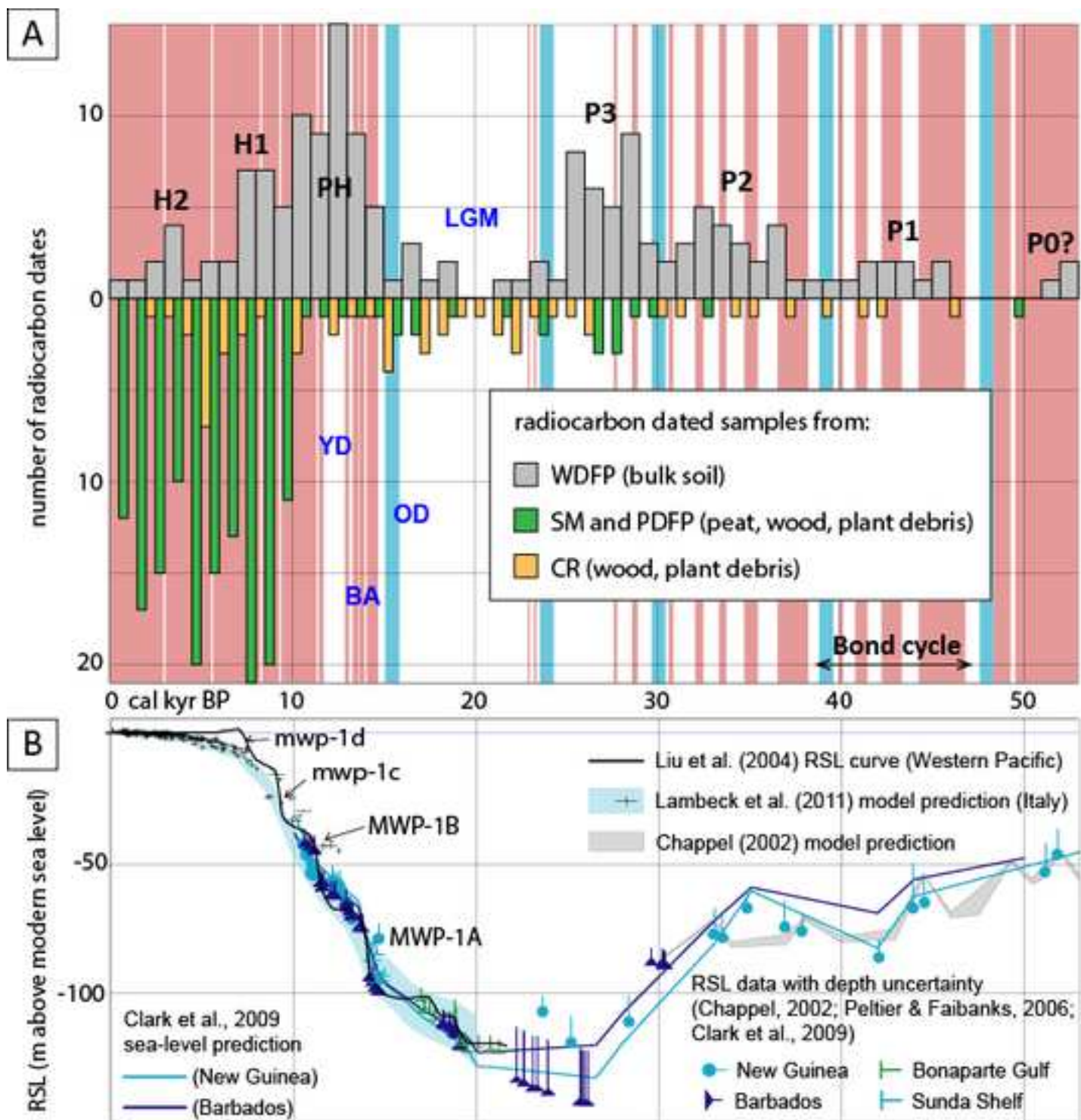


Figure 8

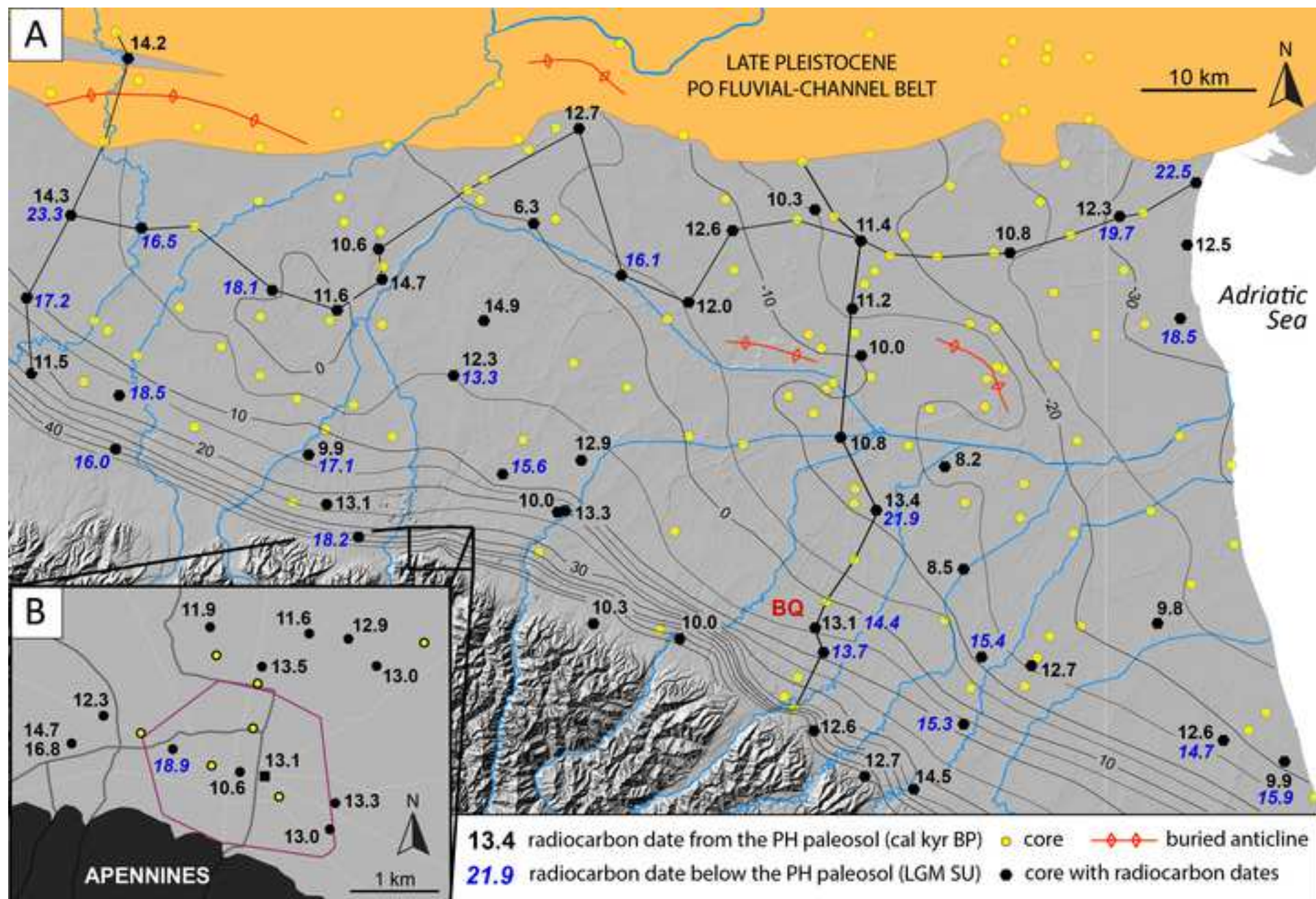


Figure 9

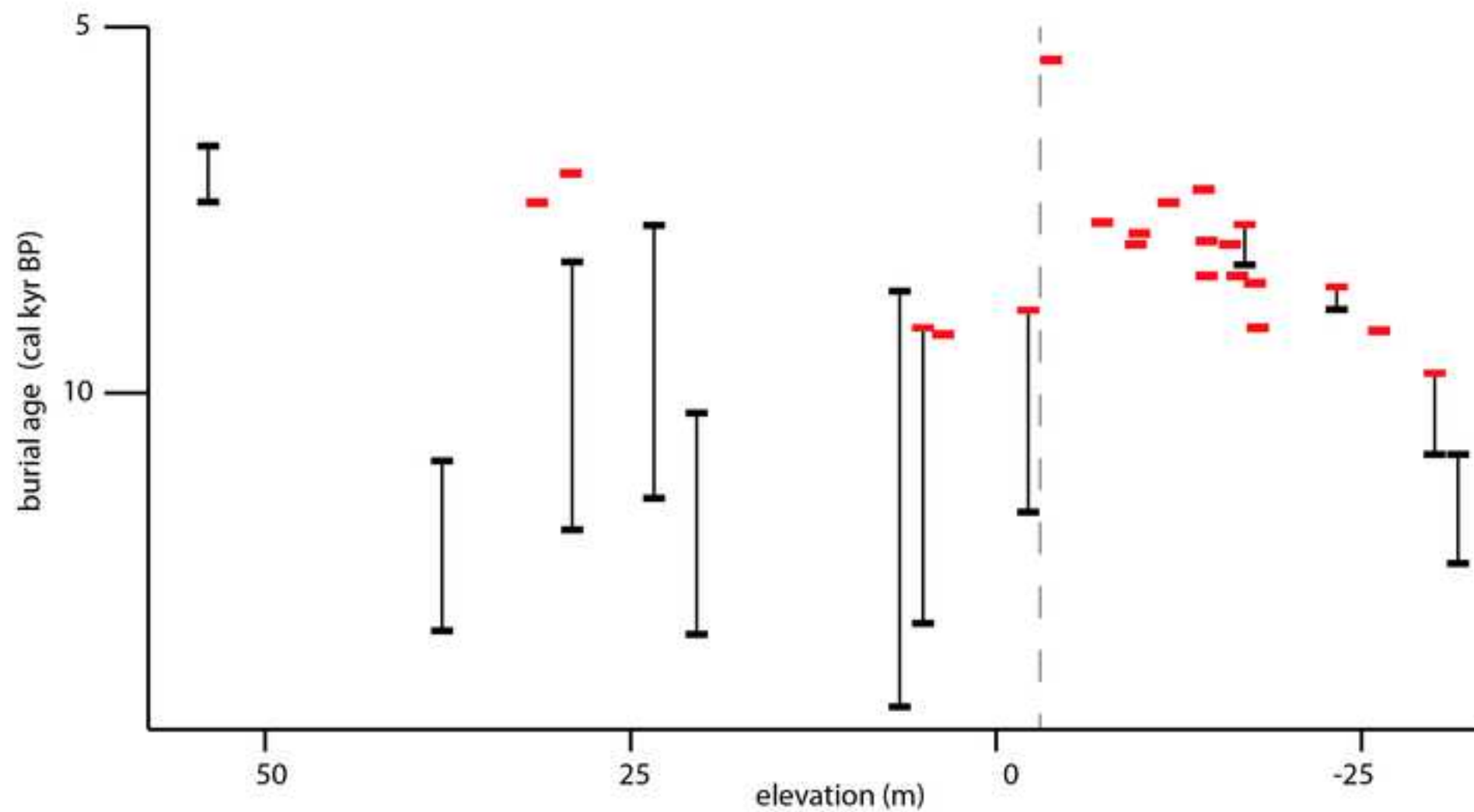
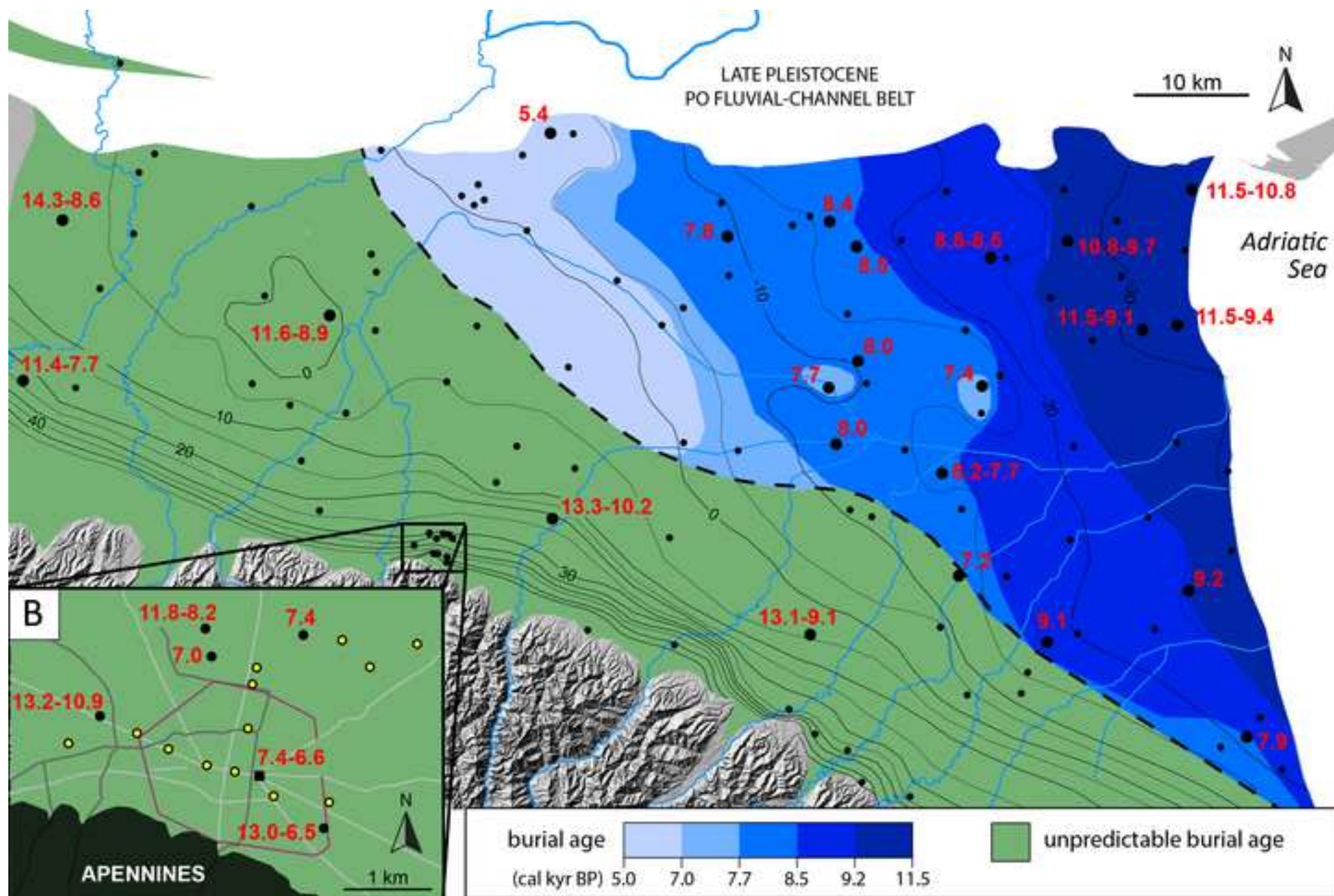


Figure 10



Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: