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Petro-chemical features and source areas of volcanic aggregates used in ancient Roman maritime concretes

F. Marra [a,*], M. Anzidei [a], A. Benini [b], E. D'Ambrosio [c], M. Gaeta [d], G. Ventura [a], A. Cavallo [e]

[a] Istituto Nazionale di Geofisica e Vulcanologia, Via di Vigna Murata 605, 00143 Rome, Italy
[b] Via Traù 3, Rome, Italy
[c] Via Cupa 5, Nettuno, 00048, Rome, Italy
[d] Dipartimento di Scienze della Terra, Sapienza-Università di Roma, P.le Aldo Moro 5, 001859 Rome, Italy
[e] CERTEMA - Laboratorio Tecnologico di Grosseto. Borgo S. Rita, Cinigiano (GR), Italy

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ABSTRACT

We present and discuss data from petrographic observation at the optical microscope, electron microprobe analyses on selected glass shards, and trace-element analyses on 14 mortar aggregates collected at the ancient harbors and other maritime structures of Latium and Campania, spanning the third century BCE through the second CE, aimed at identify the volcanic products employed in the concretes and their area of exploitation. According to Latin author Vitruvius assertion about the ubiquitous use of Campanian pozzolan in the ancient Roman sea-water concretes, results of this study show a very selective and homogeneous choice in the material employed to produce the concretes for the different investigated maritime structures, evidencing three main pumice compositions, all corresponding to those of the products of the post-Neapolitan Yellow Tuff activity of the Phlegraean Fields, and a systematic use of the local Neapolitan Yellow Tuff to produce the coarse aggregate of these concretes. However, mixing with local products of the Colli Albani volcanic district, located 20 km east of Rome, has been evidenced at two fishponds of Latium, in Punta della Vipera and Torre Astura. Based on these petrographic and geochemical data, we conclude that the selective use of pozzolan from Campania, rather than of unproved different chemical properties, was the consequence of a series of logistic, economic, industrial and historical reasons.

1. Introduction

The selective use of volcanic aggregates (pozzolan) exploited in the surroundings of *Puteoli* (Pozzuoli), deriving from the products of the activity of Phlegraean Fields and/or Vesuvius for building maritime structures in ancient Roman times is stated by Latin author Vitruvius in the book *De Architectura*. This assumption has been so far verified on a limited number of mortar aggregates collected in the harbors of Cosa (Jackson et al., 2010), Ostiae (Port of Claudius) and Anxur (modern Terracina) (D'Ambrosio et al., 2015) by means of trace element signature and EMP glass analyses.

Vitruvius (1999) uses the term "*pulvis*" (powder) to describe the material occurring in the region of Baiae (a village few kms southwest of *Puteoli* on the coast of the homonymous gulf, see Fig. 1) and in the area around Vesuvius, which "mixed with lime and rock fragments not only confers strength to the constructions, but is capable to solidify under water" (*De Arch*., 2. 6. 1). By contrast, he uses the definition of "*harena*" (sand) for the material used to produce mortar to be used in mainland constructions, despite recognizing the same volcanic origin for both: (…) And thus, just in Campania scorched earth becomes ash, so in Etruria the cocked matter becomes *carbunculus*. [Carbunculus, which translates as "charcoal", is indeed one type of *harena fossicia* (pit sand) that has been identified (D'Ambrosio et al., 2015) as deriving from the pyroclastic-flow deposits erupted by the Vico and Monti Sabatini volcanic districts of Latium (Fig. 1)] "*Both of these are outstanding for constructions, but one works in buildings on land, while the other works as well for sea moles.* (…) (*De Arch.*, 2.6.6) (D'Ambrosio et al., 2015, and references therein). This latter feature characterizes the present "hydraulic mortar", as opposite to the "aerial mortar", which cannot set underwater. However, there is no solid reason to think that the pozzolan from Latium should have a different reaction with lime when setting underwater with respect to that coming from Campania. A broad literature shows that the "hydraulic property" is due to particular chemical reactions with lime of zeolite minerals occurring in the volcanic aggregate (pozzolanic reaction), which confer to a mortar the strong binding power and the hydraulic character (e.g.: Mertens et al., 2009, and references therein). Indeed, zeolites occur in all the volcanic pozzolan of central Italy, and not only Campania.

* Corresponding author.
E-mail address: fabrizio.marra@ingv.it (F. Marra).

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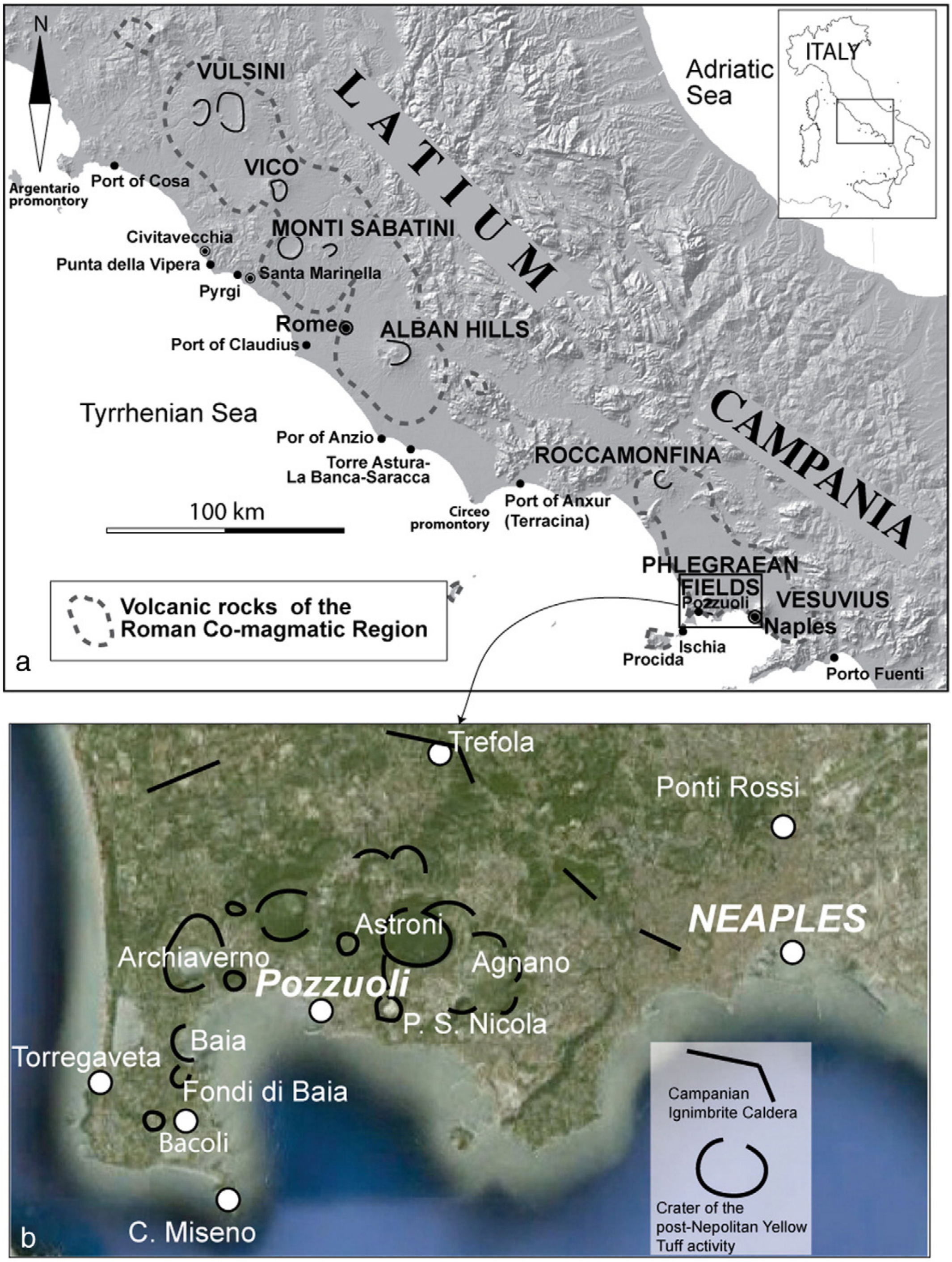

Fig. 1. a) Digital Elevation Map of central Italy showing location of the harbors and other maritime structures for which composition of the volcanic aggregate employed in the concrete has been investigated in the present study. The outcropping area of the volcanic products of the Roman co-magmatic Region (Peccerillo, 2005) is also shown. b) Detail map of the area of Pozzuoli, showing the main eruptive centers of the Phlegrean Fields volcanic activity.

D'Ambrosio et al. (2015) suggest that the selective use of pozzolan from Campania, rather than to unproved different chemical properties, may be due to a series of logistic and historical reasons. Regarding the different term used by Vitruvius for the Campanian pozzolan, *pulvis* as opposed to *harena*, after discarding the hypothesis of a finer grain size of the former, which is not supported by the study on the mortars of the Port of Claudius in Fiumicino and on the Port of Anxur (Terracina), D'Ambrosio et al. (2015) propose that the Latin author considered the whitish to light grey, high vesicular pumice clasts that make up most portion of the Campanian pyroclastic deposits as one particular, lightweight *harena fossicia*, based on the use of the adjective “*laevis*” (light) referred to the *pulvis* (Vitr., *De Arch.*, 2.6.1.).

The present study is aimed to verify the extensive use of Campanian pozzolan to produce the maritime concretes in the Tyrrhenian Sea, which a vast literature has hypothesized to extend to the whole Mediterranean region (e.g. Oleson et al., 2004; Vola et al., 2011; Brandon et al., 2014), and to investigate the possible scientific, practical, and historical reasons. At this scope, in order to integrate previous literature data, we sampled and analyzed 14 mortar aggregates collected at the ancient harbors and other maritime structures built between the third century BCE and the first century CE in Punta della Vipera, Pyrgi, Torre

Astura, and Anzio (Latium), in Ischia, and Punta Fuenti (Campania) (Fig. 1), plus one tuff caementa collected at the second century CE port of Terracina. Here we discuss data from petrographic observation at the optical microscope, electron microprobe analyses (EMPA) on selected glass shards of pumices and tuff fragments, and trace-element analyses on samples of mortar aggregates and tuff *caementa* collected at the

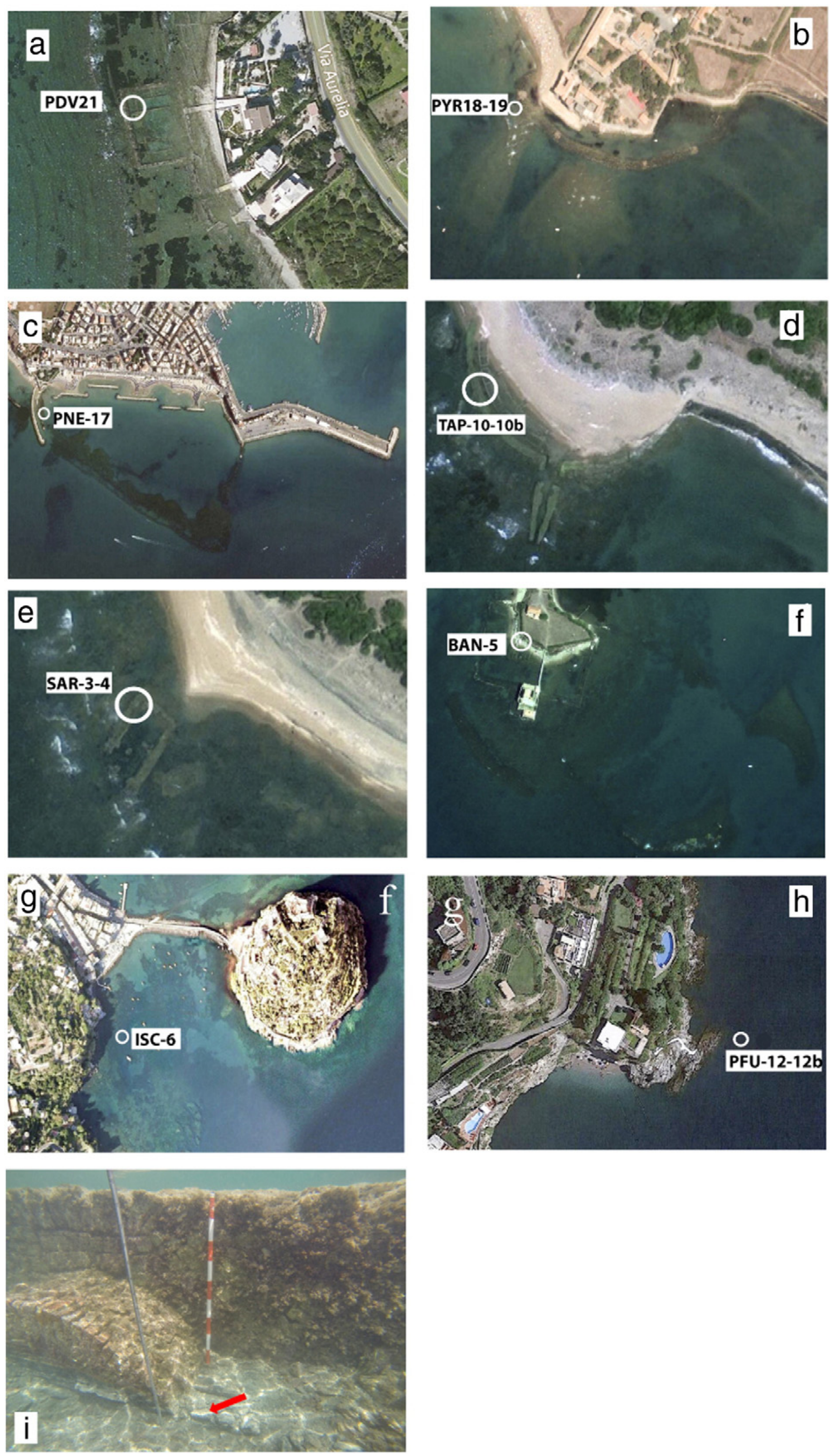

Fig. 2. Satellite photographs showing detail location of the sampled maritime structures (see also Fig. 1). a) Punta della Vipera; b) Pyrgi, photograph of the sampled portion of the fish tank; c) Anzio; d) Torre Astura; e) La Saracca; f) La Banca; g) Ischia; h) Porto Fuenti; i) underwater photo of the Roman age fish tank at punta della Vipera. The red arrow shows the sampling point of concrete, which has been collected under the paving covering the crepido (i.e. the narrow sidewalk running into the basins).

abovementioned ports and maritime structures, aimed at identifying the volcanic products employed in the concretes.

2. Methods

2.1. Archaeological sampling

Fig. 2 shows the detailed location of the investigated maritime structures and the position of the samples collected from the seawater concretes. A list of the samples and the type of analyses performed is reported in Table 1. For each of the investigated sites, we have selected the significant building parts that included mortar. A few cubic centimeters of sample, suitable for the analysis, have been extracted using a little chisel, without damaging the archaeological site. Mortar has been then retained in sealed cases and later analyzed.

2.1.1. Punta della Vipera

Punta della Vipera is the northernmost investigated maritime structure, located between Civitavecchia and Santa Marinella towns (Fig. 1a). Here a large rectangular fishpond, 55 × 34 m, is presently submerged at 1.28 ± 0.20 m (Lambeck et al., 2004). The jetty is constituted by a thick wall in *opus caementicium* (concrete) closing it on three sides. Walls faced in *opus reticulatum* with top 1 m below the height of the jetty subdivide the pond in many tanks. A circular tank is also located within the central rectangular section. Eleven feeding channels, originally covered with concrete vaults, open in the perimeter wall.

Walls in *opus reticulatum* date the fishpond to the first century BCE. Later restorations made in brick masonry refer to the first century CE or later (Higginbotham, 1997). The sample of concrete has been collected at the base of the wall in opus reticolatum separating the fish tanks, just under the pavement covering the lower *crepido* (Fig. 2i). The latter is the narrow footwalk built along the inner basins that correspond to the upper limit of sea level at the time when the site was built (Lambeck et al., 2004; Pliny the Elder (23–79 CE) "Naturalis Historia": *marginum eam partem*, *quae aquas spectat*: part of that margin that looks at the water).

A thin section of an untreated mortar sample (PDV) has been performed for petrographic analysis at the optical microscope, and electron microprobe (EMP) matrix glass of pumice clast analysis. Two large (1–2 cm) scoria clasts separated through HCl bathing from the aggregate have been selected for trace-element analysis: one red (PDV-R) and the other one dark grey (PDV-G).

2.1.2. Pyrgi

Pyrgi was one of the three ports of the Etruscan Caere, to which was connected by a road approximately 13 km long. Between 270 and 245 BCE, Pyrgi became a Roman maritime colony and retained its function as a port. It was still used at the time of Trajan and Hadrian, in the second century CE (Enei, 2004).

A mortar sample (PYR-18) of the upper portion of the submerged fish tank (Fig. 2b) at about 1.0 m below s.l. and a tuff *caementa* sample (PYR-19) of the lower portion placed at about 1.4 m below s.l. have been collected and analyzed at the optical microscope and at the EMP. A selected fraction of the fine aggregate separated after HCl bathing from sample PYR-18 and a tuff fragment of sample PYR-19 have been analyzed for trace element composition.

2.1.3. Anzio

The port of Anzio was built by Emperor Nero (54–68 CE) in the second half of the 1st century CE, as testified by Suetonius Tranquillus (1963) (Suet., Nero, 9, V), profiting of the natural creek east of Capo d' Anzio and rooting two piers into the promontory itself. Docks are today submerged. A lighthouse stood at the end of the 700 m long southern pier. The submerged portion of the piers was made in *opus caementicium*, whereas the portions above the sea level were covered by bricks (Felici, 2002, 2006).

A tuff *caementa* sample (PNE) collected in the concrete of the pier at about 1 m below sea level (Fig. 2c) has been separated from the matrix and analyzed.

2.1.4. Torre Astura

The geographer Strabo (1960) (Geogr. V, 3, 6), describing the coast of Southern Lazio, mentions a natural landing in the sea close to the mouth of the river Astura (Fig. 1a). Cicero (*Ad Att*. XII, 17, 1; 19, 1; 36,1; 37, 2; 37; 41, 4) had there a villa in which he lived between 45 and 44 BCE. The remains of a building complex, datable between the end of republican and early imperial age, are visible today. This was a seaside villa, built partly on land and partly on an artificial island, with

Table 1
Analyzed samples; [(1)]data from D'ambrosio et al. (2015).
TE: Trace element analysis; SEM: Electron microprobe matrix glass analysis; in bold: thin section.
CA: Colli Albani volcanin district; VS: Villa Senni pyroclastic-flow deposit; PR. Pozzolane Rosse pyroclastic-flow deposit; PF: Phlegraean Fileds district; NYT: Neapolitan Yellow Tuff; PF-1: pre-Campanian Ignimbrite Phlegraean Fields activity; PF-3: post-NYT Phlegraean Fields activity.
Attribution to the volcanic district based on TE and SEM analyses is provided in the last columns. *SAR-3 displays intermediate composition between PF-3 and PR (see text for discussion). **The PF-1 composition for PYR-18 and SAR-3 based on SEM analysis is not univocally determined (see text for discussion).

Label	Site name	Coordinates	Age	Structure	Aggregate	TE	SEM
PDV 21	Punta della Vipera	42.0488 N, 11.8194 E	I BCE	Fish tank	Fine		PF-3
PDV 21-G	Punta della Vipera			Fish tank	Fine	CA-VS	
PDV 21-R	Punta della Vipera			Fish tank	Fine	CA-VS	
PYR 18	Pyrgi	42.0157 N, 11.9559 E	III-I BCE	Harbor, upper jetty	Fine	PF-3	PF-1**; PF-3
PYR 19	Pyrgi			Harbor, lower jetty	Coarse	PF-NYT	PF-NYT
PCL[(1)]	Port of Claudius		I CE	Harbor dock	Fine		PF-3; PF-1
PC-1[(1)]	Port of Claudius			Harbor dock	Fine	PF-3	
PC-2[(1)]	Port of Claudius			Harbor dock	Fine	PF-3	
PNE-17	Anzio	41.4441 N, 12.6224 E	I CE	Harbor dock	Coarse	PF-NYT	PF-NYT
TAP-10	Torre Astura	41.4085 N, 12.7649 E	I BCE-I CE	Fish tank, crepido	Fine	PF-3	No fresh glass
TAP-10b	Torre Astura			Fish tank, crepido	Coarse	PF-NYT	
SAR-3	Torre Astura-Saracca	41.4208 N, 12.7451 E	I BCE-I CE	Fish tank, upper jetty	Fine	?*	PF-1**
SAR-4	Torre Astura-Saracca			Fish tank, lower jetty	Fine	CA-PR	No fresh glass
BAN-5	Torre Astura-Banca	41.4172 N, 12.7494 E	I BCE-I CE	Fish tank, crepido	Fine	PF-3	PF-3
TER[(1)]	Port of Anxur		II CE	Harbor dock	Fine		PF-3; PF-1
TER-G[(1)]	Port of Anxur			Harbor dock	Fine	PF-3	
TER-W[(1)]	Port of Anxur			Harbor dock	Fine	PF-1	
TER-TU	Port of Anxur			Harbor dock	Coarse	PF-NYT	
ISC-6	Ischia	40.7296 N, 13.9604 E	I BCE-I CE	Harbor dock	Fine	PF-3	PF-3
PFU-12	Porto Fuenti	40.6577 N, 14.7130 E	I BCE	Harbor dock	Fine	PF-3	PF-3
PFU-12b	Porto Fuenti			Harbor dock	Coarse	PF-NYT	

a large fishpond around it, and served by a port of considerable size, built in later time. The quadrangular fishpond, one of the largest today preserved, is 172 × 125 m and is set directly on the bedrock. It is bounded, towards the open sea, by a perimeter pier in *opus caementicium* in which five openings granted water circulation. All around there is a series of small rectangular rooms connected together. The terraced artificial island was built on concrete vaults faced in *opus reticulatum*. Ancient sources and construction techniques refer to an early phase dating to the second half of the first century BCE. Restorations in *opus vittatum* refer to a later phase ascribable to the later Empire. A medieval Torre, finally, was built on the ancient ruins providing the modern name of the locality (Higginbotham, 1997).

A mortar (TAP-10) and a tuff *caementa* sample (TAP-10b) have been collected from the *crepido* of the fish tank (Fig. 2d) and analyzed at the optical microscope and at the EMP. A selected fraction of the fine aggregate separated after HCl bathing from sample TAP-10 and a tuff fragment of sample TAP-10b have been analyzed for trace element composition.

2.1.5. La Saracca

The fish tank La Saracca (Fig. 2e) has a semicircular plan and is protected by a thick dam. It consists of three concentric rows of basins, now mostly submerged and partially buried by sand. On the western flank of the fishpond, a long concrete channel with posts for sluice gates connected it to the sea (Higginbotham, 1997; Piccarreta, 1977). The fishpond and the nearby Villa are dated to the late Republican period, but they both have restorations in *opus vittatum* showing they were still in use in the fourth century CE.

Two samples (SAR-3, SAR-4) have been collected in the concrete of the upper and of the lower jetty of the fishpond, respectively (Fig. 2e).

Thin sections of the untreated samples have been performed for petrographic analysis at the optical microscope and EMP interstitial glass of pumice clast analysis; however, only SAR-3 contained unaltered glass. A selected fraction of the fine aggregate separated after HCl bathing from SAR-4 and SAR-3 has been analyzed for trace element composition.

2.1.6. La Banca

The site called La Banca (Fig. 2df) hosts the remains of a seaside villa with a fishpond. It is located 1,6 km northwest of Torre Astura and dates between the end of the first century BCE and the beginning of the first century CE (Higginbotham, 1997). Walls in *opus reticulatum* of the fishpond refer to the first phase, while brick restorations may be attributed to the later phase. Villa and fishponds also shows restorations in *opus vittatum*, that refer to the fourth century CE (Rustico, 1999; Tol, 2012; Zarattini et al., 2010).

One sample (BAN-5) has been collected from the inner side of the outer walls surrounding the fishpond (Fig. 2f) and analyzed at the optical microscope and EMP. A selected fraction of the fine aggregate separated after HCl bathing has been analyzed for trace element composition.

2.1.7. Terracina

The port of *Anxur* (the modern Terracina, Fig. 1a) was built by Trajan at the beginning of the second century CE (Di Mario, 1994). One sample of tuff *caementa* (TER-TU) was collected from the inclined breakwater of the quay of the port and analyzed for trace element. This is a concrete masonry structure that in its upper portion is entirely made up of blocks of a yellow tuff, laid in rather regular rows, whereas more resistant blocks of carbonatic rock (which extensively crops out in Terracina) substitute the tuff in the lower portion of the quay.

2.1.8. Ischia

Ongoing excavations have brought into light the remains of the ancient harbor of Aenaria (the Roman name of Ischia island). A ~20 m long pier in opus caementicium has been dated between the end of the first century BCE and the beginning of the first century CE based on recovered ceramic material (unpublished data). One sample (ISC-6) has been collected from the concrete of the submerged harbor dock (Fig. 2g) and analyzed at the optical microscope and EMP. A selected fraction of the fine aggregate has been analyzed for trace element.

2.1.9. Punta Fuenti

The submerged structure located ca.1 m below the sea level is very likely a harbor pier. It is an *opus caementicium* wall with an *opus reticolatum* facing and it presents holes on the summit of the jetty which were likely deputed to host wooden poles. Employment of *opus reticolatum* technique suggests a date within the second half of the first century BCE for the structure (Benini, 2006).

A mortar sample (PFU-12) has been collected in the concrete of the harbor pier (Fig. 2h) and analyzed at the optical microscope and EMP. A selected fraction of the fine aggregate separated after HCl bathing from sample PFU-12 and a tuff *caementa* sample (PFU-12b) have been analyzed for trace element composition.

2.2. Trace element analysis

Bulk samples of the volcanic component, either single scoria or pumice clast, or, when the grain size of the aggregate was too small, several clasts selected for their homogeneous texture and mineralogy, were separated from the lime matrix through HCl bathing and analyzed for trace element at the Activation Laboratories, Canada by Lithium Metaborate/Tetraborate Fusion ICP-MS. Fused sample is diluted and analyzed by Perkin Elmer Sciex ELAN 6000, 6100 or 9000 ICP/MS. Three blanks and five controls (three before sample group and two after) are analyzed per group of samples. Wet chemical techniques were used to measure the loss on ignition (LOI) after heating at 900 °C. International rock standards have been used for calibration and the precision is better than 5% for Rb and Sr, 10% for Ni, Zr, Nb, Ba, Ce, and La, and 15% for the other elements.

Full data are reported in Online Appendix 1. Interpretation of the data has been carried out by using Zr/TiO_2 vs Nb/TiO_2 and Th/Ta vs Nb/Zr discrimination diagrams (selected element data in Table 1), following the procedure described in Marra et al. (2015) and D'Ambrosio et al. (2015).

2.3. EMP analyses of matrix glass

Electron microprobe (EMP) analyses were performed on polished thin sections at Istituto Nazionale di Geofisica e Vulcanologia (Rome, Italy) with a JEOL JXA 8200 equipped with five wavelength-dispersive spectrometers, using 15 kV accelerating voltage, 4.27 nA beam current: counting time of 10 and 5 s were used on background and peaks, respectively. To reduce alkali loss, the spot size was enlarged to 5 μm and Na and K were counted first and simultaneously with Al, Si and Ca. The following standards were adopted for the various chemical elements: Anorthoclase (Al, Si, Na), Augite (Mg), fayalite (Fe), rutile (Ti), Kfs (K), barite (Ba), apatite (P), rhodonite (Mn).

Full data are reported in Online Appendix.

3. Samples analyzed in previous studies

Based on the geochemical data reported in Jackson et al. (2010), Marra and D'Ambrosio (2013) have shown that the aggregate used in the second century CE Trajanic port of Ostiae yielded a Zr/Y vs Nb/Y signature attesting its provenance from Phlegrean Fields (Fig. 3a), whereas that of a ca. 60 BCE structure of the Cosa harbor (Tuscany) yielded a Campanian composition, which has an only equivalent in the products of the 79 CE eruption of Vesuvius (Fig. 3a). This latter event occurred well after the supposed construction of the portion of the harbor from which the pumice was sampled, thus leaving its actual identification uncertain.

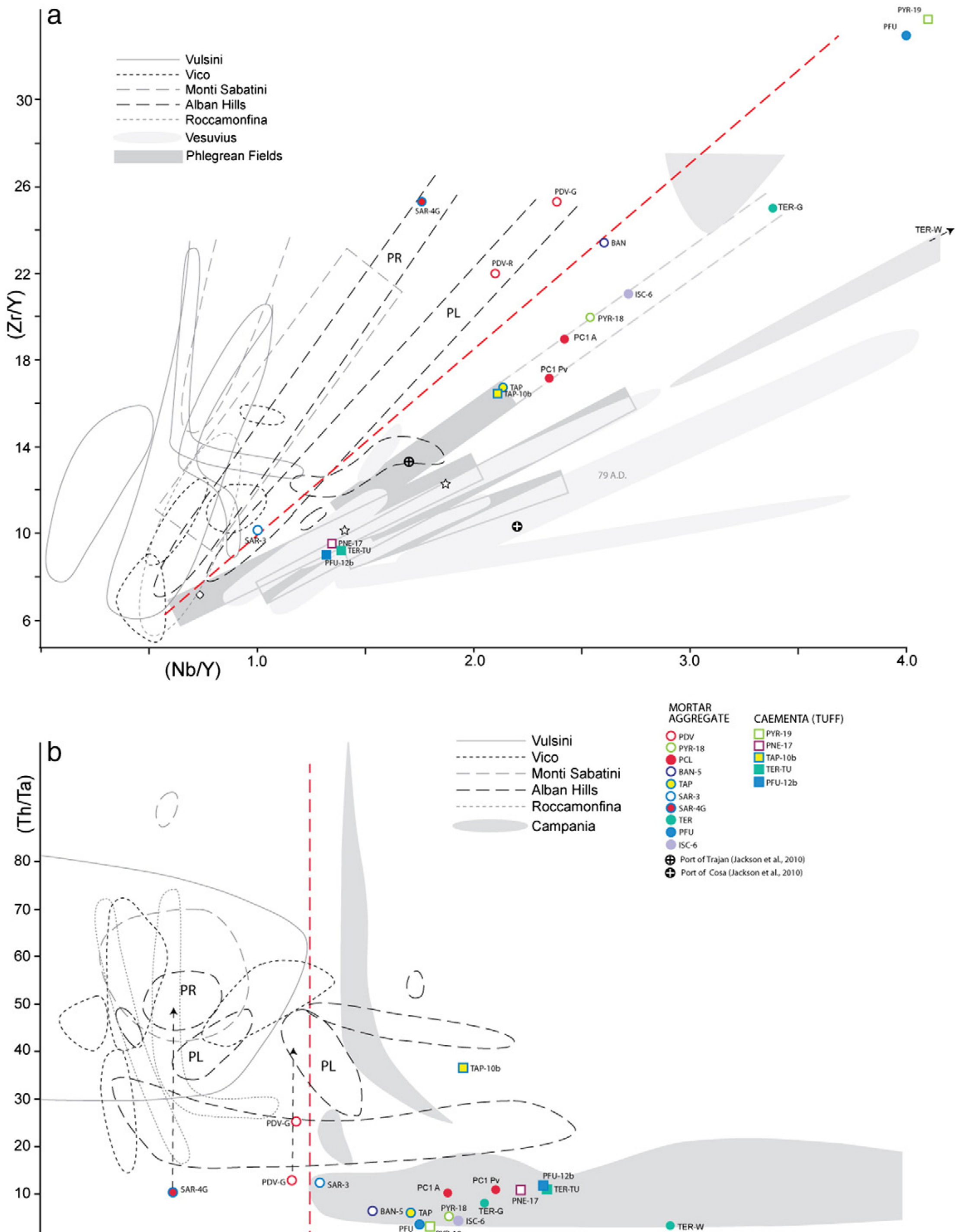

Fig. 3. a) Zr/Y vs Nb/Y and (b) Zr/TiO_2 vs Nb/TiO_2 composition of the mortar aggregates and tuff caementa analyzed in this work (except when indicated) are plotted against the compositional fields of the different volcanic products determined by literature data (Marra and D'Ambrosio, 2013; D'Ambrosio et al., 2015, Marra et al., 2015, and references therein). The dashed red lines separate the compositional field of Latium (above/left) from that of Campania (below/right). See text for comments.

Jackson et al. (2013) analyzed four pumice clasts and one tuff fragment collected from the concretes of the Roman Port of Baiae in the Gulf of Pozzuoli, along with a sample of the Bacoli Tuff, which is a semi-lithified pyroclastic deposit erupted by the homonymous local volcanic center. Zr/Y vs. Nb/Y composition of the pumices matched that of the products of most recent activity of Phlegrean Fields, while the tuff coarse aggregates and the Bacoli Tuff yielded quite similar compositions, plotting within the Neapolitan Yellow Tuff (NYT) compositional field. We have computed Zr/TiO_2 and Nb/TiO_2 for these samples from the data by Jackson et al. (2013), and plotted the values in Fig. 4 for comparison with the samples analyzed in the present work.

Finally, D'Ambrosio et al. (2015) investigated the composition of the mortar aggregates from the port of *Claudius* in Fiumicino, started by Emperor Claudius in 42 CE and inaugurated by Emperor Trajan in 64 CE, and the port of *Anxur* (the modern Terracina), which was built by Trajan at the beginning of the second century CE (Di Mario, 1994).

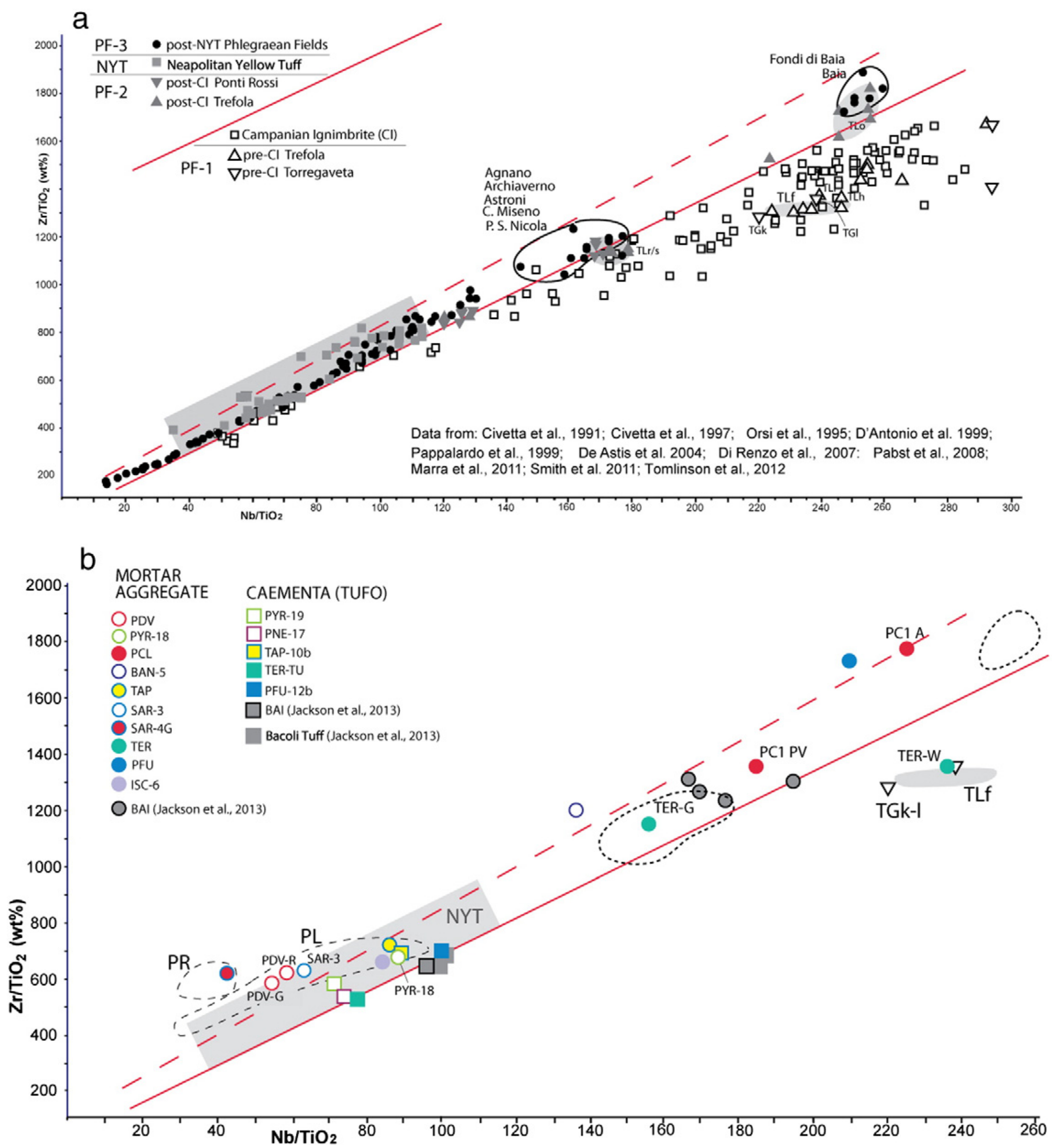

Fig. 4. a) Zr/TiO_2 vs Nb/TiO_2 compositional fields of the different volcanic products of the Phlegrean Fields activity from literature data (Civetta et al., 1991; Civetta et al., 1997; Orsi et al., 1995; D'Antonio et al., 1999; Pappalardo et al., 1999; De Astis et al., 2004; Di Renzo et al., 2007; Pabst et al., 2008; Marra et al., 2011; Smith et al., 2011; Tomlinson et al., 2012). The solid red line separates the compositions of the post Campanian Ignimbrite phase of activity (above) with respect to that of the CI and previous activity (below). The dashed red line indicates the upper compositional limit of the products of the latest eruptive phase (PF-3). b) Mortar aggregate and tuff caementa compositions analyzed in this work (except when indicated) are plotted in the discrimination Zr/TiO_2 vs Nb/TiO_2 diagram for the products of the Phlegrean Fields. See text for comments.

The mortar sample collected at Port of Claudius contained different types of pumice, displaying different textural features, but homogeneous geochemical composition. The combined Zr/TiO_2 vs Nb/TiO_2 and TAS compositions of two analyzed samples (PC1-A, PC1-PV, Table 1) evidenced their correlation with the deposits of the post-NYT activity of Phlegraean Fields (PF-3, in Fig. 4).

At Terracina, two types of pumice separated from the fine aggregate (TER-G, TER—W, Table 1) yielded different compositions. The combined Zr/TiO_2 vs Nb/TiO_2 and TAS compositions of the matrix glass of pumices suggest that the fallout deposit of Trefola (TLf, Pappalardo et al., 1999; Pabst et al., 2008) represent the product of origin for TER-W (Figs. 4 and 5). In contrast, pumice TER-G displays Zr/TiO_2 vs Nb/TiO_2 and TAS composition matching that of the products of the late Phlegraean Fields activity (Figs. 4 and 5).

4. Results

4.1. Petrographic observations in thin section

The aggregates of eight investigated mortar samples (see Table 1) are composed almost entirely of volcanic material including pumices and less vesicular scorias along with an abundant fraction of loose minerals, holocrystalline lava fragments, and a minor fraction of sedimentary lithics. The mineral assemblages, either within the juvenile fraction as well as in the loose fraction, are characterized by sanidine, plagioclase, pyroxene, biotite, olivine, iron and titan oxides. This assemblage is neither diagnostic of a specific volcanic district, nor of a volcanic region (e.g.: Marra et al., 2013; D'Ambrosio et al., 2015). The pumiceous component is characterized by aphiric to sub-aphyric texture with rare

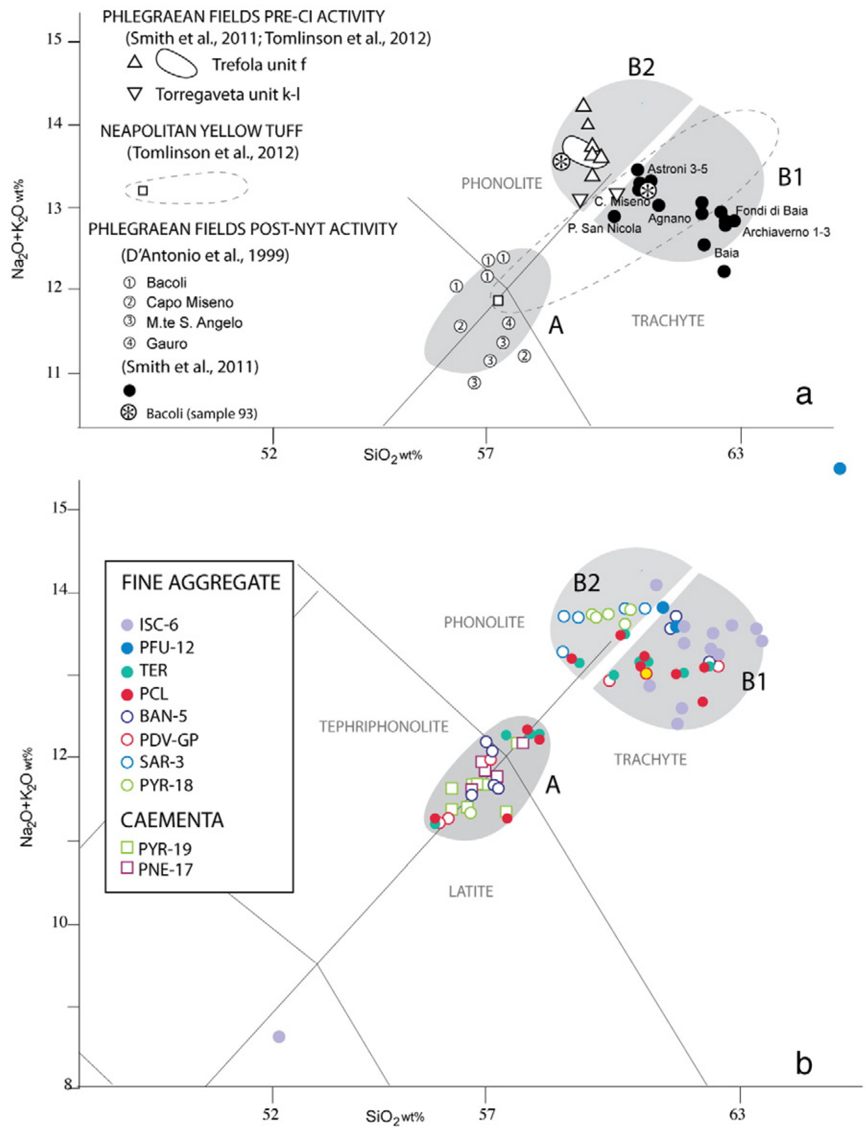

Fig. 5. a) SEM TAS compositions of matrix glass for the products of the Phlegrean Fields from literature data. b) SEM TAS composition of matrix glass for the mortar aggregate and tuff caementa analyzed in this work. See text for comments.

sanidine and/or plagioclase, and by clear, poorly to moderately vesicular glass. These characteristics are common to the Vulsini, Vico, Monti Sabatini, Phlegraean Fields and Vesuvis volcanic districts, so preventing discrimination. The scoriaceous component is present in a lesser amount with respect to the pumiceous fraction, and it is mostly represented by poorly vesicular, aphiric grey scoriae with sporadic sanidine or pyroxene. Only two mortar samples (PDV-21 and SAR-4) contain leucite-bearing reddish scoriae suggesting a provenance from the Colli Albani district, mixed with pumices. Indeed, trace-element composition of these samples excludes provenance from Campania (Fig. 3).

Two samples of coarse aggregate observed in thin section at the optical microscope (PYR-19, PNE) revealed the presence of pumice clasts, mostly aphiric or with rare plagioclase, characterized by mixing of brownish and colorless glass, a feature occurring in the Neapolitan Yellow Tuff (Orsi et al., 1995).

A summary of the textural and mineralogical features of the volcanic fraction contained in the thin sections is provided in Table 2.

4.2. Trace-element discrimination diagrams

Fig. 3 a and b shows the Zr/Y vs. Nb/Y and the Th/Ta vs. Nb/Zr discrimination diagrams (Marra et al., 2011; Marra and D'Ambrosio, 2013). These plots compare the compositions of the analyzed aggregate samples with the compositional fields of the different volcanic districts of the Roman Province (Peccerillo, 2005; Lustrino et al., 2011, and references therein). With the exception of three (PDV-G, PDV-R, SAR-4), all the investigated samples plot to the right of the red dashed line in both diagrams, within the compositional fields defined by the products of the Campania Province (including Phlegraean Fields, Vesuvius, Ischia and Procida). The two scoria samples collected from the concrete of the fishpond at Punta della Vipera (PDV-G and PDV-R) are aligned along the compositional trend of Pozzolanelle (PL, Alban Hills), displaying Zr/Nb consistent with attribution to this volcanic unit, whereas the offset from the origin of the axes is interpretable as due to Y depletion caused by the HCl treatment (Marra et al., 2015). Similarly, sample SAR-4 falls within the enlarged compositional field of the Pozzolane Rosse (PR, Alban Hills). When also the offset position of these samples in the Th/Ta vs Nb/Zr diagram is considered, suggesting a loss in Ta with respect to the original PR and PL composition (dashed arrows in Fig. 3b), the effect of leaching due to the acid treatment to separate the aggregate from the matrix is supported further.

Moreover, consistently with employment of both PR and Campanian pumices in the concrete of the upper jetty (SAR-4), mortar sample SAR-3 collected in the upper jetty at La Saracca displays an intermediate

Table 2
Petrographic features of the concrete aggregates at the optical microscope.

Label	Pumice		Scoriae		Loose fraction	
PDV 21	Aphiric, rare plagioclase, clear vesicular glass	+++	Dark grey and red with rare leucite and very rare sanidine	++	Pyroxene, mica, oxides, olivine, sedimentary lithics	++
PYR 18	Aphiric, rare plagioclase, brown glass	+++	Dark grey aphiric	+	Pyroxene, mica, oxides, lava and sedimentary lithics	++
PYR 19	Aphiric, rare plagioclase, mixed glass	+++	–		Pyroxene, lava lithics	+
PNE-17	Aphiric, rare pyroxene and oxides, mixed glass	+++	Dark grey aphiric	+	Pyroxene, mica, oxides, plagioclase	+
TAP-10	Aphiric, rare sanidine and plagioclase, elongated vesicules, brownish glass	+++			Pyroxene, plagioclase, oxides, lava and carbonatic lithics	++
SAR-3	Aphiric, rare plagioclase, clear vesicular glass	+++	Dark grey with rare pyroxene	+	Pyroxene, white mica, olivine, sandstone, carbonatic lithics	+
SAR-4	Aphiric, clear poorly vesicular glass	+	Dark grey and red with starry leucite	+++	Pyroxene, olivine, plagioclase, sanidine, oxides, mica, sandstone, carbonatic and lava lithics	+++
BAN-5	Aphiric, rare sanidine, plagioclase, pyroxene, oxides	+++	–		Pyroxene, mica, oxides, plagioclase, carbonatic lithics	++
ISC-6	Aphiric, rare plagioclase, biotite, pyroxene, oxides	+++	Dark grey with rare plagioclase	+	–	
PFU-12	Aphiric	+++	Dark grey aphiric	+	Pyroxene, mica, oxides, plagioclase	++

composition, which is transitional between PR and the Campania fields (Fig. 3), suggesting that the analyzed sample contained a mixture of these products.

In order to better constrain the provenance of the aggregate from specific volcanic products we will rely on the Zr/TiO_2 vs Nb/TiO_2 discrimination diagram (Fig. 4) which, as shown by Marra et al. (2015), is less affected by the alteration processes. The high reliability of this diagram is evidenced by Zr/TiO_2 vs Nb/TiO_2 composition of the three Alban Hills samples, which plot inside the boundaries of the respective fields for the PR and PL (dashed lines in Fig. 4b). Similarly, all the other mortar samples are more strictly constrained within or close to the boundaries of the products of the late Phlegraean Fields activity (PF-3 within the lower solid and upper dashed red lines in Fig. 4a). SAR-3 displays a slightly offset position with respect to this field, accounting for the presence of a small fraction of PR clasts within the aggregate that has been analyzed. Finally, all the tuff samples from coarse aggregates are tightly clustered to each other within the compositional field of the Neapolitan Yellow Tuff (NYT, grey box in Fig. 4b). Tuff samples analyzed by Jackson et al. (2013) also plot close to those analyzed in the present work, whereas their pumice samples have composition quite similar to those of the products of the volcanic centers around Pozzuoli (Agnano, Astroni, Archiaverno, Capo Miseno, Pigna San Nicola, thick dashed contour line in Fig. 4b). These latter have been identified by D'Ambrosio et al. (2015) as the more probable sources for the aggregates of the Ports of Claudius and Anxur (samples PC1-PV and TER-G, Fig. 4b).

Only one sample from the port of Terracina (TER-W, Fig. 4b) analyzed in D'Ambrosio et al. (2015) displays a distinct composition within the pre-NYT field (PF-1 in Fig. 4a). According to its bimodal TAS composition (see Fig. 5b), D'Ambrosio et al. (2015) attributed a fraction of the pumices employed in the concrete of the Port of Terracina to the pre-NYT products cropping out in Trefola or in Torregaveta (TLf, TGk-l in Figs. 4a, b, and 5a; Smith et al., 2011; Tomlinson et al., 2012).

4.3. Matrix glass TAS composition

Matrix glass composition of eight aggregates of maritime concrete samples investigated for this work and two samples previously analyzed in D'Ambrosio et al. (2015) are plotted in the TAS diagram of Fig. 5b. An overall bimodal composition (groups A and B) characterizes the distribution of the newly analyzed samples. Moreover, a further distinction (sub-groups *B*1 and B2) is made after comparison with TAS composition of natural products (Fig. 5a). Indeed, all the most differentiated products of the post-NYT activity at Phlegraean Fields (black dots in Fig. 5a) have trachytic composition and plot below the boundary line of the phonolitic compositional field (B1 sub-group), whereas the phonolitic composition (B2 sub-group) is exclusive of the pre-NYT products of Torregaveta and Trefola (white triangles), with the only exception of one sample of the eruptive activity of Bacoli (Smith et al., 2011). Two analyses on sample TER plotting within field B2, coupled to the peculiar Zr/TiO_2 vs. Nb/TiO_2 composition of a selected pumice from the concrete of the Port of Terracina (TER-W, Fig. 3b), allowed D'Ambrosio et al. (2015) to hypothesize the employment of the products from Trefola or Torregaveta in the mixture of volcanic ash to realize the aggregate of this concrete. Remarkably, two samples analyzed in this work (SAR-3, PYR-18) display a homogeneous composition within field B2; however, their trace-element signature is not consistent with an attribution to the pre-NYT eruptive products: indeed, both samples plot above the upper limit of the PF-1 compositional field (solid red line in Fig. 4b). It is likely, in fact, that these aggregates were realized with the most differentiated products of the activity of Bacoli, as suggested by the bimodal composition of one sample analyzed in Smith et al. (2011) (asterisk in Fig. 5a).

5. Discussion and conclusions

Geochemical data presented in this study show that the source area of the volcanic products employed in all the investigated third BCE to second CE old maritime structures of the central Tyrrhenian Sea of Latium through northern Campania is the Phlegrean Fields volcanic district. The trace-element and matrix glass compositions of the aggregates extracted from the concretes are tightly clustered within the corresponding compositional fields of the products late erupted by volcanic centers of Phlegrean Fields (PF-3). These vents are located in the immediate inland area of the roman ports of Baiae and Puteoli (modern Baia and Pozzuoli, (Fig. 1b). Only in two cases, a partial mixture with products of the Colli Albani volcanic district has been evidenced.

The products of the Vesuvius activity, which were extensively employed for land building in Pompeii since the third century BCE (Miriello et al., 2010) and exported in Rome to be employed as lightweight aggregates for vaults since the first century BCE (Marra et al., 2013) are lacking in the concretes of all the investigated maritime structure. The only possible exception is suggested by the Zr/Y vs Nb/Y composition of one mortar sample from the harbor of Cosa analyzed by Jackson et al. (2010); the composition of this sample matches that of the products of the 79 CE eruption of Vesuvius (Marra and D'Ambrosio, 2013). However, the conflicting age attributed to the sampled structure (third century BCE) questions the reliability of the geochemical correlation.

The peculiar phonolitic glass composition (sub-group B2 in Fig. 5b) of several pumices as those employed in the concretes of Pyrgi and La Saracca and previously found (D'Ambrosio et al., 2015) at the ports of Claudius and Anxur (Fig. 5b), coupled with a Phlegrean Fields Zr/TiO_2

vs. Nb/TiO_2 composition (Fig. 4b), has the only counterpart reported in the literature for the products of Bacoli (Smith et al., 2011; Fig. 5a). These latter products are the only ones among those of the PF-3 phase that are characterized by a wide range of compositions, matching all the three TAS compositional fields in which the analyzed mortar aggregates plot (A, B1, and B2, Fig. 5a). Therefore, the volcanic center of Bacoli may be considered as the source area of all the mortar aggregates analyzed in the present and previous work, with the exception of that occurring at the second century CE port of *Anxur* (sample TER-W), for which a provenance from the pre-Campanian Ignimbrite phase of activity (PF-1), either from Trefola or Torregaveta (TL-f and TGk, TG-l units, Pappalardo et al., 1999; Pabst et al., 2008) has been demonstrated based on coupled B2 TAS and PF-1 Zr/TiO_2 vs. Nb/TiO_2 composition (D'Ambrosio et al., 2015). In contrast, based on the trace-element composition excluding a pre-CI composition, the aggregates of samples PYR-18 and SAR-3 have different source area (Fig. 4b), compatible with the phonolitic B2 composition of the Bacoli products (Fig. 5a).

These observations suggest that the original area of production of the materials employed as fine and coarse aggregate for sea-water concretes was that strictly in the surrounding of Pozzuoli, corresponding to the outcropping area of the products of the PF-3 phase of activity erupted by the center of Bacoli, Baia, Fondi di Baia, Astroni, Archiaverno, Agnano, Capo Miseno, Pigna San Nicola (Fig. 1), and only later on, probably since the first century CE, based on the presence of pumices similar to those analyzed at Terracina also in the concrete of the Port of Claudius (D'Ambrosio et al., 2015), the exploitation was enlarged to more inland quarries, like that in Trefola.

Particularly interesting is the occurrence of Pozzolanelle in the aggregate of the first century BCE port of Punta della Vipera. The use of Pozzolanelle in Rome is reported only in the earliest monuments in *opus caementicium*, like the Porticus Aemilia and, the Temple of Concordia (Marra et al., 2015), while since the end of the second century BCE the extensive, almost exclusive use of Pozzolane Rosse, cultivated in dedicated tunnels (*arenarii*), is documented (Marra et al., 2015). Later on, the enlargement of the exploitation areas outside the City since the end of the second century CE was the probable cause of a renewed employment of Pozzolanelle, which were easily exploited at open-air quarries (Marra et al., 2015). The use of Pozzolanelle in maritime structures of the first century BCE in northern Latium suggests a different commercial route for the supply of building materials, with respect to the consolidated exploitation system for the urban construction in Rome. This indicates a different technical and commercial framework for sea-water with respect to the terrestrial engineering.

It is inferred by the historical sources (Vitruvius, Pliny the Elder, Seneca) that the Roman builders and architects like Vitruvius (*De Arch.2.6.1*) were convinced that only the pozzolan occurring in the surrounding of Pozzuoli, unlike the same volcanic material extensively exploited in Rome and in Etruria, was fit for seawater structures. This is much more evident by considering that Vitruvius uses different terms (*pulvis* vs. *harena*) for these volcanic aggregates. However, along with the evidence from Punta della Vipera, the concrete of Torre Astura shows that the use of *harena fossicia* in maritime structures was not a priori discarded. Indeed, the fine aggregate of the lower portion of the wall of the first century BCE fishpond in La Saracca was realized with Pozzolane Rosse imported from Rome, whereas the pozzolan from Pozzuoli was used in the upper portion of the same wall challenging the notion that the Romans considered the pozzolan from Latium unfit for seawater construction. The occurrence of a number of mainland structures in Torre Astura, including the first century BCE Cicero's Villa and its annexes, attesting for intensive use of building material from Rome, was likely the cause of availability of Pozzolane Rosse, which was integrated with the pozzolan imported from Pozzuoli in the concrete of the fishpond.

The excellent outcome of the Latial volcanic products employed in the fishponds of Torre Astura and Punta della Vipera is evidenced by the fact that these structures are perfectly preserved nowadays, just like the other maritime structures built using the products exploited at the Phlegraean Fields. It clearly shows that other volcanic products, which are quite similar from the geochemical point of view to those from Campania, along with which constitute the well-known Roman Magmatic Province (Peccerillo, 2005), would have worked as well in maritime construction. Therefore, in our opinion, the almost ubiquitary (but not exclusive) employment of pozzolan exploited in the Phlegraean Fields to produce hydraulic mortars for the Roman ports of the Tyrrhenian Sea, rather than to unattested distinctive mechanical and physical properties of these volcanic products with respect to those of the other volcanic districts of central Italy (see D'Ambrosio et al., 2015, for a discussion), is to be attributed to the large occurrence in the vicinity of one of the earliest and most important Roman harbors of Mediterranean Sea. In 194 BCE, Romans established the maritime colony of *Puteoli* (Pozzuoli) and created a new harbor system conveying to Pozzuoli the traffic of the goods coming in particular from the eastern Mediterranean Sea. In the Mediterranean Sea, *Puteoli* harbor was a hub for the commercial and raw material traffic. This fact is likely at the base of the development of the constructive technique of maritime structures and the selection of the materials, which were attested by a certification of resistance and duration. Strabo, among others (*Geography*, 5.4.6) reports that Pozzuoli "*had become a great trade center, since it has manmade harbors thanks to the natural quality of the sand. (…) by mixing the sand-ash they can run breakwaters out into the sea (…)*".

However, it seems unlikely that the Roman engineers, who demonstrated a deep empirical knowledge of the geotechnical properties of the material employed in civil construction (e.g., Lancaster, 2005; Jackson et al., 2009; Marra et al., 2013; D'Ambrosio et al., 2015) were not aware that any other kind of *harena fossicia* would have worked as well as the *pulvis puteolanis* in seawater structures. Therefore, the extensive use of the volcanic materials exploited in Pozzuoli should be considered to descend from logistic and economic reasons and not from peculiar technical properties. First of all, the possibility to exploit it in the immediate vicinity of the most important commercial port of the Roman era, which allowed for a port-to-port transportation, eliminating any other intermediate transport from inland quarries; in second instance, a well attested know-how that very likely allowed to export local manpower along with the building materials, consolidating a monopoly for Pozzuoli in the field of maritime construction.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.jvolgeores.2016.10.005.

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