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Provenance and sediment dispersal in the Po-Adriatic source-to-sink system unraveled by bulk-sediment geochemistry and its linkage to catchment geology

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ABSTRACT

The Po-Adriatic region offers an excellent case for reconstructing sediment provenance and transport pathways of a multi-sourced sediment-routing system. Through a comprehensive set of ~1400 geochemical data, a model for provenance and sediment flux was built based on distinct compositional fingerprints of 53 fluvial systems and their comparison to coastal, shelf and deep-marine sediments. Geochemically unique catchment lithologies (mafic/ultramafic rocks, limestones and dolostones) were used as end-members to assess exclusive source-rock signatures. Following calibration with sedimentary facies, selected key elements and element ratios poorly sensitive to particle size (Ni/Cr, MgO, Ni/Al_2O_3, Cr/V, Ca/Al_2O_3 and Ce/V) were adopted as provenance indicators. The high-Ni and high-Cr source-rock signature of mafic/ultramafic rocks widely exposed in the Po River watershed and along the Albanian Dinarides contrasts markedly with the high-Ca (and locally high-Mg) geochemical composition of Eastern Alpine, Apennine, and Eastern Adriatic (Montenegro, Croatia, Slovenia) river catchments, which are, instead, carbonate-rich and virtually ophiolite-free. Relatively high Ce values from Apulian river samples serve as a key marker for a minor, but very distinct sediment provenance from southern Apennine alkaline volcanic rocks.

Despite along-shore mixing and dilution with sediment sourced from other river catchments, the geochemical signature of Adriatic shelf muds primarily reflects composition of sediment eroded from the contiguous continental areas. Chromium-rich and nickel-rich detritus generated in mafic and ultramafic complexes of the Western Alps and conveyed through the Po River into the Adriatic Sea records a geochemical signal that can be traced downstream as long as 1000 km, from the Alpine zone of sediment production to the area of final deposition, offshore Apulia.

While longitudinal dispersion linked to the general cyclonic, counter-clockwise Adriatic circulation is prevailing along the Western Adriatic Sea, conspicuous detrital input from transversal pathways to the deep sea is revealed across the Eastern Adriatic shelf using heavy metals as provenance tracers. Estimates of fluvial sediment loads and compositional fingerprinting of fluvial, coastal and shelf sediments indicate that previously neglected ophiolite-rich successions of Albania represent a major sediment-conveyor to the offshore sinks (Southern Adriatic Deep and Mid-Adriatic Deep) through significant cross-shore and NNW-directed sediment transport in the Eastern Adriatic Sea. A cut-off value of the Ni/Cr ratio targeted around 0.8 represents an effective tool for the differentiation in marine sediments of Ni-rich (serpentine-rich) ophiolite detritus of Albanian origin from mafic/ultramafic sources of Alpine affinity. High trace-metal contents found within the Adriatic deep basin are mostly of natural origin and only minimally reflect metal contamination.

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1. Introduction

Reliable quantitative estimates of sediment transport and storage from a multi-sourced system, along the pathway from river catchments to the ultimate sink, require accurate identification of the sediment sources and of their role in modulating sediment supply to the downdip components of the dispersal system. Marginal seas and other semi-enclosed basins, in particular, represent a critical component of the global source-to-sink system and are ideal sites to investigate land-sea interactions, because in these areas the sediment budget and transport pathways can be more easily defined quantitatively (Liu et al., 2016; Zhang et al., 2021). In such depositional settings sediment largely consists of clay, which makes conventional petrographic studies inapplicable. Conversely, though lacking precise mineralogical connotation, bulk-sediment geochemistry represents an efficient technique that does not need time-consuming sample preparation and analysis, and that can be applied to the entire grain size distribution, including silts and clays carried in suspension (e.g. Fralick and Kronberg, 1997; Zhang et al., 2014; Garzanti, 2016).

In the coastal regions of East- and SE-Asia, where rivers provide the largest portion of global sediment discharge to the world oceans (Milliman and Meade, 1983; Milliman and Farnsworth, 2011), bulk-sediment geochemistry has been widely used for sediment provenance discrimination and for the distinction between lithogenic and anthropogenic sources. In these regions, major and trace element geochemistry of modern and Holocene deposits from the Bohai Bay (Zhang et al., 2002; Xu et al., 2016), the Yellow Sea (Yang et al., 2003; Lim et al., 2013, 2015; Xu et al., 2016; Zhu et al., 2021), the East China Sea (Xu et al., 2011; Bi et al., 2017; Zhang et al., 2021), and the South China Sea (Liu et al., 2016; Liu et al., 2018), combined with geochemical fingerprinting of modern river sediments (He et al., 2015; Nguyen et al., 2016; Sun et al., 2018), deltas (Qi et al., 2010; Strady et al., 2017), and coastal plains (Yang et al., 2002; Rao et al., 2017) has allowed to assess source-to-sink dispersal patterns from several of the world's largest rivers, including the Changjiang (Yangtze) River the Huanghe (Yellow) River, the Pearl River, the Red River, and the Mekong River.

Major- and trace-element compositions of surficial sediments and their natural levels in relation to the source rocks have also been investigated in several other semi-enclosed basins or large bays worldwide, including the Black Sea (Kiratli and Ergin, 1996), the Baltic Sea (Shahabi-Ghahfarokhi et al., 2021), the Baffin Bay (Loring, 1984), the Gulf of Mexico (Armstrong-Altrin et al., 2015), the Kara Sea (Loring et al., 1998), the Bengal Bay (Tripathy et al., 2014; Li et al., 2017; Sun et al., 2019), and the Japan Sea (Ohta et al., 2004; Cha et al., 2007).

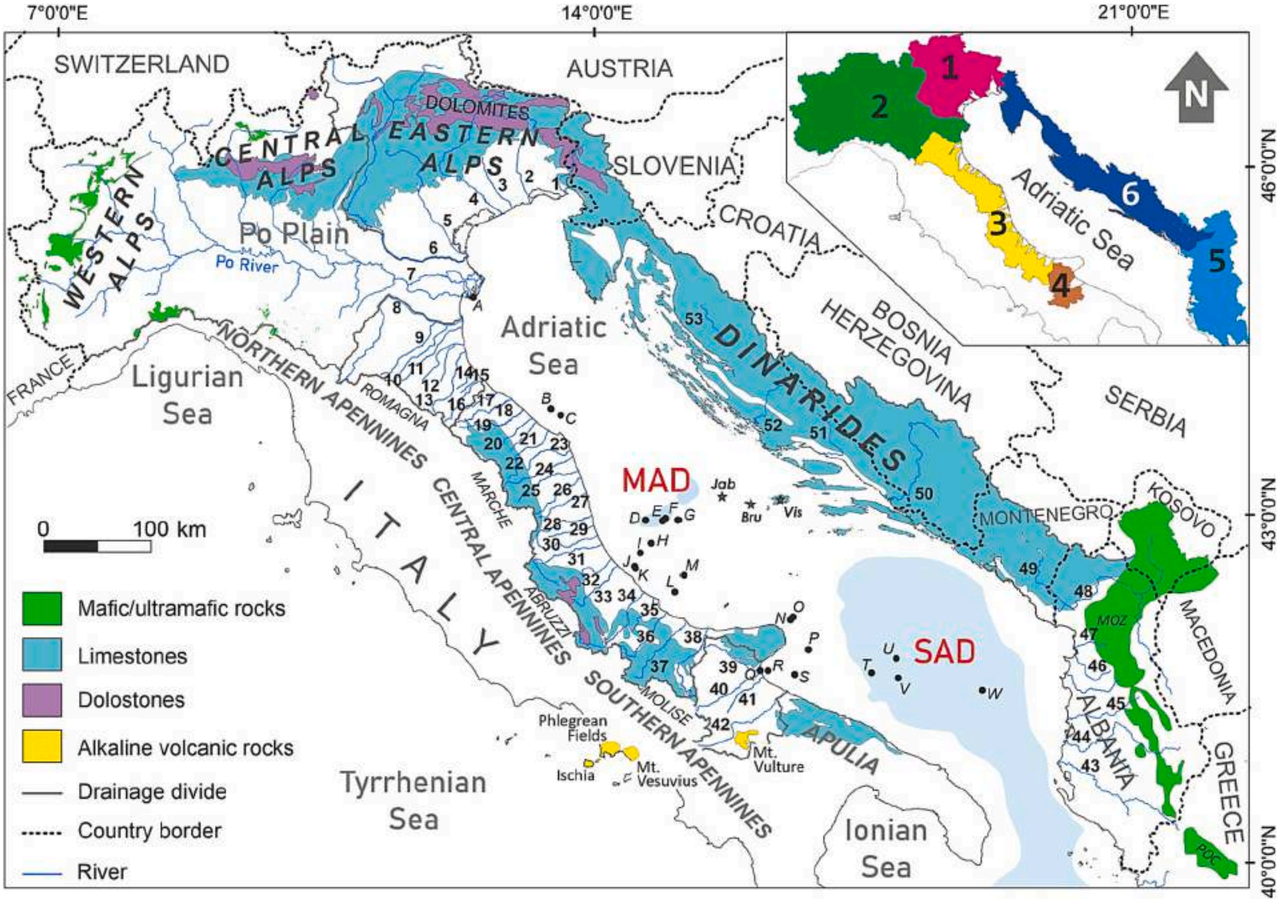

Fig. 1. Geological sketch map of the Adriatic sediment routing system, with indication of Alpine, Apennine and Dinaric rock units with high potential to provide geochemical tracers, and six compositional end-members (1: Eastern Alps, 2: Po River catchment, 3: Apennines, 4: Apulia, 5: Albania, 6: Dinarides: Montenegro-Croatia-Slovenia). River catchments (1–53) and cores (A-W) used in this study are also indicated. 1: Isonzo, 2: Tagliamento, 3: Livenza, 4: Piave, 5: Brenta, 6: Adige-Isarco, 7: Po, 8: Reno, 9: Santerno, 10: Senio, 11: Lamone, 12: Fiumi Uniti, 13: Savio, 14: Rubicone, 15: Uso, 16: Marecchia, 17: Conca, 18: Foglia, 19: Metauro, 20: Cesano, 21: Misa, 22: Esino, 23: Musone, 24: Potenza, 25: Chienti, 26: Tenna, 27: Aso, 28: Tronto, 29: Vibrata, 30: Salinello, 31: Tordino, 32: Vomano, 33: Aterno-Pescara, 34: Sangro, 35: Sinello, 36: Trigno, 37: Biferno, 38: Fortore, 39: Candelaro, 40: Cervaro, 41: Carapelle, 42: Ofanto, 43: Vjosa, 44: Seman, 45: Shkumbin, 46: Ishëm, 47: Mat, 48: Bojana-Drin, 49: Morača, 50: Neretva, 51: Cetina, 52: Krka, 53: Lika. A: Core 1, B: KS02–219, C: AD76–01, D: INV12–06, E: AMC99–01, F: IN68–21, G: PAL94–66, H: PAL94–08, I: PAL94–09, J: PRAD02–04, K: KS02–357, L: RF95–13, M: IN68–22, N: LSD02–40, O: LSD02–38, P: COS01–16, Q: MAN, R: INV12–15, S: SI08–27, T: INV12–10, U: SA03–11, V: ST04–01, W: IN68–05. MAD: Mid-Adriatic Deep, SAD: Southern Adriatic Deep, MOZ: Mirdita Ophiolitic Zone, POC: Pindos Ophiolitic Complex. Vis: Vis Island, Jab: Jabuka shoal, Bru: Brusnik shoal.

Most of the above studies, however, did not involve a comprehensive source-to-sink analysis, but dealt only with segments of the system.

The Po-Adriatic is a multi-sourced foredeep system fed by three mountain belts: the Alps to the north, the Apennines in the West and the Dinarides in the East. In such a strongly heterogeneous geological setting, which hosts a variety of source rocks, exclusive catchment lithologies can be used to delineate basin-wide markers of sediment provenance (Weltje and Brommer, 2011) and trace detrital signatures across the downdip segments of the sediment dispersal system (Sømme et al., 2009): river watersheds, alluvial plains, coastal plains/deltas, shelves and basin floor (Fig. 1).

Conventional techniques of provenance analysis (sand petrography) in the Po-Adriatic region have focused mostly on the onshore, sand-prone portions of the system: notably, the Po Plain (Garzanti et al., 1998, 2006, 2011b, 2012; Marchesini et al., 2000; Vezzoli and Garzanti, 2009; Lugli et al., 2007; Fontana et al., 2015, 2019; Bruno et al., 2021; Tentori et al., 2021) and the Venetian Plain (Stefani, 2002; Monegato et al., 2010; Piovan et al., 2010; Fontana et al., 2014), with additional petrographic data from modern rivers and beaches (Gazzi et al., 1973; Gandolfi et al., 1982). In the Adriatic, except for the comprehensive study by Pigorini (1968), sand petrography has been limited to the analysis of isolated transgressive coastal deposits drowned in place during the Holocene phase of relative sea-level rise (Moscon et al., 2015).

Earlier geochemical studies in the Po-Adriatic system have been carried out on relatively small areas only or have relied upon limited numbers of analyzed samples. Despite a small database based on 21 riverbed samples and 76 prodelta samples, a quantitative sediment budget model for the Adriatic Sea was tentatively produced by Weltje and Brommer (2011). Sample analyses from 34 Italian rivers were carried out by Dinelli and Lucchini (1999). Concentration and distribution of major and trace elements were examined separately for the northern Adriatic Sea (De Lazzari et al., 2004; Romano et al., 2013; Spagnoli et al., 2014) and the central and southern Adriatic Sea (Lucchini et al., 2003; Spagnoli et al., 2008, 2010; Goudeau et al., 2013; Ilijanić et al., 2014). Frignani et al. (2005) and Lopes-Rocha et al. (2017a) focused, instead, on the narrow mud wedge along the Western Adriatic shelf. In the most proximal segment of the Po-Adriatic system, extensive previous work from the Po Plain has documented the relative impact of source-rock composition and changes in particle size on trace-metal distribution (Amorosi et al., 2002, 2019, 2020; Bianchini et al., 2002; Curzi et al., 2006; Amorosi and Sammartino, 2007; Amorosi, 2012; Dinelli et al., 2012; Greggio et al., 2018).

Unlike the Western Adriatic Sea, where sediment fluxes have been widely examined and quantified (Cattaneo et al., 2003), only scattered and largely incomplete compositional data are available from the Eastern Adriatic Sea, i.e. the Slovenian, Croatian, Montenegrin and Albanian coastal provinces, with few exceptions (Dolenec et al., 1998; Miko et al., 2003a; Rivaro et al., 2004; Halamić et al., 2012; Ilijanić et al., 2014; Razum et al., 2021). Although it is well established that smaller mountainous Albanian rivers discharge larger percentages of their sediment loads to the Adriatic Sea than do larger rivers (Milliman and Syvitski, 1992; Ciavola et al., 1999), previous research has largely neglected the possible influence of sediment supplied from Albanian sources on Adriatic seabed composition (Correggiari et al., 1996; Cattaneo et al., 2003; Syvitski and Kettner, 2007; Goudeau et al., 2013). For the purpose of modelling sediment supply, in particular, a negligible sediment fraction has been assumed to have originated from erosion in the Eastern Adriatic hinterland (Brommer et al., 2009; Weltje and Brommer, 2011). This hypothesis was based on the assumption that along the Eastern Adriatic (especially Croatian) margin, karstic phenomena are prevailing onland and shore-parallel traps may prevent fluvial sediment from contributing widely to the Adriatic sediment budget (Cattaneo et al., 2003). As a consequence, the potential contribution of Eastern Adriatic rivers to the Adriatic Sea sediment budget is virtually unexplored and sediment budget calculations at the scale of the entire Po-Adriatic system have relied upon unrepresentative datasets.

This study examines for the first time the entire Po-Adriatic sediment routing from source to sink, through the characterization of the three major areas where sediment is generated: the Alps, the Apennines and the Dinaric orogenic belt. To comprehensively assess the patterns of sediment pathways across the whole system, we present a thorough and systematic review of the literature from the whole Adriatic area, integrated by bulk-sediment geochemistry from both onshore and offshore regions. A total of 628 alluvial samples from 53 river catchments were used as compositional end-members to fingerprint source-rock composition through key elements or element ratios (Fig. 1). Prodelta and shelf deposits (444 samples) were then analyzed to investigate sediment mixing and dispersal at the fluvial-marine transition. Finally, the geochemical characterization of 326 samples from slope to deep-marine sediment cores and a comprehensive dataset of river sediment loads were used to assess the relative contribution of all potential sediment sources.

In order to capture the geochemical signatures of Alpine, Apennine and Dinaric sediment sources for the Adriatic basin, we subdivided river samples into six end-members (groups 1–6 in Fig. 1) that highlight sediment contribution from the Northern Adriatic (Group 1), Western Adriatic (Groups 2–4) and Eastern Adriatic (Groups 5–6) river catchments. Group 1 includes Eastern Alps rivers. Group 2 corresponds to the Po River catchment, with all its Alpine and Apennine tributaries. Group 3 includes Apennine rivers flowing directly into the Adriatic Sea, further subdivided into Romagna, Marche and Abruzzi-Molise catchments. Group 4 is the small province of Apulia rivers. Group 5 includes the drainage basins of Albanian rivers. Finally, Group 6 includes the Eastern Adriatic (Montenegro, Croatia and Slovenia) watersheds.

2. Oceanographic setting and sediment supply to the Adriatic basin

The Adriatic Sea is a micro-tidal epicontinental sea dominated by a cyclonic circulation driven by thermohaline processes (Artegiani et al., 1997a, 1997b; Paschini et al., 1993; Poulain, 2001). Water masses characterized by annual and inter-annual fluctuations in density, flow and gyre strength, and sediment transport capacity define the general oceanographic setting (Turchetto et al., 2007).

The Northern Adriatic Sea is shallow, with maximum depth of about 75 m. The Central Adriatic Sea has a narrower and steeper shelf, with a small basin about 250 m deep (the Mid-Adriatic Deep or MAD, also known as Jabuka Pit, Dolenec et al., 1998). The Southern Adriatic Sea is a deep basin (Southern Adriatic Deep, SAD, as deep as 1200 m) linked to the Mediterranean Sea through the Otranto Strait (Trincardi et al., 2014).

The Adriatic surface circulation is dominated by gyres centered in the north, middle and south Adriatic Sea (Fig. 2). These gyres are associated with currents that flow southward and northward along the Italian and Albania-Croatia coasts, respectively, resulting in an overall geostrophic circulation (Artegiani et al., 1997b). At a local scale, anticyclonic vorticity has been documented, such as offshore of the Po River delta (Zavatarelli and Pinardi, 2003) and the Albania-Montenegro coast (Marini et al., 2010).

The Levantine Intermediate Water (LIW) is a denser and salty water mass (29.0 kg/m^3) that forms in the Levantine Basin through evaporation during the summer and cooling during winter (Lascaratos et al., 1999). The LIW enters the Adriatic Sea through the Otranto Strait and flows in the SAD with a cyclonic path in a water depth range of 200–700 m, with average velocity of 0.13 m/s (Orlić et al., 1994). During spring and autumn, a vein of the Levantine Intermediate Water can intrude the middle and north Adriatic Sea concurring to the formation of the North Adriatic Dense Shelf Water (NAdDW; Artegiani et al., 1997a), The seasonally-modulated NAdDW is the densest water in the whole Mediterranean Sea, with densities up to 1.030 kg/m^3 and mean temperatures of ~11 °C during extreme events (Vilibić, 2003). It moves southwards

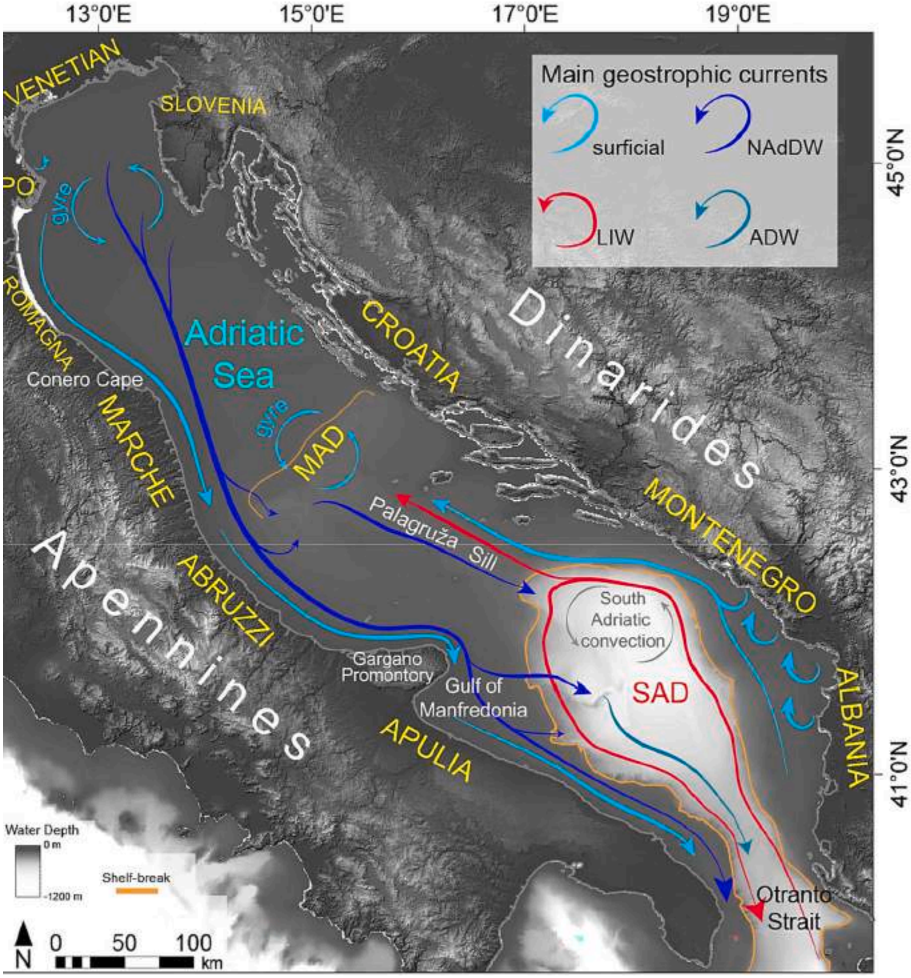

Fig. 2. Pathways of surface circulation in the Adriatic Sea: Levantine Intermediate Waters (LIW), North Adriatic Dense Waters (NAdDW) and Adriatic Deep Water (ADW) (from Artegiani et al., 1997a, 1997b; Poulain, 2001; Marini et al., 2010; Bonaldo et al., 2015).

along the isobaths of the Italian continental shelf and slope, driven by its excess density as a bottom-hugging gravity current deflected rightwards by the Coriolis force (Vilibić and Supić, 2005). The western part of the NAdDW flows nearly parallel to the western Adriatic coast, at an average velocity of 0.10–0.30 m/s (Chiggiato et al., 2016). Occasionally, storm-induced pulses of kinetic energy cause the current speed on the shelf to rise to >0.40 m/s (Benetazzo et al., 2014). A branch of the North Adriatic Dense Shelf Water sinks towards the MAD, lifting and pushing the older water masses (Artegiani and Salusti, 1987; Chiggiato et al., 2016; Langone et al., 2016; Marini et al., 2016) and promoting water exchange between the MAD and the SAD through the Palagruža sills, as well as the migration of small-scale sediment drifts (Marini et al., 2016).

Along the western shelf, the NAdDW flows around two main capes (Conero and Gargano promontories in Fig. 2), reducing across-shelf transport consistently and concurring with coastal currents to transport and deposit the sediment along the coast (Cattaneo et al., 2003, 2007; Lee et al., 2005; Palinkas and Nittrouer, 2006; Harris et al., 2008; Pellegrini et al., 2015, 2021). The NAdDW reaches the South Adriatic two months after its generation (Vilibić and Orlić, 2001; Langone et al., 2016). It strongly interacts with slope topography by enhanced turbulent mixing and finally sinks to the bottom of the SAD (Trincardi et al., 2007; Benetazzo et al., 2014). Through this process, the North Adriatic Dense Shelf Water cascades across the south Adriatic slope along its steepest sector, reaching below the depth range impacted by the contour parallel LIW (Trincardi et al., 2007; Canals et al., 2009; Bonaldo et al., 2016; Chiggiato et al., 2016; Langone et al., 2016). By continuing exchanging waters with the surrounding masses, the NAdDW mixes with the south Adriatic Deep Water (ADW, Vilibić and Orlić, 2001; Manca et al., 2002; Mantziafou and Lascaratos, 2004), formed by open-ocean vertical convection and recognized as one of the major contributors to the ventilation of deep waters in the whole Eastern Mediterranean Sea (Roether and Schlitzer, 1991). Due to its dynamic properties (buoyancy and kinetic energy), a portion of the NAdDW remains on the western shelf, flowing as a contour-parallel bottom current over several hundreds of kilometers (Fig. 2; Benetazzo et al., 2014; Bonaldo et al., 2015), generating a variety of depositional and erosional sedimentary features (Pellegrini et al., 2016; Rovere et al., 2019).

Freshwater is discharged into the Adriatic Sea from Alpine rivers (including the Po) to the northwest, Apennine rivers to the west, and Dinaric rivers in the southeast. The Po River is by far the largest Adriatic river, draining 74,000 km^2 and with average flow of 1500 m^3/s, followed by the Bojana-Drin (Albania; 20,000 km^2) and Adige (northeast Italy; 12,000 km^2). Collectively, these three rivers drain ~60% of the total catchment area of ca. 173,000 km^2. Headwaters of rivers draining the Italian Alps and Albanian Dinarides lie at elevations >2000 m, with the Po being by far the highest at 4800 m. In contrast, the vast majority of Apennine rivers have headwaters lower than 1000 m in elevation. However, in the Apennines, rivers are known to be steep and mud-rich, with intense seasonal runoffs that can produce high-density fluvial flows ("dirty rivers" of Syvitski and Kettner, 2007; Pellegrini et al., 2021). In the southeastern Adriatic Sea, the Bojana-Drin River has the largest single discharge (about 700 m^3/s) and can be considered as the southeastern Adriatic counterpart to the Po River. Several additional Albanian rivers contribute to the freshwater flux, their combined discharge being about 1250 m^3/s (Verri et al., 2018). Coastal plumes from the Albanian and Montenegrin rivers discharge extend northwards along the coast for approximately one hundred kilometers (Marini et al., 2010).

The main fluvial sediment sources of the Adriatic Basin are located along its northern, western and south-eastern sides, with a combined current delivery of ca 120 10^6 t/y of mean suspended load, with contributions of 3 10^6 t/y from eastern Alpine rivers, 15 10^6 t/y from the Po River, 32.2 10^6 t/y from the eastern Apennine rivers, 1.5 10^6 t/y from rivers south of the Gargano Promontory (Frignani et al., 1992; Milliman and Syvitski, 1992; Cattaneo et al., 2003), 59 10^6 t y^{-1} from Albanian rivers (Milliman and Syvitski, 1992; Pano, 1992; Ciavola et al., 1999; Milliman and Farnsworth, 2011; Çomo et al., 2018), and 14 10^6 t/y from Croatian rivers (Milliman and Farnsworth, 2011). Based on chronostratigraphic reconstructions and accumulation rates of Holocene sediments deposited on the shelf, the Western Adriatic Sea has been subdivided into two main depositional settings (Palinkas and Nittrouer, 2006): (i) a northern sector, between the Po Delta and Conero Cape, where reworking exceeds sediment supply by local rivers, resulting in sediment bypassing this sector (input 16.9 10^6 t/y vs sediment accumulation 7.5 10^6 t/y), compared to (ii) a southern sector, between the Conero Cape and Gargano Promontory, where net sediment accumulation is greater than the input from local rivers (input 7.8 10^6 t/y vs sediment accumulation 13.9 10^6 t/y; Frignani et al., 2005). Similar estimations for the entire Eastern Adriatic shelf remain currently unknown.

3. Catchment geology

3.1. Eastern Alps

The Alpine geological history of the Southern Alps is linked to the gentle deformation of the Tethyan passive continental margin of Adria and to the evolution of the Alpine Jurassic Tethys (Castellarin and Cantelli, 2000). The Triassic platforms and build-ups preserved in the Dolomite Mountains of the Eastern Alps (Winterer and Bosellini, 1981), exposed in spectacular natural outcrop sections (Bosellini, 1984), include thick successions of dolostones and limestones that are drained by the major North Adriatic rivers (Piave, Livenza, Tagliamento, and Isonzo in Fig. 1). A significant felsic volcanic component is associated to carbonate successions in the Eastern Alpine province, which includes granite and granodiorite of the Adamello massif and rhyolite and riodacite of the Permian Athesian volcanic group.

Modern rivers draining the sedimentary cover rocks of the eastern Southern Alps contain a very high proportion ($>$ 50%) of carbonate rock fragments (Gazzi et al., 1973; Stefani, 2002; Garzanti et al., 2006; Picard et al., 2007; Monegato et al., 2010; Monegato and Vezzoli, 2011). Owing to the abundance of these rocks in onshore catchments, dolomite is generally considered to be the main tracer of Eastern Alpine sediment provenance in the northern Adriatic region (Marchesini et al., 2000; Ravaioli et al., 2003; Spagnoli et al., 2014). The dolomite content is typically between 15 and 30% in the Venetian rivers (Brondi et al., 1979) and up to 60% close to the Venice lagoon (Ravaioli et al., 2003; Spagnoli et al., 2014). East of the Venice lagoon, in modern beach sediments between Piave and Tagliamento river mouths, dolomite is invariably more abundant than calcite (Gazzi et al., 1973).

3.2. Po River catchment

The Po River is a trunk river, 652 km long, that flows from the Alps to the Adriatic Sea and that receives sediment from a number of tributaries that drain the Alps and the Northern Apennines (Garzanti et al., 2011b – Fig. 1). Its drainage basin is made up of a variety of sedimentary, metamorphic and magmatic rocks, spanning from the Paleozoic to the Quaternary.

Composition of modern Po River sediments reflects heterogeneous sediment contributions from the Western and Central Alps, and from Northern Apennine source rocks. Diagnostic lithic signatures range from sedimentaclastic to metamorphiclastic (Garzanti et al., 2010b). Three distinct petrofacies have been recognized in the Po watershed (Tentori et al., 2021): a metamorphiclastic signature is the characteristic feature of headwater branches in the upper Po drainage basin (sediment supplied from Western and Ligurian Alps). In its upper catchment, the Western Alpine orogen consists primarily of metamorphic source-rocks, locally associated with granitoid gneisses and metaophiolites (Fig. 1). A sedimentaclastic signature with abundant limestones and dolostones and common volcanic lithic fragments typifies the lower Po drainage basin (contribution from Central Alpine Rivers – Fig. 1); whereas a sedimentaclastic signature with a larger siliciclastic component is characteristic of Apennine tributaries. Shales, hybrid arenites and thick turbidite sandstone-marl alternations are the predominant rock types exposed in the Northern Apennines. Additional mafic to ultramafic detritus is derived from ophiolites exposed at the NW tip of the Apennines and conveyed through Po right-bank (southern) tributaries.

3.3. Northern and Central Apennines

Northern and Central Apennine rivers that debouch directly into the Adriatic Sea predominantly drain sedimentary rocks. These sedimentary successions are exposed in the small catchments of 31 relatively short rivers of Romagna, Marche and Abruzzi-Molise regions (Fig. 1). Thick Miocene foredeep turbidites (sandstones and marls) crop out extensively in the Northern (Romagna) Apennines (Ricci Lucchi, 1986). Pre-Neogene (Jurassic-Cretaceous) source rocks exposed along the upper reaches of Apennine rivers consist chiefly of sedimentary successions formed during the opening of the Alpine Tethys seaway, including a dominantly pelagic domain in the Northern Apennine (Marche) area and a persistent carbonate platform in the Central Apennine (Abruzzi) region (Centamore et al., 1971; Santantonio, 1993; Rusciadelli et al., 2009). The Latium-Abruzzi platform, in particular, consists of a 5000-m-thick, discontinuous succession of limestones and only minor dolostones, of late Triassic to late Miocene age. Pliocene-Pleistocene sandstones and clays form a continuous, narrow belt cropping out at the Apennine foothills (Ricci Lucchi et al., 1982; Amorosi et al., 1998).

3.4. Apulia

Apulia rivers cut into a series of carbonate and siliciclastic sedimentary rocks exposed in the Gargano Promontory and in the Bradanic Trough, respectively (Fig. 1). The Gargano promontory is a carbonate block that is part of the slightly deformed foreland of the southern Apennine thrust belt (Bosellini et al., 1999). Mesozoic carbonates of the Apulia platform also crop out in the Murge region. In contrast, turbidites, silty clay hemipelagic deposits and conglomerates of Pliocene to Pleistocene age supplied mostly from the Southern Apennines form the dominant lithology of the Bradanic Trough (Ricchetti et al., 1992). Mudrocks, marls and chaotic clays from Miocene thrust-top basins of the Apennine belt crop out in the upper reaches of Ofanto River, the longest river of Apulia.

Highly undersaturated alkaline-potassic to ultrapotassic volcanic rocks represent a particular source material that is exposed in the Mt. Vulture volcanic complex (Fig. 1). The volcanic activity of Mt. Vulture, which was coeval with the mid-Pleistocene deformation of the southern Apennine chain, includes pyroclastic flow, pyroclastic fall and epiclastic deposits (Corrado et al., 2017). Tephra layers related to Mt. Vulture have been recognized over a wide area, about 150 km away from the volcanic center (Petrosino et al., 2015; Corrado et al., 2017). The Mt. Vulture volcanic province is drained by Ofanto River and by some of its tributaries. Sand of the Ofanto River includes a volcaniclastic component that has been partly related to the Mt. Vulture (the southern-augite province of Pigorini, 1968), but that may also reflect, at least in part, the explosive eruptions of Mount Vesuvius and other volcanoes of central and southern Italy, as recently documented by Donato et al. (2022) based on the Sr-Nd isotopic composition of pyroxenes.

3.5. Albania

The Dinaric system is a NW-SE oriented orogenic belt that includes distinct tectonic units. The External Dinarides, characterized by generally SW verging structures, consist almost entirely of Mesozoic carbonate platform successions. Ultramafic rocks associated with genetically related sedimentary successions represent the most characteristic and widespread unit of the Internal Dinarides (Dinaric Ophiolite Zone of Pamić et al., 1998). The Mirdita tectonic zone, in Albania, is the best preserved Jurassic ophiolite sequence of the Eastern Mediterranean Sea: it consists of peridotites, gabbroid rocks, tectonized harzburgite and dunite with extensive chromite deposits (Dilek et al., 2008), associated with cherts, carbonates, turbidites and mélanges units.

Several Albanian rivers flow into the Adriatic Sea. The most important ones are: the Drin, Mat, Ishëm, Shkumbin, Seman, and Vjosa. Despite relatively small basins and water discharges, Albanian rivers are active hydrological systems, extremely effective in delivering sediment to the sea (Ciavola et al., 1999). Their hydrographic basins include a great variety of rock formations, with spectacular outcrops of highly weathered ophiolite units (Xhaferri et al., 2020). Weathering of Albanian source rocks results in the disappearance of less stable components (olivine and augite) and concentration of epidote and chromite as residual minerals downstream (Pigorini, 1968). A recent petrographic analysis from beach sands of the Mat and Vjosa delta systems has shown that magnetite and chromite, which impart characteristic dark/black colour to the deposit, represent the most abundant heavy minerals (Xhaferri et al., 2020).

3.6. Montenegro, Croatia and Slovenia

The Eastern Adriatic coast is a part of the High Karst Zone (External or Outer Dinarides) that extends for about 500 km from the Italian to the Albanian borders, across Slovenia, Croatia, and Montenegro (Fig. 1). The Middle Permian to Eocene carbonate succession of the Croatian Karst Dinarides has a huge cumulative thickness locally exceeding 8000 m (Palinkaš et al., 2010). Even in Montenegro, Mesozoic carbonate rocks, highly fractured and karstified, cover over 60% of the region (Radulovic et al., 2012). Due to the high degree of karstification, permanent surface streams are rare in the region.

While the Triassic succession may include clastic rocks, dolostones and volcanogenic sedimentary formations, a thick limestone succession of Lower Jurassic to Cretaceous age forms the Adriatic Carbonate Platform (Vlahović et al., 2005). Like the coeval Apennine and Apulian carbonate platforms, the Adriatic Carbonate platform was characterized by predominantly shallow-marine deposition. Bauxites mark regional unconfomities at distinct stratigraphic levels. Tertiary rocks are most widely distributed in Dalmatia and in the central part of Istria and include flysch units (breccias, calcarenites, calcirudites, marls and shales) that crop out mostly along the Eastern Adriatic coast.

Permian and Triassic magmatism in the Dinarides produced gabbro, diorite-syenite, and granite intrusions and an extrusive sequence of basalt, andesite, and dacite lavas with abundant pyroclastic rocks (Pamić and Balen, 2005). Metabasalt, diabase, basalt, and andesitic basalt are found in Central Croatia and in Central Adriatic Sea: diabase and spilite of Late Ladinian-Late Norian age crop out on the island of Vis (Lozić et al., 2012), whereas augite diabase and gabbro are the dominant rock types on the Jabuka and Brusnik shoals in the Central Adriatic (Golub and Vragović, 1975; De Min et al., 2009; Palinkaš et al., 2010) (Fig. 1).

4. Geochemical dataset and analytical procedures

4.1. Sediment data collection

This study presents a compilation of samples and data from several research projects and from the existing literature. A total of 1398 samples (433 unpublished data and 965 data from previously published studies) were analyzed from the onshore and offshore segments of the Po-Adriatic sediment routing system (Table 1). The complete dataset is shown in Supplementary Table 1.

In the onshore part of the system, 628 modern fluvial and delta plain sediments were collected from 53 river systems and merged into Groups 1 to 6 (Fig. 1, Table 1). Samples were retrieved from exposed bars or subaqueous channel beds, and all particle sizes, from coarse sand to mud, were considered. Natural levee, crevasse and floodplain deposits were also collected through hand drilling, using Eijkelkamp Agrisearch equipment (01.11.SO hand auger set for heterogeneous soils). Where possible, we carried out a further subdivision into facies association (Supplementary Table 1): fluvial-channel sand (FC), distributary channel sand (DC), crevasse-levee sand-silt alternation (CL), floodplain clay (FP), and beach-ridge sand (BR) collected at (now inactive) fluvial mouths. In all other cases, we simply distinguished sand from mud (Supplementary Table 1).

We also considered 444 samples from 13 Adriatic shelf cores (Core 1, KS02–219, AD76–01, PAL94–09, PRAD02–04, KS02–357, RF95–13, LSD-40, LSD2–38, MAN, INV12–15, SI08–27, and COS1–16) and from previously published material, collected at depths lower than 100 m (Table 1). Finally, 326 samples were retrieved from 3 cores of the Adriatic slope (INV12–06, PAL94–08, and IN68–22), between 129 and 187 m depth; 3 cores of the Mid-Adriatic Deep (MAD - PAL94–66, IN68–21, and AMC99–01), between 214 and 255 m depth; and 4 cores of the Southern Adriatic trough (SAD - INV12–10, ST04–01, IN68–05, and SA03–11), between 566 and 1225 m depth. In order to minimize the effects of compositional changes through time, only sediment accumulated during the present sea-level highstand, with notably uniform geochemical composition, was considered.

4.2. Laboratory analyses, corrections for total concentration and statistical analysis

Bulk samples from Italian rivers (1–42 in Fig. 1) and from shelf and deep-marine cores shown in Fig. 1 were geochemically analyzed at University of Bologna laboratories according to the same sample-preparation methods. Geochemical analyses were each held to the same standards for error and reproducibility, allowing for comparable datasets. In preparation for chemical analyses, samples were oven dried at 50 °C, powdered and homogenized in an agate mortar and analyzed by X-ray fluorescence (XRF) spectrometry using a Panalytical Axios 4000 spectrometer. The matrix correction methods of Franzini et al. (1972), Leoni and Saitta (1976), and Leoni et al. (1982) were followed. The estimated precision and accuracy for trace-element determinations was 5%. For elements with low concentration (<10 ppm), the accuracy was 10%.

The Eastern Adriatic dataset includes analyses from earlier studies that were partly obtained through aqua regia digestion and inductive coupled plasma mass spectrometry (ICP-MS) analysis. It is well known that digestion with aqua regia does not fully dissolve silicates and other refractory oxides, such as Cr-bearing chromite (Bryant and Hardwick, 1950; Johnson and Maxwell, 1981; Ščančar et al., 2000; Pueyo et al., 2001; Wagner et al., 2001; Tsolakidou et al., 2002; Sutherland et al., 2004; Mäkinen et al., 2006; Chander et al., 2008). In sediments from the Po Plain, in particular, it has been documented that aqua regia digestion

Table 1

The geochemical dataset, subdivided by depositional environment, location, and published vs unpublished data.

Data Provenance					Unpublished	Published
Fluvial end-members	Group	1	Eastern Alps		12	6
		2	Po River		7	166
		3	Apennines	Romagna	8	84
				Marche	91	-
				Abruzzi-Molise	54	2
		4	Apulia		28	51
		5	Albania		-	40
		6	Montenegro & Croatia		-	79
Shelf	Northern Adriatic				-	16
	Po Delta			Core 1	-	39
	Western Adriatic		Marche	KS02-219	8	-
				AD76-01	-	15
			Abruzzi-Molise	PAL94-09	-	31
				PRAD02-04	-	43
				KS02-357	31	-
				RF95-13	12	-
	Apulia		Gargano	LSD-40	15	-
				LSD-38	7	-
			Gulf of Manfredonia	MAN	8	-
				INV12-15	37	-
				SI08-27	22	-
				COS1-16	36	-
	Eastern Adriatic		Albania		-	36
			Montenegro		-	36
			Croatia		-	29
			Slovenia/Gulf of Trieste		-	23
Slope to deep Basin	Slope			INV12-06	9	-
				PAL94-08	-	25
				IN68-22	-	20
	MAD (Mid-Adriatic Deep)			PAL94-66	-	21
				IN68-21	-	12
				AMC99-09	-	43
				various	-	13
	SAD (Southern Adriatic Deep)			INV12-10	-	37
				ST04-01	-	55
				IN68-05	-	11
				SA03-11	48	-
				various	-	32
				TOTAL	433	965

provides results that are significantly lower than those obtained through XRF analysis (approximately 45–60% for Cr and 81% for Ni, Amorosi and Sammartino, 2011). Similar results were obtained extracting metals with aqua regia by Bianchini et al. (2013), who documented that about 50% of total Cr and 90% of total Ni are recovered. From soils of Croatia, Halamić et al. (2012) reported aqua regia extraction concentrations of 57.2% for Cr, 73.4% for Ni, 77.0% for Ca, 37.1% for Al, 59.5% for Mg, and 61.1% for V.

With the aim of comparing data obtained through metal extractions with total or pseudo-total determinations (XRF; four acids mixtures: HF–HCl–HNO_3–$HClO_4$) and avoid large discrepancies among the datasets, on the basis of the above results we assumed incomplete removal and systematic underestimation of selective extractions and corrected data based on aqua regia extractions, accordingly, increasing Cr contents by 1.80%, Ni by 1.25%, V by 1.65%, Al by 2.70% Ca by 1.30% and Mg by 1.70% (Supplementary Table 1).

As the inherent inhomogeneity and incompleteness of the dataset represent an obstacle to rigorous statistical analysis (Verhaegen et al., 2019), in this study we focused on qualitative indications for sediment provenance. However, identification of potential outliers was performed through box-and-whiskers plots, where values exceeded 1.5 times the interquartile range (difference between the 75th and 25th percentiles). Following outlier removal, descriptive statistics was carried out for nickel, chromium and Ni/Cr ratios, resulting in the characterization of fluvial, shelf and deep-marine deposits in terms of: number of detects, minimum detected concentration, maximum detected concentration, sum, mean, standard error, variance, standard deviation, median, 25th percentile, 75th percentile, skewness, kurtosis, geometric mean, and coefficient of variation (Supplementary Tables 2–4).

4.3. Normalization of geochemical data

It is well established that sediment provenance reconstructions can be affected significantly by large variability in sediment texture (Weltje and von Eynatten, 2004; Garzanti et al., 2009; Liu et al., 2010; von Eynatten et al., 2012; Laceby et al., 2017) and that source-rock comminution and hydrodynamic sorting are important controlling factors of sediment composition (von Eynatten, 2004). As hydraulic-sorting effects on grain size distribution can notably impact the primary provenance signal (Garzanti et al., 2010a, 2011a; Garzanti, 2016), in order to compensate for grain-size variability of geochemical element concentration, we:

(i) controlled possible effects of size-selective transport on a sedimentological basis, by grouping samples into facies associations that represent transport invariant sub-compositions (Weltje, 2004; Garzanti et al., 2009; Razum et al., 2021); we did not adopt size fractionation, as this approach does not ensure source representativeness and commonly results in different elemental compositions based on the particle size fraction selected (Laceby et al., 2017);

(ii) normalized geochemical data using one element as grain size proxy (Loring, 1991; Daskalakis and O'Connor, 1995; Rubio et al., 2000).

Aluminium is typically used in provenance research as an effective normalization factor (Covelli and Fontolan, 1997; Menon et al., 1998; Liaghati et al., 2003; Goodbred et al., 2014). Element ratios that were adopted for the discrimination of ultramafic versus non-ultramafic source-rock composition in Po-Adriatic sediments and that proved very effective in reducing the textural effect are Cr/Al_2O_3 (Amorosi et al., 2002, 2007; Lucchini et al., 2003; Curzi et al., 2006; Dinelli et al., 2007, 2012; Amorosi, 2012; Ilijanić et al., 2014; Buscaroli et al., 2021), Cr/V (Amorosi and Sammartino, 2007; Dinelli et al., 2012; Amorosi et al., 2014) and Ni/Al_2O_3 (Dinelli and Lucchini, 1999; Amorosi et al., 2007; Ilijanić et al., 2014; Greggio et al., 2018). These indices can be used almost interchangeably. Proxies used for carbonate (dolostone) versus siliciclastic contribution based on single elemental ratios are MgO/Al_2O_3 and Mg/Ni (Picone et al., 2008; Greggio et al., 2018).

Proxies that are widely used in geochemical studies of fine-grained marine sediments, such as the Ca/Ti ratio, proved to be poorly effective in the differentiation between biogenic and terrigenous sedimentation (Ingram et al., 2010) on the variety of sedimentary facies considered in this study, as they may reflect pedogenic processes largely taking place on land (Ca) or represent particle-size sensitive elements (Bábek et al., 2015) subject to selective transport/accumulation (Ti).

5. The ‘Source’: geochemical signature of fluvial end-members

Each section of the hinterland carries unique geochemical provenance signals that reflect distinct catchment geology (Fig. 1): mafic/ultramafic rocks represent one particular component for both Po River and Albanian stream sediments, whereas an abundance of carbonates typifies Eastern Alps (dolostone-rich) and Apennine/Dinaric (dolostone-poor) deposits. The Apulian watersheds are characterized by a minor, but discernible volcanic influence.

In the following sections, we examine the six provinces that contribute detritus to the Adriatic sediment routing and suggest elemental ratios that can be used for their discrimination.

5.1. Fingerprinting ultramafic and dolostone-rich sediment sources: the Ni/Al_2O_3 vs MgO diagram

For the differentiation of mafic/ultramafic sediment sources (Groups 2 and 5 in Fig. 1) and dolostone-dominated end-members (Group 1) from ophiolite-poor and dolostone-poor Adriatic catchments (Groups 3, 4, and 6), we used the Ni/Al_2O_3 vs MgO diagram (Amorosi et al., 2007; Greggio et al., 2018, Fig. 3).

Nickel enrichments in the Western Adriatic (Dinelli and Lucchini, 1999; Amorosi et al., 2002, 2014, 2019, 2020; Bianchini et al., 2002, 2013; Amorosi and Sammartino, 2007; Curzi et al., 2006; Amorosi, 2012; Greggio et al., 2018) and Eastern Adriatic (Ilijanić et al., 2014) provinces have been widely used, normalized to alumina, as a proxy to denote material of ophiolitic inheritance. The typical mineralogical fingerprint of fine-grained, Po River-sourced material is an abundance of phyllosilicates: nickel in this size fraction is sequestrated by serpentine (Amorosi et al., 2002; Spagnoli et al., 2014). On the other hand, magnesium has been used to discriminate dolomite-rich source areas, where dolostones are largely exposed and potentially represent an effective tracer of sediment supplied from Eastern Alps rivers (Dinelli and Lucchini, 1999; Ravaioli et al., 2003; Curzi et al., 2006; Picone et al., 2008; Greggio et al., 2018; Amorosi et al., 2019, 2020).

In the binary plot of Fig. 3, most river samples exhibit a general positive correlation between Ni/Al_2O_3 and Mg values, consistent with the common association of Ni and Mg in the same minerals (e.g., serpentine). Samples from Apennine catchments, Apulia and Croatia cluster at relatively low (< 8) Ni/Al_2O_3 and low ($< 5\%$) MgO values, denoting their predominantly carbonate (limestone) or siliciclastic composition, with no particular Ni or Mg enrichments. In contrast, Po River samples are characterized by higher (8–15) Ni/Al_2O_3 levels and higher (3–6%) MgO values. Samples from Albanian rivers are markedly distinct, being characterized by very high (5–15%) MgO values and extremely high (> 30) Ni/Al_2O_3 values. This trend of increasing Ni and Mg values in Po River and Albanian river samples is interpreted to reflect increasing contribution from ophiolite rocks.

Samples from Eastern Alps rivers plot considerably off this general trend and can be readily differentiated from all other sediment sources, revealing a distinctive geochemical facies in which very high (5–15%) MgO contents, comparable to those recorded from Albanian rivers, are coupled with very low (< 5) Ni/Al_2O_3 values. The poor covariance between Ni and Mg indicates that Mg in this area is not associated with Ni-rich minerals typical of mafic or ultramafic rocks, deriving chiefly from erosion of Ni-poor (dolostone) sedimentary precursors.

5.2. Reducing particle size impacts on sediment fingerprints: the Cr/V diagram

Several studies have documented that large ultramafic (ophiolite) complexes cropping out extensively in the Western Alps and at the NW tip of the Apennine chain (Fig. 1) may deliver large volumes of Cr-rich

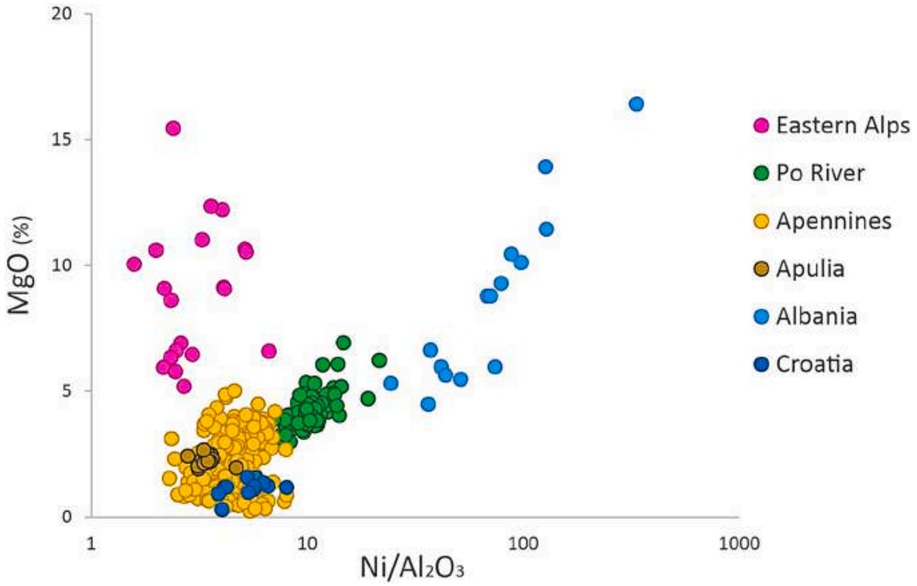

Fig. 3. Scatterplot of MgO versus Ni/Al_2O_3. Note the high-Ni and high-Mg sediment composition of (ophiolite-rich) Albanian and Po River catchments, compared to the geochemically distinct (low-Ni and high-Mg) signature of dolostone-rich, Eastern Alps rivers. Samples from Apennine, Apulia and Dinaric catchments have low-Ni and low-Mg contents. Albanian stream sediments from Shtiza et al. (2005, 2009); Croatian samples from Miko et al. (2003a), Peh et al. (2003), Salminen et al. (2005), Halamić et al. (2012), and Ilijanić et al. (2022).

detritus downstream, to the alluvial, coastal and marine segments of the sediment routing system (Amorosi et al., 2002; Amorosi and Sammartino, 2007; Greggio et al., 2018). Carrier minerals for Cr in the coarse tail of the size distribution (bed load) include weathering-resistant heavy-minerals, such as Cr-spinel (detrital chromite and magnesium chromites) and Cr-magnetite, which are common in serpentinites (Bonifacio et al., 2010). At the other end of the size distribution, within suspended-load clay-dominated deposits, Cr is largely trapped by chlorite (Bianchini et al., 2013).

Chromium-rich and nickel-rich deposits co-occur in Po Plain sediments (Amorosi, 2012) and also high Cr/Al_2O_3 values have proved to be reliable markers of ophiolite contribution from the Po River (Amorosi et al., 2002; Curzi et al., 2006; Dinelli et al., 2007, 2012). Ilijanić et al. (2014) documented that sediment from Albania yields the highest Cr/Al values in the entire Adriatic area, in response to weathering and erosion of ophiolite-rich successions of the Mirdita zone (Fig. 1).

As an additional basin-wide indicator of ophiolite-rich detritus, we adopted in this study the Cr/V ratio (Fig. 4), a key index that is commonly used to distinguish sediment derived from ultramafic source rocks (Feng and Kerrich, 1990; Garver et al., 1996; Bauluz et al., 2000; von Eynatten, 2004; Lužar-Oberiter et al., 2009; Sarti et al., 2020). In the Cr/V diagram of Fig. 4, trace metals correlate positively one another across different provinces. In particular, samples from the Po River catchment are aligned along a distinct straight regression line with the highest Cr/V ratio reflecting clear ultramafic signature. In contrast, Eastern Alps and Apulia river systems consistently show very low Cr/V values that reflect a lack of ultramafic sources in the drainage basins (Fig. 1). Relatively high Cr values from Apennines and Croatian Dinarides streams are interpreted to reflect an abundance of recycled grains of ophiolitic provenance that are common within turbidite/flysch formations in these regions (Ricci Lucchi, 1986; Lenaz et al., 1996). For scattered data from Albania, see Salminen et al. (2005) in Supplementary Table 1.

In order to emphasize compositional variations induced by selective transport and extract the provenance signal of the Cr/V ratio, we subdivided Po-related sediments into four lithofacies assemblages that reflect unique combinations of grain size and transport mechanisms across the alluvial and deltaic depositional environments (Fig. 4): (i) fluvial-channel (fine to very coarse sand), (ii) beach ridge (fine to coarse sand), (iii) distributary channel (silty sand to fine sand), and (iv) floodplain (silt and clay). For Apennine river sediments, we similarly addressed grain size effects by separating sandy (bed-load) fluvial sediment from silt-clay (suspended load) particles.

From the Cr/V diagram (Fig. 4), it is clear that Cr sourced from the Po River and from 31 Apennine river catchments is notably enriched in the finer size fractions (Amorosi and Sammartino, 2007). The generally positive correlation of chromium with vanadium can be observed across all grain-size grades, regardless of the sedimentary facies, and primarily reflects hydraulic differentiation between bed load and suspended load (see the distinct beahavior of sand and mud in Apennine samples). The linear distribution also indicates that although Cr concentration may vary considerably with grain size, the Cr/V ratio remains constant. This implies that the Cr/V index is poorly sensitive to differences in particle size and can be used effectively to trace sediment provenance.

5.3. *The geochemical signature of carbonate rocks: the CaO/Al_2O_3 diagram*

The CaO/Al_2O_3 ratio has been used in the Western Adriatic province as a key geochemical marker of sediment provenance to emphasize the carbonate versus siliciclastic contribution of river catchments (Dinelli and Lucchini, 1999; Lucchini et al., 2003). With the notable exception of dolomite-rich (Mg-rich) carbonate rocks that typify the Eastern Alps river sediments (Fig. 3), carbonate successions in the Adriatic region (Croatia Dinarides and Apennines) are made up almost entirely of limestones (Fig. 1): calcite-enriched fluvial sediments, thus, deliver remarkable amounts of Ca to the Adriatic system via distinct entry points of detritus.

Across the whole Adriatic region, CaO inversely correlates with Al_2O_3 (Fig. 5). The compositional signature of rivers draining thick limestone successions is sharply distinct and a cut-off value of CaO% around 15 clearly separates carbonate-rich from carbonate-poor drainage basins. Maximum CaO values in fluvial sediments are observed where limestones represent the most abundant sediment sources: the Croatian Dinarides, Eastern Alps, and Central-Southern (Marche and Abruzzi) Apennines. Whereas Apulia and Romagna river sediments display slightly lower CaO content (10–15%). On the other hand, remarkably lower CaO values (< 10%) characterize the Po River catchment and fluvial sediments from Albania, consistent with smaller volumes of limestones in the related catchments (Fig. 1). Soil samples from carbonate-rich provinces, such as the Croatia coastal region, may exhibit remarkably low (1–4%) CaO contents in response to pedogenesis and carbonate dissolution (Miko et al., 2003a; Peh et al., 2003; Halamić et al., 2012 in Supplementary Table 1).

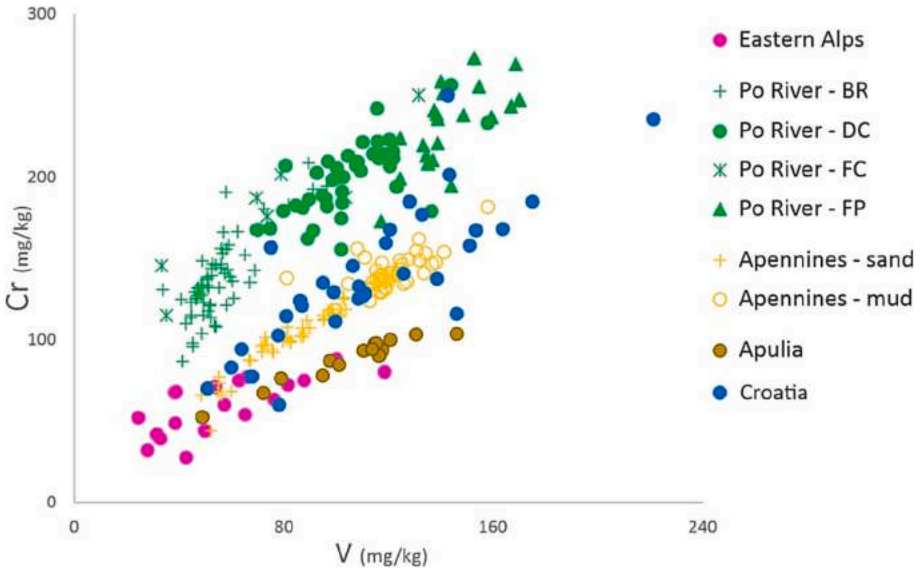

Fig. 4. Scatterplot of V versus Cr from fluvial and deltaic deposits of the Po-Adriatic system, showing metal concentration as a function of sediment provenance and particle size: samples sourced from the ophiolite-rich Po river catchment display the highest Cr/V ratios, suspended load (floodplain) deposits showing the highest Cr content. All other samples exhibit lower Cr/V values. BR: beach-ridge, FC: fluvial-channel, DC: distributary-channel, and FP: floodplain facies associations. Croatia samples from Miko et al. (2003a, 2003b), Salminen et al. (2005), Cukrov et al. (2008), and Halamić et al. (2012).

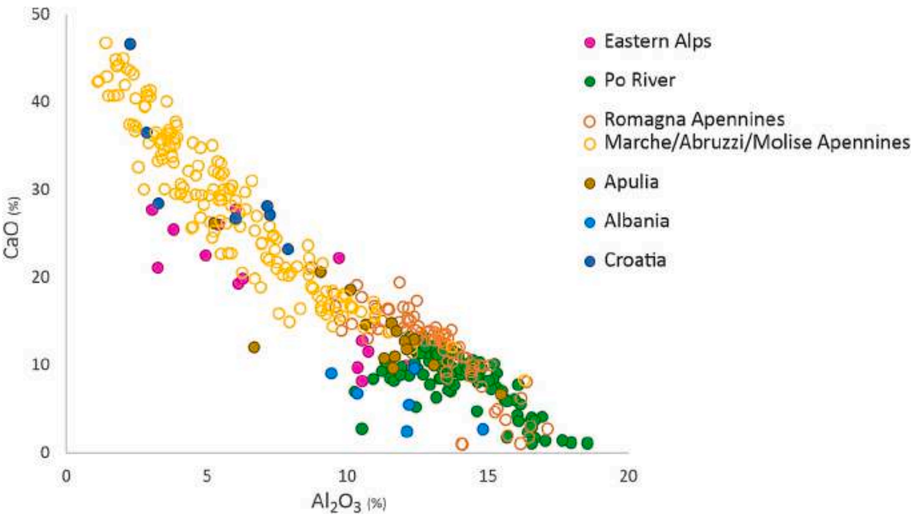

Fig. 5. Scatterplot of CaO versus Al_2O_3 from fluvial, deltaic and lacustrine deposits of the Po-Adriatic system. The highest Ca values reflect erosion and transport of detrital calcite from limestone-rich sediment sources (Croatian Dinarides, Marche/Abruzzi Apennines and Eastern Alps - see catchment geology in Fig. 1). Albanian samples from Salminen et al. (2005) and Shtiza et al. (2009); Croatian samples from Miko et al. (2003b), Salminen et al. (2005), and Ilijanić et al. (2022).

5.4. *The geochemical signature of alkaline volcanic rocks: the Ce/V diagram*

Proximity to several volcanoes of the Mediterranean area makes Apulian river catchments areas of potential accumulation and drainage of volcanic material from different sources (Corrado et al., 2017). A key element that has been used to trace sediment input from Southern Apennine volcanic sources to the Adriatic is cerium, the most abundant Rare Earth Element (Spagnoli et al., 2008; Goudeau et al., 2013). According to Spagnoli et al. (2008) and Goudeau et al. (2013), Ce is notably enriched in the Mt. Vulture volcanic rocks and conveyed through right-bank (southern) tributaries into the Ofanto River and to the coastal system of Apulia, via the Ofanto river mouth (Fig. 1).

If plotted in the Ce/V binary diagram, samples from Apulian rivers south of the Gargano Promontory are separated from all other sediment sources by significantly higher Ce values and plot towards the field of Mt. Vulture volcanic-rock composition (Fig. 6). Cerium concentration in Apulian rivers, in particular, typically falls in the 80–120 mg/kg range, whereas all other Adriatic rivers have Ce content invariably <90 mg/kg (Fig. 6). A general positive correlation between Ce and V typifies all study samples: this implies that Ce tends to be concentrated primarily in suspended material.

The relative abundance of Ce in fluvial systems of Apulia (Candelaro, Cervaro and Carapelle rivers) that, unlike the Ofanto, do not drain volcanic rocks (Fig. 1) suggests a possible additional contribution of Ce from widespread dispersal and accumulation of recent Ce-bearing pyroclastic products originating from farther volcanic centers (Fig. 1), such as the Phlegrean Fields or Mt. Vesuvius (Sulpizio et al., 2014, and references therein). Volcanic products from these volcanoes are typified by relatively high Ce concentration (Peccerillo, 2005; Smith et al., 2011) that have also been reported from tephra deposits in the Adriatic Sea (Lowe et al., 2007) and in the Mediterranean Sea (Tomlinson et al., 2012, 2015).

5.5. *Albanian catchments: A huge source of nickel and chromium for the Adriatic sinks*

The huge volumes of ophiolite rocks exposed in mountain regions of

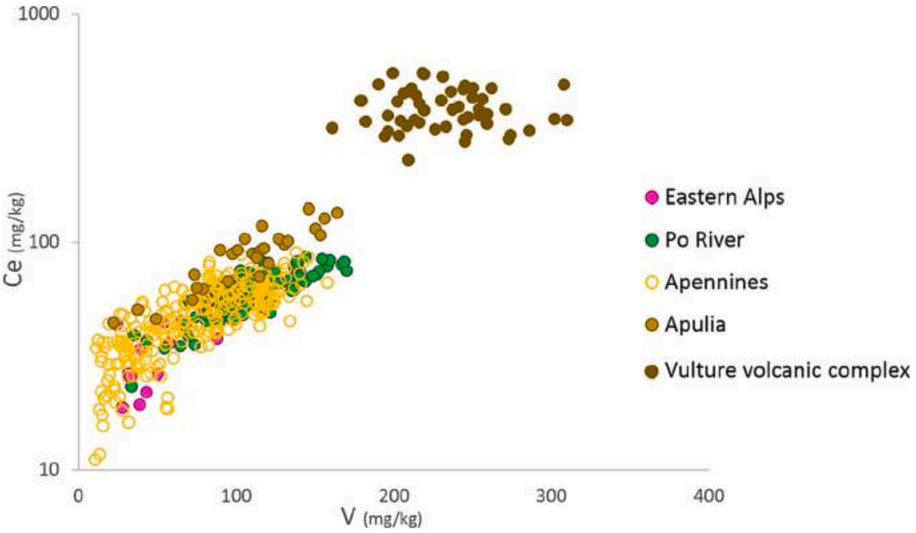

Fig. 6. Scatterplot of Ce versus V from fluvial deposits of the Po-Adriatic system and their comparison to volcanic rock composition from the Mt. Vulture complex (data from Stoppa and Principe, 1997; Beccaluva et al., 2002, and De Astis et al., 2006). Apulia river sediments are notably enriched in Ce and plot towards the Mt. Vulture end member.

Albania and in the Greek river watersheds (Mirdita Ophiolite Zone and Pindos Ophiolite Complex, Fig. 1) represent the primary sediment sources for Ni-rich and Cr-rich detritus that accumulates through erosion of highly weathered hillslopes (Dolenec et al., 1998; Ilijanić et al., 2014; Xhaferri et al., 2020). Where serpentine-bearing soils are dominant, magnesian minerals, such as serpentine (Dolenec et al., 1998; Rivaro et al., 2005) and ferromagnesian minerals, such as olivine, orthopyroxene and spinel (Albanese et al., 2015) supply large amounts of Ni from the upland catchments. On the other hand, elevated Cr contents are largely derived from rivers that drain catchments with an abundance of chromite (Xhaferri et al., 2020). Albania ranks third in the world's nickel mineral deposits and first in chromite mineral deposits (Rabchevsky, 1985).

Fig. 7 summarizes total nickel concentration from the whole Adriatic region, showing in detail metal distribution in the Eastern Adriatic provinces (Albania and Dinarides). Nickel contents from Albanian soils and from Albanian river sediments are almost one order of magnitude larger than in Western Adriatic regions and nearly two orders of magnitudes larger than in Eastern Alps sources (Fig. 7). The same holds for Cr, whose common concentration in soils and stream sediments of Albania are much higher than in all other Adriatic regions (Supplementary Table 1).

In general, a progressive decrease in trace metal concentration is recorded from the upland catchments, where Ni and Cr are presumably hosted within serpentinite rock fragments and Cr-spinel in coarse-grained bedload detritus, to relatively more distal river systems (Fig. 8). The downstream decrease in Ni content mostly reflects dilution through progressive addition of detritus from ophiolite-poor watersheds in the lower reaches of Albanian rivers, though a contribution of physical weathering (break-up and comminution) of ultramafic rock fragments during transport cannot be ruled out. This trend of depletion is best documented for both Ni and Cr along the Shkumbin River course, where Cr and Ni concentrations are reduced by approximately 45%, and 65%, respectively, along a 75 km transect (Fig. 8).

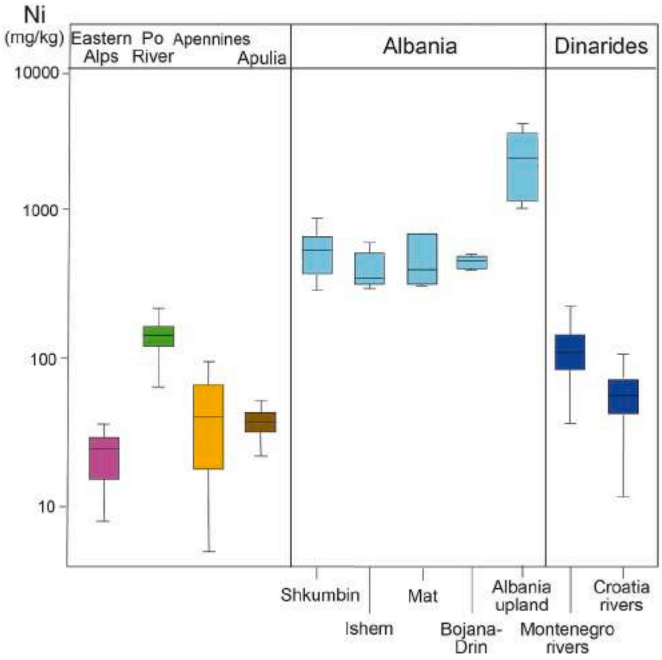

Fig. 7. Geochemical fingerprint of Eastern Adriatic (Albania and Dinarides) river catchments based upon Ni content and comparison with Northern Adriatic (Eastern Alps) and Western Adriatic (Po/Apennines/Apulia) river sediments. Highly weathered, serpentine-bearing soils from upland regions of the Albania Dinarides supply Ni-rich material to Albanian rivers. Data from the Eastern Adriatic province are corrected for total metal determinations: Albanian samples are from Shallari et al. (1998), Salminen et al. (2005); Shtiza et al. (2005, 2009), Neziri and Gössler (2006), Gjoka et al. (2011), Bani et al. (2013), and Pepa et al. (2020). Montenegro and Croatia stream sediments are from Miko et al. (2003a), Peh et al. (2003), Salminen et al. (2005), Cukrov et al. (2008), Halamić et al. (2012), Vemic et al. (2014), Jurina et al. (2015), and Milić et al. (2021). Descriptive statistics in Supplementary Table 2.

North of the Bojana river delta, catchment lithologies along the Dinaric mountain belt contain smaller volumes of ophiolite rocks. This is reflected in alluvial and coastal plain sediments of Montenegro and Croatia by progressively lower Cr and Ni contents (Fig. 7). Apart from high Ni concentration (uncorrected 145–177 mg/kg values) north of the Albanian/Montenegrin border (Milić et al., 2021) and at scattered locations (Krivokapić, 2021), mean Ni and Cr contents in alluvial to coastal sediments of Montenegro are very low (Mugoša et al., 2016) and typically <80 mg/kg (uncorrected value) at the Morača River mouth (Vemic et al., 2014; Kastratović et al., 2016; Krivokapić, 2019).

The Dinaric region of Croatia exhibits relatively higher Cr concentration (mean value from 1144 samples: 116 mg/kg – Halamić et al., 2012), with maximum values clustered in the central part of the region, between Split and Zadar, where Cr is enriched due to weathering of bauxite deposits (Halamić and Miko, 2009). The mean (uncorrected) Cr value in the Krka estuary is 91 mg/kg (Cukrov et al., 2008). Increased Cr content in the central part of the Istrian peninsula, south of Rijeka, is associated with oucrops of siliclastic flysch containing spinels, chromite, magnetite and ilmenite (Lenaz et al., 1996; Halamić et al., 2012). Nickel has a significantly lower mean value (74 mg/kg), with maximum concentration in southern Croatia, in the Split area (Halamić and Miko, 2009; Halamić et al., 2012). The mean (uncorrected) Ni value in the Krka estuary is 46 mg/kg (Cukrov et al., 2008). Along the Neretva River (Fig. 5), Ni values range between 25 and 75 mg/kg (Jurina et al., 2015; Giglio et al., 2020). Sand deposits at the Neretva River mouth exhibit Cr concentration lower than 20 mg/kg (Bogner et al., 2005), whereas a mean value of 56 mg/kg is recorded in the clay fraction (Giglio et al., 2020). Very low trace metals values are also recorded in lacustrine sediments of coastal lowland areas surrounded by karstified carbonate rocks, such as Cres Island (Miko et al., 2003b) and Pag Island (Ilijanić et al., 2022).

6. The Adriatic shelf: sediment mixing and dispersal patterns

Sediment plumes from the contributing 53 Adriatic river systems examined in this study mix with sea water at river mouths. The interaction of river and marine processes at the fluvial to marine transition zone severely impacts Adriatic shelf sediment composition. In this section, the compositional features of fluvial end-members outlined in Figs. 3-8 were adopted to assess sediment mixing at the sea bottom and trace detrital signatures and sediment pathways along the Adriatic shelf.

6.1. Northern Adriatic shelf

The primary provenance signal imparted by the particular Eastern Alps source-rock composition, typified by the abundance of dolostones in river catchments (Fig. 1), is reflected in the significant proportion of dolomite along the Northern Adriatic mud wedge, with contents generally >20% north of Po River delta (Ravaioli et al., 2003). Dolomite decreases rapidly southwards and eastwards, due to dilution by siliciclastic fine-grained sediments contributed by the Po River and Croatian river catchments. Dolomite is also abundant in offshore transgressive sands of the Adriatic that accumulated during the post-Last Glacial Maximum relative sea-level rise (Marchesini et al., 2000; Ravaioli et al., 2003).

Plotting the Venetian shelf dataset of Spagnoli et al. (2014) onto the Ni/Al_2O_3 vs MgO diagram reveals, as expected, a large overlap with the geochemical composition of Eastern Alps stream sediments (Fig. 9). The particular, high-MgO (8–11%) and low-Ni/Al_2O_3 (< 4) signature of suspended load (offshore) particles from this area is consistent with the abundance of dolomite (10–42%) observed in Venetian shelf muds

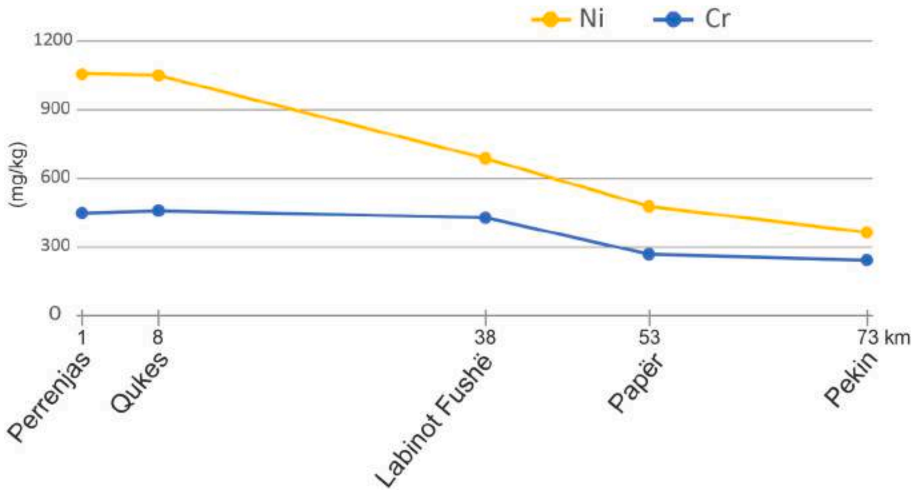

Fig. 8. Downstream decrease in Cr and Ni contents along the Shkumbin River course, in Albania (data from Pepa et al., 2020), reflecting progressive mixing with sediment from non-ophiolitic sources. Trace metal values are corrected for total metal determinations, to allow comparison with XRF data presented in this study.

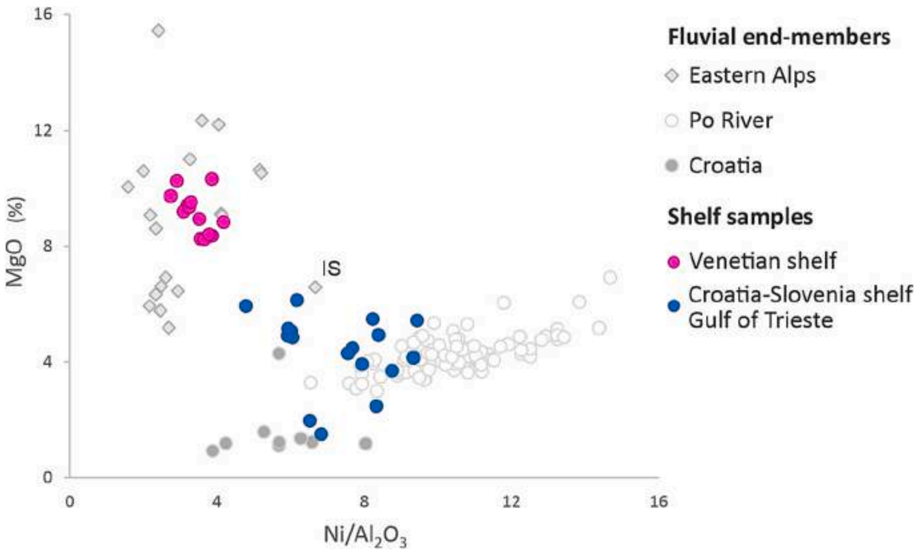

Fig. 9. Scatterplot of MgO versus Ni/Al$_2$O$_3$, showing geochemical composition of Northern Adriatic shelf sediments (in colour) compared to their putative sediment sources (Eastern Alps river, Po River and Eastern Adriatic coast). Venetian shelf dataset from Spagnoli et al. (2014); Croatia-Slovenia-Gulf of Trieste dataset from Dolenec et al. (1998), Covelli et al. (2006), Spagnoli et al. (2014), and Rožič et al. (2022). For fluvial end-members composition, see Fig. 3 (Croatia river samples from Miko et al., 2003a, 2003b; Peh et al., 2003; Halamić et al., 2012; Ilijanić et al., 2022). IS: Isonzo River sediment composition (from Dinelli and Lucchini, 1999).

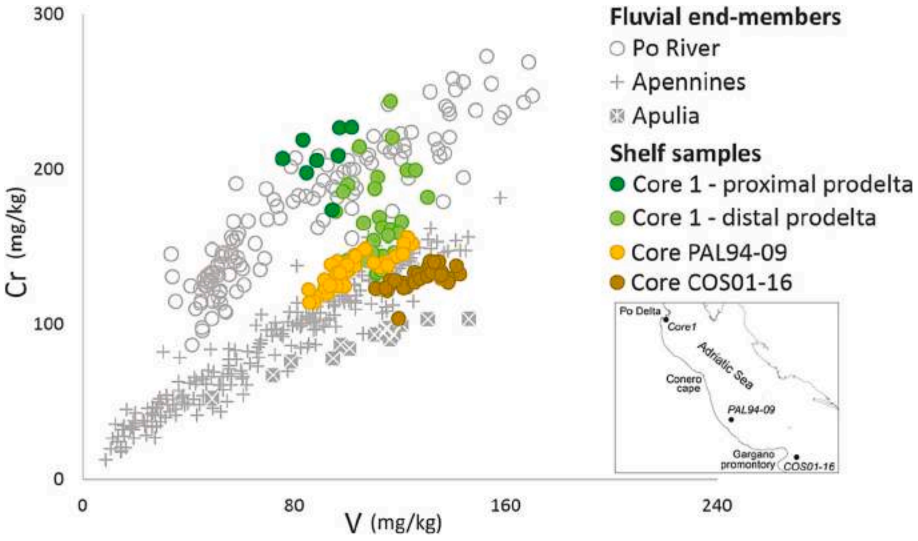

Fig. 10. Scatterplot of Cr versus V from samples of the Western Adriatic shelf (in colour: cores 1, PAL94–09 and COS01–16) and their comparison to modern fluvial sediment composition (in grey – compare with Fig. 4). Proximal prodelta muds overlap the field of Po River deposits. Sediment mixing of Po, Apennine and Apulia rivers at offshore locations is outlined by intermediate compositions of shelf samples between those of all possible fluvial end-members.

(Spagnoli et al., 2014) and denotes dolostone-derived detritus contributed by Eastern Alps sources, with almost no sediment dilution from other (Po River or Croatian) sources.

On the other hand, carbonate and flysch units from Istria and from the Northern Dalmatian islands do not appear to be the only clastic sources for sediments off Croatia, Slovenia and the Gulf of Trieste (Covelli et al., 2006; Acquavita et al., 2010; Rožič et al., 2022). Samples from the eastern Northern Adriatic shelf do not plot in the same fields as their local source rocks (Fig. 9). Higher Mg and Ni/Al$_2$O$_3$ values suggest a possible mixing effect with sediment sourced from Alpine rivers (Isonzo River, in particular) and detritus delivered by the Po River (Fig. 9). This influence of northern/western Adriatic rivers along the eastern Adriatic coast is likely due to the negligible sediment load of eastern Adriatic rivers.

6.2. Po delta and the Western Adriatic mud wedge

Sediment composition in the Adriatic basin changes distinctly south of the Po Delta (Spagnoli et al., 2014; Greggio et al., 2018). The Po River is the main source of trace metals into the Western Adriatic Sea and accounts for 45–50% of Cr and Ni delivered to the mud wedge along the Western Adriatic shelf (Lopes-Rocha et al., 2017a). Geochemical trends from prodelta and shelf clay retrieved close to the Po River mouth (Amorosi et al., 2008; Spagnoli et al., 2014; Barra et al., 2020) and their comparison to modern fluvial sediment composition unambiguously depict a consistent general picture of Po-Apennine sediment mixing in the marine environment (Fig. 10). Shelf deposits generally have similar composition as their putative fluvial sources, but yield slightly different signatures that reflect sediment mixing.

In proximal prodelta (Core 1) deposits, which accumulated at relatively low depths (< 10 m) in front of the Po river mouth, Cr/V values have a clear end-member signature, overlapping the characteristic ratio of Po River sediments (Fig. 10) and thus highlighting the highest fluvial influence. In contrast, metal contents within distal prodelta clay outline a remarkable depletion in Cr, with distinctly lower values than in fluvial or distributary-channel Po end-members. This remarkable decrease in total metal concentration is interpreted to reflect significant sediment dilution of Po-derived material by non-ophiolitic sources with increasing distance from the Po River mouth.

With the aim of assessing the degree of sediment mixing between Cr-rich (Po River-sourced) material and Cr-poor deposits from Apennine sources along the Western Adriatic mud belt, we plotted compositional data from a Central Adriatic borehole (PAL94–09) against the Apennine fluvial end-members (Fig. 10). Samples along the Western Adriatic mud belt appear to contain a mixture of Apennine and Po-derived sediments. In particular, samples from borehole PAL94–09 show consistently higher Cr content than their fluvial counterparts (Fig. 10), suggesting addition of Po River-sourced material to sediment delivered from the Apennine fluvial mouths via shore-parallel transport to the SE (Cattaneo et al., 2003; Palinkas and Nittrouer, 2007). Higher Cr values with increasing distance from the shoreline oberved along the western Adriatic shelf suggest non-random spatial metal distribution and sediment mixing along isobath-parallel pathways.

6.3. Apulian shelf

Due to the prominence of the Gargano Promontory, the Gulf of Manfredonia represents a partly shaded area relative to sediment flux from the Western Adriatic current (Cattaneo et al., 2003 – Fig. 2). In this region, the particular (Ce-rich) compositional signature of the Apulian hinterland (Fig. 6) is reflected by consistently high (> 80 mg/kg) cerium values recorded in shallow-marine deposits of core MAN, between 16 and 25 m depth, and in offshore Apulia (core INV12–15, retrieved at 15 m depth) (Fig. 11).

Samples from open shelf cores SI08–27 (30 m depth) and COS01–16 (76 m depth) reveal a different geochemical composition, plotting instead onto the field of ‘normal’, low-Ce Central Apennine/Po River composition (Fig. 11): this suggests alongshore mixing with sediment from northern sources. A model of mixed provenance for core COS01–16 is also suggested by significantly higher Cr values than those that typify the Apulian rivers end-member and that cannot be contributed from the Apulian hinterland (Fig. 10).

Consistent with inferred sediment pathways reconstructed south of the Gargano Promontory (Spagnoli et al., 2008; Goudeau et al., 2013; Pellegrini et al., 2015), the pronounced trends of increasing Cr concentration and decreasing Ce values from the shallow, protected Gulf of Manfredonia towards the open shelf reveal discernible sediment contribution from the Po River to the deep Apulian offshore, via the

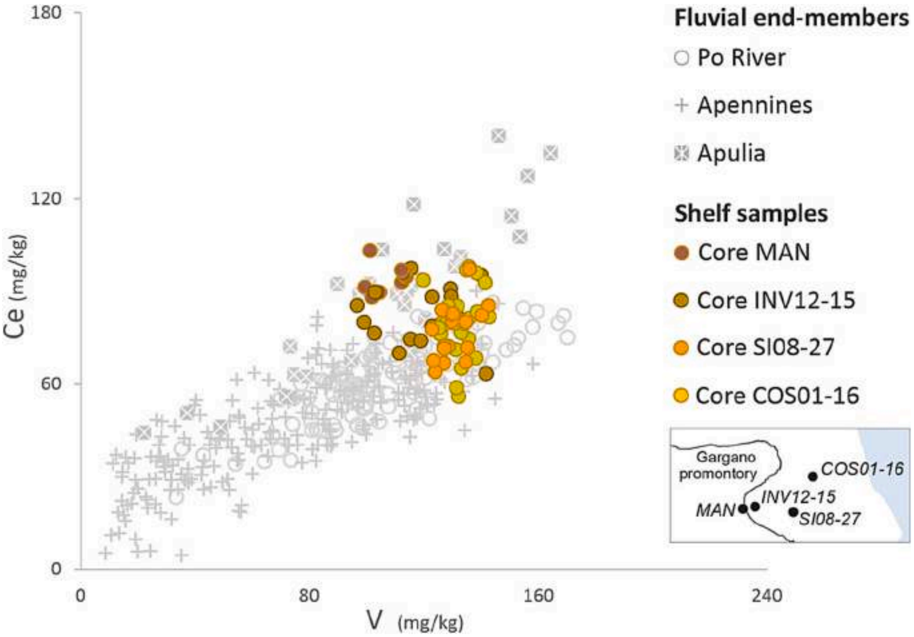

Fig. 11. Scatterplot of Ce versus V from Apulian shelf core samples (MAN, INV12–15, SI08–27, and COS01–16, in colour) and their comparison to modern fluvial sediment composition (in grey – see Fig. 6, for fluvial end-members).

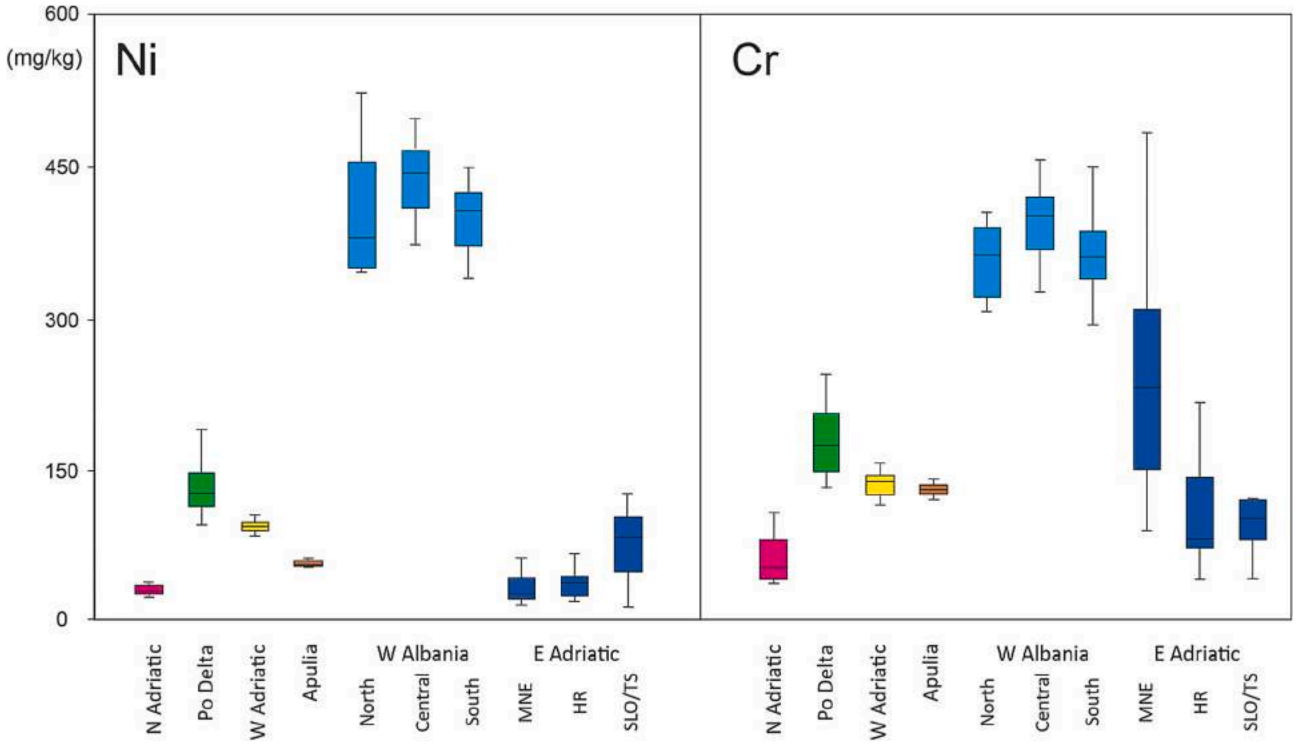

Fig. 12. Box plots of nickel and chromium concentration for shelf sediments of the Adriatic Sea Trace metals content on the Western Albanian shelf largely exceeds Ni and Cr concentrations from all other shelf areas. Nickel and Cr values from Eastern Adriatic samples are corrected for total concentration. North Adriatic dataset from Spagnoli et al. (2014). Albanian dataset from Rivaro et al. (2004). Montenegrin (MNE) dataset from Joksimović et al. (2011, 2019) and Tanaskovski et al. (2014). Croatian (HR) dataset from Dolenec et al. (1998), Bogner et al. (2005), Kljaković-Gašpič et al. (2009), Mikulic et al. (2008); Oreščanin et al. (2009), and Komar et al. (2015). Slovenian/Gulf of Trieste (SLO/TS) dataset from Dolenec et al. (1998), Covelli et al. (2006), and Ščančar et al. (2007). Descriptive statistics in Supplementary Table 3.

Western Adriatic Current in transit around the Gargano Promontory.

6.4. Albanian shelf

Comparison with trace metal values reported for all other Adriatic shelf deposits (Fig. 12) reveals that Albanian shelf sediments are markedly enriched in Ni and Cr along a > 100-km-long stretch of coast (Dolenec et al., 1998; Rivaro et al., 2004; Shehu and Lazo, 2010; Spagnoli et al., 2010; Ilijanić et al., 2014). Based on the dataset of Rivaro et al. (2004), there is no significant alongshore variation in metal concentration along the coasts of Albania, in N-S direction (Fig. 12): mean Ni values, corrected for total metal determinations, are narrowly constrained between 400 and 438 mg/kg for Ni, whereas mean Cr contents vary between about 403 and 455 mg/kg (Supplementary Table 3). Comparable trace metal contents for Ni and Cr have been reported from the same area by Shehu and Lazo (2010) and from the Durres Bay by Lazo et al. (2003).

Absolute Ni and Cr concentrations on the Albanian shelf partly overlap trace metal contents from the lowest reaches of Albanian rivers (compare Figs. 12 and 8), thus supporting the hypothesis of direct Ni and Cr derivation from the Albanian watersheds to the shoreline via fluvial sediment supplied at river mouths (Milliman et al., 2016). Consistent with geochemical data for Albanian river sediments, which place the highest trace metal values along the Shkumbin River (Fig. 7 and Supplementary Table 2), the highest Ni and Cr values along the Albanian shelf are recorded off the Shkumbin river mouth, in the central part of the Western Albanian shelf (Fig. 12 and Supplementary Table 3). The relatively lower mean metal concentration and higher data dispersion observed off the Bojana-Drin river mouth (north Albanian shelf in Fig. 12) are interpreted to reflect dilution of ophiolitic detritus released from the Mirdita tectonic zone by non-ophiolitic sediment carried by the Morača River, from Montenegro (Fig. 1).

6.5. Eastern Adriatic shelves

North of the Bojana delta, the Eastern Adriatic shelf of Montenegro exhibits a notably different geochemical signature relative to the adjacent Albanian shelf, being typified by high, though lower Cr concentration that is paralleled by very low Ni content (Joksimović et al., 2011, 2019; Tanaskovski et al., 2014 – Fig. 12). This difference in sediment composition mimics the distinct geochemical signature of the Montenegrin rivers relative to Albanian rivers (Fig. 7).

Along the Croatian coast, where carbonate rocks commonly make up 50–95% of source rocks in river catchments (Fig. 1), typical Cr values measured along the shelf or in protected bays are much lower than in Montenegro (Ujević et al., 2000; Bogner et al., 2005; Mikulic et al., 2008; Oreščanin et al., 2009 – Fig. 12), with rare exceptions (Obhođaš et al., 2006; Komar et al., 2015). Nickel is also present invariably in very low proportions (Kljaković-Gašpič et al., 2009; Mikulic et al., 2008; Oreščanin et al., 2009; Komar et al., 2015). Metal concentration in marine sediments increases with decreasing grain size (Bogner et al., 2005) and with increasing distance from the coast, reflecting the fate of suspended fluvial material entering the sea (Bulatovic and Kljajic, 2009).

Chemical composition of the Slovenian shelf and of the Gulf of Trieste partly shows higher Ni concentration, but similar Cr values to the Croatia shelf (Fig. 12), which are interpreted to reflect recycling of ultramafic material through erosion of flysch successions from Istria (Ščančar et al., 2007; Acquavita et al., 2010; Rožič et al., 2022).

7. The ‘Sink’: fingerprinting sediment provenance of MAD and SAD deposits

The Mid-Adriatic Deep (MAD), in the Central Adriatic Sea, and the Southern Adriatic Deep (SAD) represent the ultimate sites of deposition in the Po-Adriatic sediment routing system. To assess patterns of provenance and sediment pathways from the Adriatic catchments to the deep basin, we analyzed sediment composition of three cores from the MAD and four cores from the SAD (Fig. 1).

Through seismic profiles interpretation, previous work has documented a 40 km shelf-edge progradation of the Po River lowstand wedge during the Last Glacial Maximum that partially filled the MAD (Trincardi et al., 1994; Pellegrini et al., 2017, 2018). Previous geochemical investigations of Adriatic sediment cores suggested an enrichment (up to 5%) of ultramafic detritus into the MAD, likely conveyed by the Po River, thus supporting the hypothesis of enhanced contribution of the major trunk river to deep-marine sedimentation in the Central Adriatic Sea (Lucchini et al., 2003). The possible mixing between Po-derived and Apennine sediment sources in the deep-marine environment was also postulated by Weltje and Brommer (2011), consistent with petrographic data (Pigorini, 1968).

In the diagram of Fig. 13, the nickel content of 227 deep-sea core samples normalized by Al_2O_3 (Fig. 13) reveals a largely distinct sediment population, with Ni values higher than (and almost entirely nonoverlapping with) the Po end-member signature. These data are clearly at odds with the hypothesis of a unique supply to the deep basin from Western Adriatic sources and indicate that the Adriatic sinks were fed, at least in part, by sediment sources other than the Po and Apennine rivers. The Ni/Al_2O_3 signature of MAD and SAD sediments is distinctly intermediate between Eastern Adriatic (Albanian) and Western Adriatic (Po-Apennine) fluvial and shelf sediment sources (Fig. 13). A mixing pattern of multiple source areas, with a major Ni contribution from the Albanian watersheds, thus emerges as a reliable picture of sediment provenance for SAD and MAD deposits.

8. Sediment dispersal pathways in the Adriatic Sea reflect catchment lithology

Partitioning the sediment flux into its different components on a source-to-sink scale, from the Alpine, Apennine and Dinaric mountain belts to the Adriatic Sea, is not straightforward. A comprehensive set of compositional data, including (i) onshore catchments, (ii) zones of temporary storage, and (iii) final sinks is generally unavailable, and previous work has calculated sediment budgets on the basis of local investigations or isolated tie points only. In this study, characterization of 53 fluvial end-members for six compositionally distinct catchment-river systems (Groups 1 to 6 in Fig. 1) helps disentangle provenance mixing in the Adriatic Sea, within coastal/shelf to deep-water depositional environments, assessing the relative contribution of individual detrital sources.

There is a good match between source-rock lithology (Fig. 1), the general pattern of oceanic circulation (Fig. 2) and the overall spatial distribution of geochemical properties observed in Adriatic sediments (Fig. 14). Unique sources of detritus (ophiolites, limestones, dolostones and alkaline volcanic rocks - Fig. 1) drive the spatial variation of compositional tracers at the basin scale and carry distinct geochemical fingerprints that may propagate to deltaic, coastal, and shallow- to deep-marine depositional systems, and that can be applied with equal success to sand, silt and clay fractions (Amorosi et al., 2019).

Trace metals hosted preferentially in mafic and ultramafic rocks, such as Ni and Cr (Hiscott, 1984; Garver et al., 1996; Armstrong-Altrin et al., 2015) are known to carry clear provenance signals even in distal segments of the sediment routing system (von Eynatten et al., 2003; Garzanti, 2016). The spatial distribution of Ni and Cr across the Po-Adriatic region is overwhelmingly controlled by the source-rock lithology of river catchments combined with sediment loads at fluvial mouths (Fig. 14a, b). Ophiolite rocks extensively cropping out along the Dinaric chain and subordinately in the Western Alps (Fig. 1) represent major sources for these trace metals that can be traced downstream to the deepest parts of the Adriatic Sea (the MAD and SAD). Large volumes of carbonate rocks stored in the Eastern Alps, Apennines and the Dinarides (Fig. 1) deliver a significant proportion of Ca to the Adriatic Sea, where detrital carbonate mixes with an intrabasinal, biogenic component (Fig. 14c). Finally, Mg-bearing minerals (mostly dolomite) released by the dolostone-rich formations of Eastern Alps (Fig. 1) are trapped almost entirely in the Northern Adriatic Sea, but huge amounts of Mg are transferred to the Adriatic depocenter (SAD) from Albanian ophiolite sources (Fig. 14d).

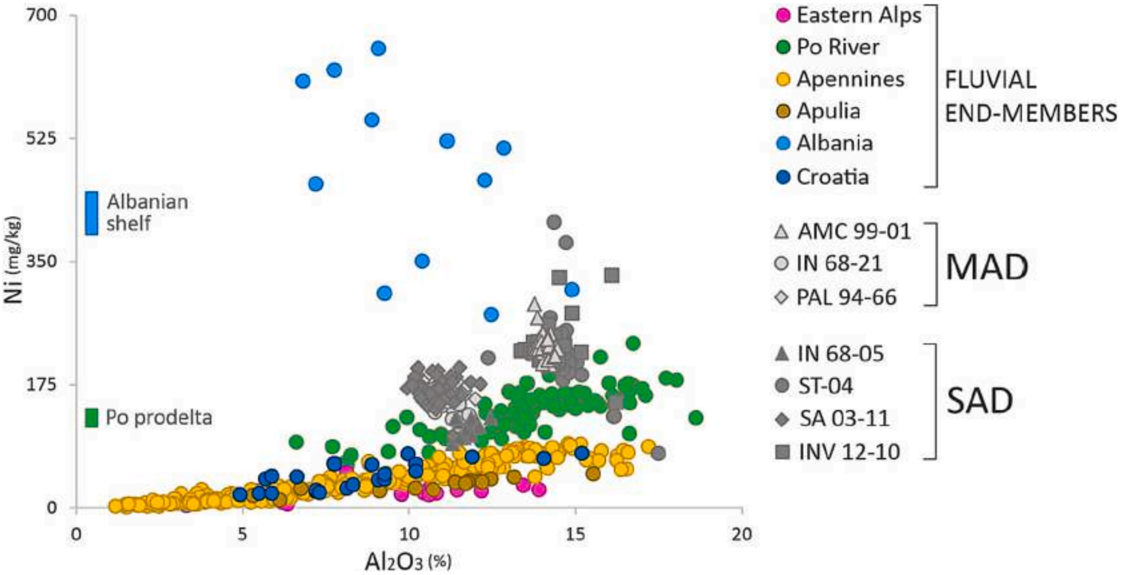

Fig. 13. The Ni/Al_2O_3 plot allows discrimination of all potential sediment sources for the deep Adriatic basins. Core data from the MAD (light grey) and SAD (dark grey) fall between the Albanian and Po fluvial end-members, suggesting mixing of two distinct sediment fluxes in the deep basins, with remarkable contribution from Albanian ophiolites (Albanian samples with Ni > 700 mg/kg are not shown). Mean nickel contents for Albanian shelf and Po prodelta sediments (Fig. 12) are reported for comparison on the y axis (data from Rivaro et al., 2004 and Amorosi et al., 2008). MAD: Mid-Adriatic Deep, SAD: Southern Adriatic Deep. Albanian end-members from Shtiza et al. (2005, 2009) and Salminen et al. (2005); Croatia end-members from Peh et al. (2003), Salminen et al. (2005), Halamić et al. (2012), and Jurina et al. (2015).

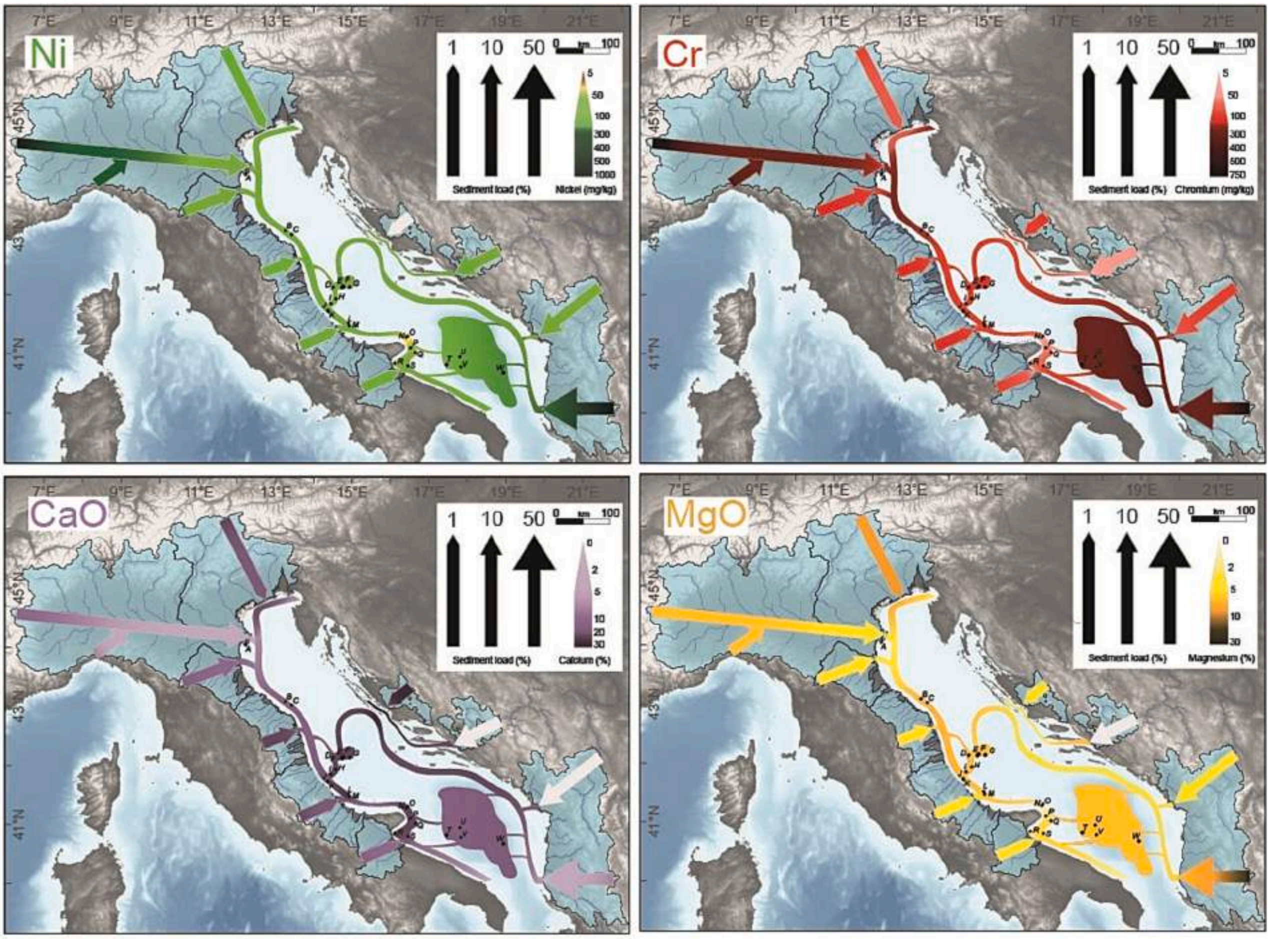

Fig. 14. Provenance interpretation maps depicting Ni, Cr, Ca, and Mg in transit across the Po-Adriatic routing system. High element signals originating in the watersheds are transferred by fluvial systems to the coast, and further across areas of temporary storage (prodeltas and shelves) to the sinks, via both longitudinal and transversal pathways. Colour intensities of idealized sediment pathways are proportional to geochemical element concentration. The size of the arrowhead reflects mean sediment loads measured at fluvial mouths. MAD: Mid-Adriatic Deep; SAD: Southern Adriatic Deep.

8.1. *Sediment dispersal in the Northern Adriatic Sea*

Modern shelf sediments in the Northern Adriatic Sea are relatively depleted in Cr and Ni (Figs. 12 and 14a, b), reflecting the remarkably low trace metal content of the Eastern Alpine hinterland (Romano et al., 2013): in this region, river catchments are virtually ophiolite-free (Fig. 1). Eastern Alps rivers flow through Mesozoic carbonate (limestone and dolostone) formations directly into the Adriatic Sea and thus contribute much CaO and MgO to the northern Adriatic shelf (Figs. 3, 5 and 14c, d). The relatively low MgO values found south of the Po River mouth (Fig. 14d) indicate that the Po Delta acts as a barrier to the influence of the Northern Adriatic counter-clockwise current.

Sediment yield from the Dinarides is negligible (Figs. 9 and 14), because of the intensely fractured and karstic nature of the catchments that trap water and sediment influx in basins close to most of the northern Croatian coastal area (Pellegrini et al., 2018). As a result, sediments from the Dinaric region of Croatia do not contribute significantly to the Northern Adriatic dispersal system.

8.2. *Sediment dispersal at the Po River mouth and in the Western Adriatic Sea*

Sediment carried by the Po River and by Apennine rivers is trapped near the western side of the basin by the cyclonic circulation of the Adriatic Sea (Cattaneo et al., 2003; Harris et al., 2008; Pellegrini et al., 2021). Model simulations of river-supplied sediment have shown that rapid-settling material tends to accumulate near the river mouths, such as in the context of the Po River, whereas unflocculated material is transported farther along the coast and accumulates in elongated deposits along the shelf (Sherwood et al., 2004).

Mixing model calculations from offshore sediment south of the Po Delta have shown that Po-derived material decreases to just 60% a few km away from the river mouth, whereas 40% would be supplied from Apennine sources (Weltje and Brommer, 2011). Progressive dilution of Po River contribution in the offshore region has also been ascertained by De Lazzari et al. (2004), who documented that Cr and Ni steadily decrease with increasing distance from the Po River mouth, and from the Italian coast towards the Croatian coast. Our geochemical characterization of the Po prodelta area (Fig. 10) fully supports these considerations.

As outlined in the previous sections, sediment dispersal from the Po River delta occurs preferentially through SSE-directed transport pathways. A powerful longshore drift under dominant southerly currents has been ascertained by a large number of studies in the Western Adriatic area (Trincardi et al., 1994, 2004a,b, 2020; Cattaneo et al., 2003; Ravaioli et al., 2003; Sherwood et al., 2004; Correggiari et al., 2005a, 2005b; Frignani et al., 2005; Weltje and Brommer, 2011; Goudeau et al., 2013; Spagnoli et al., 2014, 2021; Lopes-Rocha et al., 2017a; Pellegrini et al., 2021). Remarkable amounts of sediment sourced from the central Apennine rivers, north of the Gargano Promontory, is conveyed towards the southern Adriatic basin (Frignani et al., 2005). The decreasing influence of the Po River across this long segment of the Po-Adriatic system is mainly ascribed to the increasing dilution by local (Apennine) solid river yields.

Chromium- and nickel-rich sediment delivered by the Po River is discharged into the Western Adriatic Sea and deflected with an important southward component for about 700 km (Fig. 14 a, b). A continuous dilution of the Western Alpine metamorphic detrital signature through progressive mixing with sediment sourced from the Apennines is the dominant feature along the Western Adriatic mud belt (Fig. 10). Due to sediment dilution of Po-derived sediments by non-ophiolitic (Cr- and Ni-poor) sources, average trace metal contents progressively decrease with increasing distance from the Po River mouth. Data on metal fluxes (Tankéré et al., 2000) have shown that only 13% of dissolved nickel carried into the sea through riverine input in the Po Delta area is transferred longitudinally to the Central Adriatic.

In the Southern Adriatic Sea, Cr and Ni contents reach their minimum values (Fig. 14 a, b). Though subordinate, a plausible sediment contribution from the Po River to the Apulian offshore via the Western Adriatic Current (Goudeau et al., 2013) is indicated in this study by higher Cr and Ni values recorded on the Apulian shelf relative to metal contents typical of Apulian rivers (Fig. 10). The Po contribution to sediment transport in the Southern Adriatic Sea has been estimated to be around 15% and 30%, for the silt and clay size fractions, respectively (Weltje and Brommer, 2011).

Sediment supplied to the Adriatic from the Apulia hinterland is fingerprinted by relatively high Ce/V values (Fig. 6). Cerium in the Gulf of Manfredonia, however, is present in significant proportion only at shallow depths (Fig. 11), thus supporting the hypothesis of sediment mixing offshore Apulia between Po River and Apulian sources.

8.3. Sediment dispersal in the Eastern Adriatic and the deep sea

Whereas particulate fluxes through the western margin of the Southern Adriatic Sea have been investigated in detail (Cattaneo et al., 2003; Langone et al., 2016), little information is available for the eastern side of the Adriatic Sea, with rare exceptions (Dolenec et al., 1998; Ilijanić et al., 2014). In particular, there is poor documentation of sediment transport along transversal pathways (Pigorini, 1968).

Nickel and chromium concentrations along the Albanian shelf are remarkably higher than those found on the Western Adriatic shelf and at the Po River mouth (Fig. 12). This difference mostly reflects closer proximity of the Albanian coastal area to Cr-rich and Ni-rich source rocks cropping out in the adjacent hinterland (Figs. 1 and 14a, b). Trace metal values comparable to those that are typically recorded on the Albanian shelf can be found only in very proximal sectors of the Po routing system, in the Turin area (Madrid et al., 2006), at a considerable distance (about 300 km) upstream of the Po river mouth and almost 1000 km away from the Southern Adriatic Deep (Fig. 14a, b).

In the MAD, and especially in the SAD, a much larger proportion of Ni and Cr than values for Po River-sourced material hint at sediment mixing between the NW-directed Eastern Adriatic Current, fed predominantly from Ni-rich and Cr-rich Albanian sources, and the SE-directed Western Adriatic Current, partly contributed by the Po River (Fig. 14a, b). The hypothesis of cross-shelf sediment contribution to the SAD from Albanian sources is strongly supported by the high Mg content observed in deep-marine sediments (Fig. 14d), which is interpreted to reflect short transport from Mg-rich (serpentine-rich), ophiolite-bearing rocks. Furthermore, this hypothesis is consistent with high sediment yields reported from Albanian rivers (Fig. 14) and documented in detail by Ciavola et al. (1999). Close to the MAD, far from Albanian ultramafic sediment sources, a potentially additional supply of Cr and Ni to the Adriatic sink is from ultramafic rocks of the Central Adriatic Sea (Fig. 1) exposed on the island of Vis (Lozić et al., 2012) and in the small Jabuka and Brusnik shoals (Golub and Vragović, 1975; De Min et al., 2009; Palinkaš et al., 2010).

The overall sediment dispersal pattern depicted in Fig. 14 on the basis of trace-metal (Ni-Cr) and major oxides (CaO-MgO) distribution is in agreement with large-scale circulation in the southern Adriatic inferred from heavy-mineral assemblages (Pigorini, 1968) and clay mineralogy (Tomadin, 2000). These studies suggested a possible mixed sediment contribution to the Adriatic deep basin from Po River/Apennine and Dinaric sediment sources.

Sediment supplied by Albanian rivers, therefore, only partly accumulate in the Eastern Adriatic coastal prism. The transport of ophiolitic detritus along north-directed (along-shore) sediment pathways (the "Albanian flux" of Tomadin, 2000) is presumably driven by the Eastern Adriatic Current (Marini et al., 2010; Ilijanić et al., 2014), which could account for anomalously high Cr concentration along the Montenegrin shelf (Fig. 12) and decreasing values towards Croatia (Fig. 12).

Considering basin configuration, most ophiolite-rich detritus partly by-passed the Albanian-Montenegrin shelf, feeding the Southern Adriatic bathyal basin, reasonably through transversal paths (Fig. 14). High sediment loads from the steep Albanian rivers could be delivered at concentrations that made the river waters denser than sea water, triggering hyperpycnal flows and enhanced cross-margin sediment transfer from Eastern Adriatic sources to the deep-sea basin. Flocculation, break up and resuspension of fine-grained particles by bottom currents, storms and waves, in conjunction with transfer of suspended material from river plumes beyond the shelf via downslope density flows (dilute turbidity currents) are possible key depositional mechanisms of cross-shelf sediment transport. This hypothesis is consistent with the common occurrence of graded turbidites in cores of the SAD. Transport to the ultimate sink, however, could alternatively take place through repeated deposition and resuspension events.

9. Tracing Adriatic deep-sea sediments back to their catchment sources: the Ni/Cr ratio

In order to discriminate robustly Western Adriatic vs Eastern Adriatic sediment sources and assess their relative contribution to the deep-marine Adriatic sedimentary record, we matched the particular geochemical composition of Adriatic river catchments and adjacent shelf areas against sediment composition of MAD and SAD deposits (Fig. 15). The Ni/Cr ratio is commonly considered a robust indicator of sediment provenance from ultramaphic rocks in the source region (Garver et al., 1996; Ilijanić et al., 2014). This index proved to be an effective tool for very fine sand, silt and clay, where systematic covariation in Ni and Cr makes this ratio uniform for provenance from a given drainage basin.

Among all fluvial end-members and related shelf deposits analyzed in this study, despite largely increased Cr values due to correction for total metal determinations (e.g. Rivaro et al., 2004), alluvial and shelf sediments from Albania consistently exceeded the Ni/Cr threshold value of 0.8 (Fig. 15), with median values of 1.30 for fluvial sediments and 1.11 for shelf deposits (Supplementary Table 4). Serpentine minerals (lizardite, antigorite and chrysotile), which are particularly abundant in Albanian ophiolites, are inferred to represent the most likely carrier mineral for Ni, as confirmed by Ni/Cr ratios typically >2 from serpentine-bearing soils in Albania upland catchments (Shallari et al., 1998; Bani et al., 2013).

Nickel/chromium ratios almost invariably lower than 0.8 depict the geochemical signature of all other Adriatic deposits (Fig. 15). Po River and Romagna Apennines samples have median Ni/Cr values in the range of 0.60–0.73 (Supplementary Table 4). River deposits from Montenegro, Croatia, Eastern Alps, Marche-Abruzzi Apennines (see Surricchio et al., 2019) and Apulian yielded, instead, markedly lower (0.32–0.49) Ni/Cr values (Supplementary Table 2). There is almost no particle size influence on this elemental ratio, as documented by almost equal Ni/Cr values obtained from sand vs mud fractions in Po and Apennine river samples (Fig. 15).

Samples from seven cores of the MAD and SAD, with the notable exception of core SA03–11, systematically display Ni/Cr values >0.8 (Fig. 15 – median values between 0.84 and 1.16 in Supplementary Table

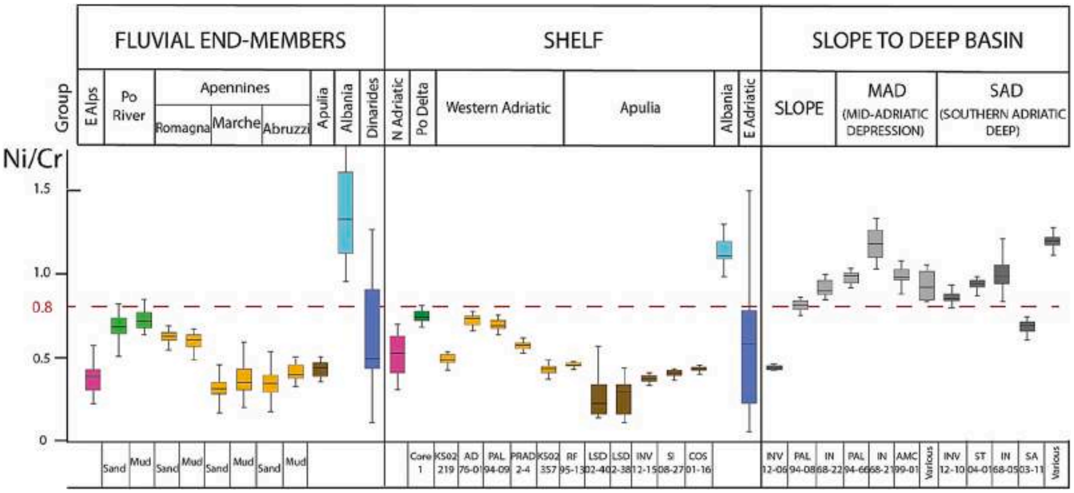

Fig. 15. Ni/Cr signature of Adriatic fluvial end-members and shelf deposits and their comparison with deep-sea sediments. Diagnostic Ni/Cr values >0.8 from the MAD and SAD suggest that Albanian watersheds contribute a significant proportion of sediment to the Adriatic sinks. Albanian fluvial samples from Shtiza et al. (2005, 2009), Neziri and Gössler (2006), Gjoka et al. (2011), and Pepa et al. (2020). Montenegrin and Croatian alluvial samples from Salminen et al. (2005), Cukrov et al. (2008), Halamić et al. (2012), Vemic et al. (2014), and Milić et al. (2021). Northern Adriatic shelf deposits from Spagnoli et al. (2014). Albanian shelf deposits from Rivaro et al. (2004). Eastern Adriatic (Montenegro, Croatia, Slovenia and Gulf of Trieste) shelf deposits from Dolenec et al. (1998), Covelli et al. (2006), Ščančar et al. (2007), Mikulic et al. (2008); Oreščanin et al. (2009), Joksimović et al. (2011), Tanaskovski et al. (2014), Komar et al. (2015), and Rožič et al. (2022). Descriptive statistics in Supplementary Table 4.

4), hinting at a major sediment contribution to the Adriatic sinks from Albanian sources, with limited Po influence. Our data are consistent with previously published material from the MAD and SAD (see "various MAD" and "various SAD" in Fig. 15, Dolenec et al., 1998; Ilijanić et al., 2014; Spagnoli et al., 2014), which also invariably provide median Ni/Cr ratios between 0.91 and 1.18 (Supplementary Table 4).

From relatively shallow shelf cores recovered along the Western Adriatic mud belt, we generally obtained low (< 0.5) Ni/Cr values, which we interpreted as supplied from Apennine sources. On the other hand, relatively deeper shelf cores (PAL94–09, AD76–01 and PRAD2–4) yielded median Ni/Cr values between 0.57 and 0.72 (Supplementary Table 4), which typify a Po River signature (Fig. 15). This particular distribution of Ni/Cr ratios is consistent with clay mineral analysis from bottom sediments in the Western Adriatic, which recognized two separate, SE-directed sediment fluxes: an Apennine flux close to the Italian coastline and a parallel, Po-influenced flux, in the open sea (Tomadin, 2000).

In summary, sediment appears to have been transferred to the Southern Adriatic sink by multiple fluvial sources (Fig. 14): Po River, Apennine rivers and Albania Dinarides rivers. Based on trace metal concentrations, Ni/Cr ratios and the distance between the mixing area (SAD) and contributing river deltas, the relative contribution of Albanian sources to the SAD is estimated to be around 75%, with a minor (25%) supply from mixed Po/Apennines sources.

10. Do high chromium and nickel contents in the Adriatic Sea reflect metal contamination?

Continental margins act as the final sink for many of the contaminants brought to the sea from the land. High Cr and Ni contents found in scattered samples of the Po-Adriatic system have raised the question whether such high trace metal values in the fluvial environment (Shallari et al., 1998; Shtiza et al., 2005, 2009; Pepa et al., 2020) or in the coastal/shelf region (Rivaro et al., 2004, 2005; Ščančar et al., 2007; Mikulic et al., 2008; Acquavita et al., 2010; Spagnoli et al., 2010; Halamić et al., 2012; Vemic et al., 2014; Lopes-Rocha et al., 2017b; Joksimović et al., 2019; Rožič et al., 2022) can be attributed to localized or diffuse anthropogenic influence. As an example, the high Cr concentration recorded in Boka Kotorska Bay, in Montenegro, an area on the UNESCO's World Heritage List, has been interpreted to reflect heavy pollution (Tanaskovski et al., 2014). Absolute metal concentrations measured in bay sediments, however, are lower than natural metal contents detected in the near Albanian coast (Rivaro et al., 2004) and might reflect southern provenance via the Eastern Adriatic current (Ilijanić et al., 2014).

In the Po Plain area, detailed documentation on modern soils and on sediment cores up to 200 m long proved that source-rock composition is the primary control on the natural spatial distribution of trace metals. "Anomalously" high Cr and Ni values, even exceeding the threshold limits designated for contaminated lands, represent the natural background and do not reflect metal contamination (Amorosi et al., 2002; Amorosi et al., 2014; Pignotti et al., 2018).

Based on the distribution of dissolved Ni close to the Po River mouth, Tankéré and Statham (1996) concluded that the Adriatic Sea is not contaminated with trace metals and that elevated concentrations reflect the natural release of metals from the source area. Similarly, Ilijanić et al. (2014) did not find any Cr or Ni enrichment in deep Adriatic sediments. Studies on metal speciation of Albanian shelf sediments have proved that the highest amounts of Cr and Ni are bound to the residue, which demonstrates that up to 80% of total metal concentration is associated to the most refractory phases, and thus related to catchment geology and only minimally ascribed to mining activities or industrial wastewater discharge, with the only exception of localized entry points in shelf regions, such as the Drin Bay (Rivaro et al., 2005; Shehu and Lazo, 2010).

Similar to what has been documented for Po Plain sediments and subsurface deposits (Amorosi et al., 2002, 2007, 2014; Amorosi and Sammartino, 2007), where previous work did not identify strong overprint of anthropogenic activity, high trace metal values in the deepest sectors of the Adriatic Sea are interpreted as due to transport and accumulation of Cr and Ni of natural origin supplied from ophiolite-rich sediment sources stored in the Albanian-Dinaric chain.

11. Conclusions

Based upon an extensive geochemical dataset and using selected geochemical indicators (trace metals and major oxides) and their downstream trends, a model for sources and routing of sediment was reconstructed across all segments of the Po-Adriatic source-to-sink system.

The geochemical composition of sediments from 53 modern streams delineates six geology-based, highly contrasting sediment sources (Eastern Alps, Po River, Apennines, Apulia, Albania Dinarides and Montenegro-Croatia Dinarides) that act as primary end-members, with distinguishable mineralogical and geochemical fingerprints. Diagnostic geochemical features from onshore catchments can be used to trace sediment dispersal at fluvial mouths onto the Adriatic continental shelf and their combination to generate complex sediment flux in the deepest parts of the basin.

Geochemical tracers used to assess the detrital fingerprint and reconstruct sedimentary transport paths through the Adriatic include Ni/Cr, MgO, Ni/Al_2O_3, Cr/V, Ca/Al_2O_3, and Ce/V. Unique source rocks, such as ophiolites, limestones, dolostones, and alkaline volcanic rocks control the spatial variation of elemental tracers on a basin scale and are ideally suited for provenance analysis in the Adriatic system. Distinct geochemical fingerprints can propagate downstream from the catchments across the alluvial plain, the coastal zone, the shelf and the deep-marine environment. The influence of hydraulic sorting on sediment composition appears negligible for the element ratios used in this work, which are equally applicable to sand, silt and clay fractions.

There is a good match between the general pattern of Adriatic oceanic circulation and the overall spatial distribution of geochemical properties observed in Adriatic sediments. Elevated concentrations of elements hosted preferentially in mafic and ultramafic rocks, such as Cr and Ni, proved to be effective, basin-wide tracers of Albanian and Po-derived sediment provenance along a 1000-km-long transect, from the Western Alps to the Southern Adriatic deep basin. The enrichment in calcite (from limestones) and dolomite (from dolostones) is reflected in the geochemistry of all grain size fractions by an abundance of carbonate-related elements, such as Mg, Ca (and their oxides MgO and CaO) and Sr in samples from the Eastern Alps, Apennines and Croatian Dinarides.

Provenance signals are not modified in transit from the subaerial alluvial system to the delta, and can be delineated to the shelf and down to the sinks. Sediment supplied from the Po River and its tributaries was tracked along its pathway southwards to the distal part of the Western Adriatic mud wedge. In this area, the ophioliticlastic (Ni-rich and Cr-rich) signature of sediment contributed by the Po River is progressively diluted by along-shore transport of siliciclastic detritus fed by Apennine sources. High MgO values in the Northern Adriatic trace the likely sediment contribution from Eastern Alpine sediment sources, with dolomite as a major carrier mineral. Whereas an abundance of MgO in the Southern Adriatic reflects sediment delivery from Albanian ophiolite-bearing successions to the shelf margin and beyond. Volcanic lithic fragments related to the Vulture/Vesuvius magmatic province of the southern Apennines can account for remarkably high Ce concentration within fluvial and shallow-marine deposits of Apulia.

The Ni/Cr ratio can be used as a reliable tracer of Albanian versus Po River ophiolite detritus. In Albania, where headwater streams flow through large serpentinite outcrops, sediment with a particular (Ni-rich) geochemical signature (Ni/Cr > 0.8) is delivered to the upstream reaches of the tributaries and conveyed by fluvial sources to the shelf/coastal zone. In the Adriatic deep basin, Ni/Cr values typically >0.8 reflect remarkable contribution of detritus generated from erosion of the Albanian orogen and transferred to the Adriatic sea floor via selective entrainement of resuspended silt and clay.

Previous work in the Adriatic area has almost ignored the contribution of Albanian rivers to the overall sediment budget. Our data challenge early assumptions about the negligible contribution of Eastern Adriatic rivers to the deep-sea stratigraphic record. Through analysis of elemental composition of bulk sediment in the Adriatic deep basins, sourced by multiple river systems, we assess the dominant role of local supply from hinterland Albanian rivers draining the Dinaric orogen versus long-distance supply from the Po River. In this scenario, this study provides background values of element concentration that can be used to highlight anthropogenic contamination in the subaqueous realm.

Sediment-budget modelling, especially from ancient successions, is rarely constrained by accurate compositional data. This study improves our understanding of sediment dispersal patterns in the Adriatic Sea and demonstrates that source discrimination based on bulk-sediment geochemistry is an effective tool for a sound assessment of the routing system. This study has highlighted the role of selected trace metals and major oxides as provenance tracers in the Adriatic area. Future research needs to build a quantitative model of sediment generation, transport, mixing, and accumulation. To this purpose, it would be beneficial to access a much larger dataset, with extensive coverage and comprehensive mapping of the Adriatic sea bottom.

This study also serves as a warning against the uncritical use of geochemical data in basin-scale reconstructions. Analytical procedures (total/pseudo-total determinations vs aqua regia extractions) may play a key role in assessing precisely major elements and trace metal contents. As data gathered from several laboratories can involve different analytical techniques of metal determination, uncorrected geochemical data obtained from acid digestion for poorly extractable geochemical elements, such as Cr, should be taken cautiously for quantitative sediment budget assessment, especially on a basin scale. The element ratios shown in this work provide qualitative indications for provenance and can be used as a guide for interpreting accurately the Quaternary sedimentary record of the Po-Adriatic system, but their values are not meant to be predictive unless fully comparable datasets are used.

Declaration of Competing Interest

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.

Data availability

Research data used in this study are available in the Supplementary material.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.earscirev.2022.104202.

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