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A workflow for analysis of compositional data in sedimentary petrology: inferring provenance changes in sedimentary basins from spatio-temporal variation in heavy-mineral assemblages

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## Supplementary Material

**Appendix 1 - Literature review of the stratigraphy and paleogeography of the southern North Sea Basin**

1. **Aquitanian and Burdigalian (early Miocene)**

Marine sedimentation commenced in the Burdigalian, following a long hiatus in the Aquitanian. These early Miocene sediments are the result of a marine transgression from a north to northwest direction (Louwye 2005). In the Antwerp-Campine basin, the Edegem and Kiel members of the Berchem Formation were deposited (Louwye et al. 2000; Louwye, 2005). These members occur in a limited area to the south of Antwerp. The base gravel contains dark rounded flint pebbles and reworked septaria from the underlying Oligocene clay. The Edegem Member consists of dark green, fine grained, clayey and very glauconiferous sand (De Meuter & Laga, 1976). Reworked foraminifers and glauconite provide evidence for substantial reworking of sediment from the underlying Oligocene (Vandenberghe et al., 1998). The Kiel Member comprises fine to medium grained decalcified sand (De Meuter & Laga, 1976). Vandenberghe et al. (1998) interpret the Edegem and Kiel members as two separate incomplete sequences, formed due to competing tectonic uplift and rising sea level.

The time-equivalent unit of the lower Berchem Formation in the Limburg Campine area is the Houthalen Member of the Bolderberg Formation (Laga et al., 2001). The Houthalen sand has a Burdigalian age and is dark green, medium fine grained, micaceous, slightly ligniferous and glauconiferous (De Meuter & Laga, 1976; De Meuter, 1970; Hooyberghs & De Meuter, 1972). These sediments are similar to the Berchem Formation though the average glauconite content is significantly lower (17%) which can be explained by a more landward position of the Bolderberg Formation (Adriaens, 2015). At the base of the Houthalen sand a transgressive gravel with reworked Oligocene components and phosphate pebbles is present (Vandenberghe et al., 1998).

Towards the north, in the Netherlands, the Lower Miocene is present in a poorly defined basal part of the Breda Formation. According to Slupik et al. (2007) no Lower Miocene sediments are present northwest of the study area, resulting in a hiatus of about 13 Ma with the underlying Oligocene. In the Venlo block northeast of the study area, the Veldhoven Formation is of Chattian to middle Burdigalian age (Munsterman & Brinkhuis 2004). The Veldhoven Formation is a predominantly sandy unit (Voort Member) followed by glauconitic grey clays (Veldhoven clay Member) and coarsening upward once more into sands (Someren Member) (Van Adrichem Boogaert and Kouwe, 1997). The top of the Lower Miocene in the RVG is represented by the marine glauconitic Breda Formation (Munsterman & Brinkhuis, 2004). To the southeast in Germany, the Lower-Middle Miocene Breda Formation laterally interfingers with the fluvial Ville Formation. Near the top of the Lower Miocene these fluvial deposits are represented by the Morken coal seam (Prinz et al., 2017; age assessment based on Berggren et al., 1995).

1. **Langhian and Serravallian (middle Miocene)**

The Antwerpen Member of the Berchem Formation was deposited in the late Burdigalian to early Serravallian in the Antwerp and Antwerp-Campine area (Louwye et al., 2000). The bulk of the Antwerpen Member was deposited from the late Langhian onwards (Louwye, 2005). It consists of dark green, medium fine grained, slightly clayey and very glauconiferous sand (De Meuter & Laga, 1976). The average glauconite content is the highest of any unit in the Campine Basin (Adriaens, 2015), and, in contrast to the Lower Miocene members, contains mostly authigenic glauconite (Vinken, 1988). The dinoflagellate cyst assemblage indicates an overall shallowing up of the depositional environment (Louwye et al., 2010). The Antwerpen Member is the third incomplete sequence of the Lower to Middle Miocene. The presence of authigenic glauconite, phosphate pebbles and hardgrounds near the base of the Antwerpen Member are interpreted as a transgressive surface. Each member of the Berchem Formation represents the transgressive and/or highstand phase of a different sequence (Vandenberghe et al., 1998). Maximum flooding occurred in the early Middle Langhian (Louwye, 2000; Louwye et al., 2000).

In the Limburg Campine area, the near shore to continental Genk Member of the Bolderberg Formation is deposited as a lateral equivalent of the Antwerpen Member (Louwye & Laga, 2008; Deckers & Louwye, 2017). It consists of yellow white sand varying from fine to coarse grained and including lignite and quartzite banks in the Opgrimbie facies (De Meuter & Laga, 1976).

The basal sequence of the Breda Formation to the northwest is a Middle Miocene sequence with a basal gravel of phosphorite pebbles followed by maximum flooding and a coarsening upward trend, which can be correlated to the Antwerpen Member (Slupik et al. 2007). In the Venlo block, the Middle Miocene of the Breda Formation is about 150 m thick (Munsterman & Brinkhuis, 2004). In the southeastern RVG, the Heksenberg Member of the Ville Formation is deposited in between the Morken and Frimmersdorf lignite layers. These are a continental lateral equivalent of the marine Breda Formation (Van Adrichem Boogaert & Kouwe, 1997). The MMU (Mid Miocene Unconformity) in the LRE is interpreted as a siliciclastic interlayer in the Frimmersdorf coalseam, which is dated at 13.2-14.8 Ma, near the Langhian – Serravalian boundary (Schäfer & Utescher, 2014). In the German North Sea sector the MMU is a hiatus comprising the mid Serravallian-earliest Tortonian age, dinoflagellate cyst zones DN6 and DN7 (ca. 11.2-13.3 Ma), which is similar to that determined for well Goirle, in proximity of the Belgium-Dutch boundary (Kothe et al., 2008). In the Danish offshore sector, the MMU is correlated to a condensed section of glaucony-rich mud, spanning several millions of years from ca. 13 Ma (Rasmussen and Dybkjaer, 2014). The Heksenberg Member and coal seams can be correlated with the regressive top of the Bolderberg Formation, followed by a period of non-deposition in the Campine Basin during the Serravallian related to the MMU (Demyttenaere, 1989;Wouters & Vandenberghe, 1994;Louwye & Laga, 2008)**.** In the Belgian part of the RVG, 40 m of marine sediments occur below the Upper Miocene which have no known counterpart in the Campine Basin, possibly due to continued deposition in the RVG during this period when the Campine Basin underwent emersion and erosion (Vandenberghe et al., 2005; Vandenberghe et al., 2014).

1. **Tortonian and Messinian (late Miocene)**

Sedimentation resumed in the Campine Basin in the Tortonian. The Diest Formation consists of mostly decalcified coarse glauconite-rich marine sand and has a basal gravel of rounded flints. The Dessel Member can be recognized at the base of the Diest Formation based on its finer grain size (De Meuter & Laga, 1976). The Deurne and Dessel members of the Antwerp and Antwerp Campine area have an early to middle Tortonian age. The bulk of the Diest Formation has a late Tortonian to early Messinian age (Louwye & Laga 1998, 2008; Louwye et al. 2007). Both the Dessel and Diest sand are highly diachronous units, younging towards the northwest in the Campine Basin as a progradational system (Louwye et al., 1999; Louwye, 2002).

The most distinctive feature of the Diest Formation is its erosional base, forming a deep gully in the Hageland area, which erodes up to 100 m of Oligocene clay. In the Campine area, erosive channels can also be recognized at the base, but are less pronounced (Gulinck, 1962; Vandenberghe et al., 1998; Vandenberghe et al., 2014). The strong incision is classically related to subsidence of the RVG and uplift of the Brabant Massif combined with the opening of the English Channel, which causes strongly erosive coast-parallel tidal currents in a sea strait connecting the English Channel with the RVG (Gullentops, 1957; Houbolt, 1982). Diest sediments filled this gully and formed prograding tidal sand banks, which are preserved in the current Hageland hilly morphology due to rapid emersion and the formation of iron banks. In this model, the iron-rich top sediments of the Flemish Hills west of Brussels are considered as belonging to the Diest Formation and linked up with the Hageland Diest Formation. The stratigraphic position of these sediments, however, remains a point of discussion as no biostratigraphic data are available (Houthuys, 2014). Vandenberghe et al. (2014) propose a model for the deposition of the Hageland and Campine Diest sand in two different sequences. During the early Tortonian, the Deurne and Dessel members were deposited in the Antwerp and Antwerp-Campine area. At the same time, the Hageland gully was flooded, followed by the deposition of large sand waves which are preserved in the Hageland landscape. During the second sequence in the late Tortonian, the Hageland Hills were raised above sea level due to tectonic uplift of the Brabant Massif and the Campine Diest sand is deposited.

The final Miocene transgression led to the deposition of the marginal marine Kasterlee Formation. This formation consists of decalcified slightly glauconitic gray fine micaceous sand with micaceous clay lenses (De Meuter & Laga, 1976). The paleoenvironmental analysis of dinoflagellate cysts confirms a shallow marine restricted environment with an important continental influx. Due to the very shallow depositional conditions strong lateral variations in facies occur and a basal gravel can only be found in the most proximal environment to the south (Louwye et al., 2007; Louwye & De Schepper, 2010; Verhaegen et al., 2014). The Kasterlee Formation was deposited during the Messinian (Louwye et al., 2007). After this depositional sequence, a hiatus of 2 Ma occurred before the Pliocene Poederlee and Mol formations were deposited in the Antwerp-Campine and Limburg-Campine (Louwye & De Schepper, 2010).

The second sequence of the Breda Formation northwest of the study area was deposited above the MMU during the Late Miocene, separated from the first sequence by a phosphorite pebble lag (Slupik et al., 2007). The more continuous record in the Venlo Block contains approximately 60 m of Upper Miocene Breda Formation (Munsterman & Brinkhuis, 2004). The extensive peatlands and marches which developed in the RVG during the transgression in the early to middle Miocene, were again reclaimed by fluvial systems of the pre-Rhine in the late Miocene and Pliocene. The main sediment sources for these fluvial systems were the Ardennes and Rhenish Massifs (Schäfer et al. 2005; Schärer et al., 2012; Tatzel et al., 2017). These sediments have a distinctive delta front architecture with large prograding foresets, up to approximately 500 m, as visible in wells SMG-01 and HSW-01 (Van Adrichem Boogaert & Kouwe, 1997). The late Tortonian to Pliocene fluvial to deltaic Kieseloölite Formation progrades much further northwest into the RVG than the Inden Formation. It is laterally equivalent to the marine Breda and Oosterhout formations. The Late Miocene part of the Kieseloölite Formation is represented by the Waubach Member which consists of coarse sand and gravel (Van Adrichem Boogaert & Kouwe, 1997).