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# Unravelling the enigmas of the 'silver sands' in the Dutch/German/Belgian border area

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## Abstract

Bright white sands consisting almost exclusively of quartz (sometimes called 'silver sands') occur throughout the world; those in Europe commonly date from the late Paleogene and early Neogene. They have a clearly sedimentary origin, and they may have originated in various types of environments, but precise data are lacking because sedimentary research into these deposits (that have a high economic value) has been scarce. It is most likely that diagenetic processes are largely responsible for their exceptional appearance, but it is highly unlikely that all silver sands were subject to the same diagenetic conditions.

The precise origin of most of the silver sands is still enigmatic. In the case of the silver sands in the Dutch/German/Belgian border area, it appears that long-lasting *in situ* leaching by humic acids (resulting in an extremely low percentage of heavy minerals), in combination with differential cementation (and later partial dissolution of the cement), must be held responsible for the wide variety of the characteristics of these sands, including locally sharp boundaries with the underlying sands, lack of precipitates at the contact plane with the underlying sands, and the joint occurrence of strongly weathered and fresh specimens of the same heavy-mineral species.

***Keywords***: leaching, weathering, heavy-mineral analysis, unlithified sediments, diagenesis

## Introduction

The most common constituent of siliciclastic sands and sandstones is quartz. Sandstones consisting exclusively of quartz do not exist, however, although some quartzites come fairly close. The same holds for sands: they commonly contain a few to some tens of percents of other sand-sized minerals, small rock fragments and, in addition, a certain amount of clay minerals. Iron is commonly present in the form of coatings or as cement in the form of oxides and/or hydroxides. The less iron is present and the higher the percentage of quartz, the more the colour of the sand changes from brownish-yellow to white. Bright white sands are exceptional, but they occur; in Europe such deposits – sometimes called 'silver sands' – are not uncommon (O'Driscoll, 2004), although the extent of the individual silver-sand bodies is often restricted.

The occurrences of the silver sands dealt with here occur in a WNW-ESE trending zone that is, at least partly, bounded by faults that are related to the Rhine Graben tectonics. The westernmost sands are found in the Belgian Kempen (Campine) region (among other locations present in the Sibelco sandpit near Maasmechelen); other sands occur in the southern part of the Dutch province of Limburg, and the easternmost representatives are located in the adjoining westernmost part of Germany. The distance from the westernmost to the easternmost silver-sand pits dealt with here is about 35 km (Fig. 1), and the belt with these sands is some 10-15 km wide in the Netherlands.

### *Stratigraphic context*

There has long been much uncertainty about the precise stratigraphic position of the various silver-sand occurrences in the Dutch/German/Belgian border area (Fig. 1), mainly because they cannot be dated themselves; the German silver sands are, however, intercalated between browncoal seams, and this is partially true for the Dutch occurrences, which makes it possible – on the basis of palynology – to attribute both a maximum and a minimum age to the German and Dutch silver sands. This

*Fig. 1. Location map, showing the silver-sand pits studied in the Netherlands, Germany and Belgium.*

is not possible, however, for the sands that occur in Belgium close to the border with the Netherlands, and it has been presumed for a long time that these Belgian occurrences had a different age. It seems now, however, that all silver sands in the Dutch/German/Belgian border area have an Early to Middle Miocene age, but that they slightly young from West to East (Fig. 2).

In the Netherlands, silver sands occur in the southern part of the province of Limburg (Kuyl, 1973), where they are nowadays exploited in two quarries: Sigrano and Heerenweg (commonly called 'Beaujean'; in fact the latter quarry consists of two units, one West and one East of the Heerenweg), both situated near the village of Heerlerheide (Van der Waals et al., 1962). They constitute the Heksenberg Member of the Breda Formation (Westerhoff et al., 2003) and form bodies of up to about 90 m thick, with a lateral extent of a few kilometres. These deposits were considered as Late Oligocene to Miocene by Muller (1943) and De Jong & Van der Waals (1971), but the position between under- and overlying browncoal wedges (the Morken and Frimmersdorf seams, respectively) of the Ville Formation point at a Miocene age (Manten, 1958; Utescher et al., 2002), more specifically a late Early Miocene to early Middle Miocene age (Utescher et al., 2002).

The sands in the westernmost part of Germany (Neurather Sands), situated between the underlying Frimmersdorf and the overlying Garzweiler browncoal seams (Fig. 2), form part of the Ville Formation that are dated as Middle Miocene (Hilden, 1988) and that can be correlated with the lower part of the Vrijherenberg Member of the Breda Formation in the Netherlands (Van Adrichem Boogaert & Kouwe, 1997). They are exploited in the Nivelstein sandpit, situated near Herzogenrath, just past the Dutch/German border East of Heerlen), where also a well-cemented facies occurs.

In the Belgian quarry of Sibelco at Maasmechelen, silver sands occur that are known as the Opgrimbie Sands (named after a now abandoned quarry that was situated only a few hundreds of metres away from the operational Sibelco quarry), which belong to the Bolderberg Formation (Wouters & Vandenberghe, 1994). The Opgrimbie Sands have also a Miocene age (presumably late Burdigalian: Wouters & Vandenberghe, 1994), as can be deduced from a browncoal layer of up to 7 m thick that subdivides the white sands into a lower and an upper unit (Buffel et al., 2001).

## *Economic importance*

The silver sands in all three countries are exploited because their extremely low content of iron, in combination with the exceptionally high percentage of quartz, makes them a good resource for the glass industry. The fine-grained character, in combination with the good rounding of the grains, makes some of these occurrences a good resource for the manufacturing of household abrasives. The ceramics and chemical industries are

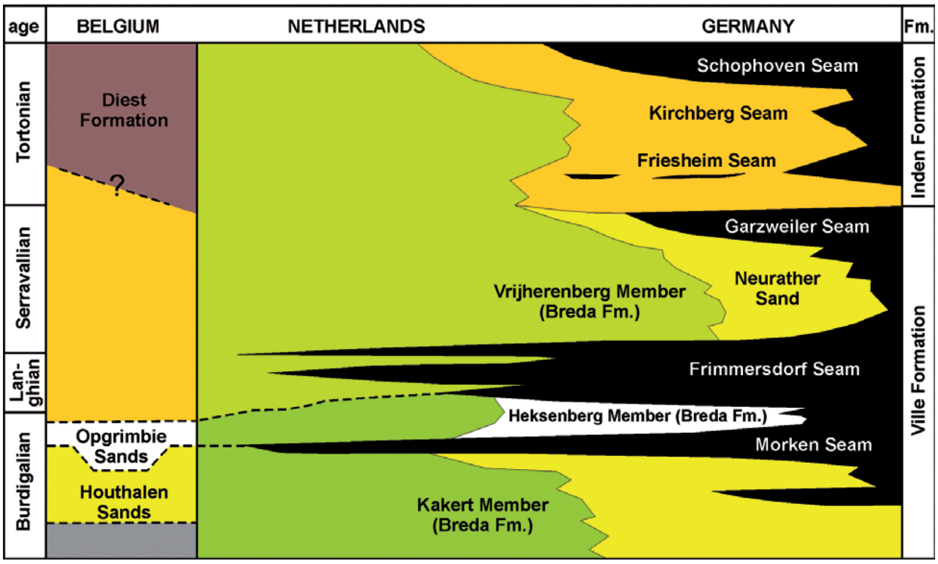


*Fig. 2. Tentative stratigraphy of the Miocene sands and browncoal occurrences in the German/Dutch/Belgian border area. Adapted from Wouters & Vandenberghe (1994) and Utescher et al. (2002).*

also important consumers of these sands (Reihe, 1999). The sands have locally been cemented (Fig. 3; see also figs 4-5 in Gulinck, 1961), and the sandstones – which are known from the Opgrimbie, Beaujean and particularly the Nivelstein quarries (in Germany they are called 'Nivelstein Sandstone') – commonly show a light-brown to yellow colour after weathering. They have been used from Roman times on, until some 75 years ago, as building stone. Particularly some churches, also in the Netherlands, illustrate this use (Fig. 4).

The most important property of the silver sands that makes them so valuable nowadays is that they consist now for at least 98% of quartz (see also Table 4 in Van der Meulen et al., 2009). The iron content in the sands that occur between the Morten and Frimmersdorf seams is extremely low (up to 0.01%) and aluminium oxides form maximally 0.025% (Laban, 2007). The content of heavy minerals in this unit is exceptionally low (sometimes not more than 0.003%: De Jong & Van der Waals, 1971); most of the heavy minerals that are still present belong to leucoxene-like opaque specimens; among the translucent grains, ultrastable heavy minerals dominate (Fig. 5) (see also Gullentops, 1972/1973, for heavy-mineral analyses of the Opgrimbie Sands). The silver sands at other stratigraphic levels show a somewhat lower quality (~97% $SiO_2$, ~0.35% $Al_2O_3$ and 0.03% $Fe_2O_3$: Dubelaar & Menkovic, 1998).

Fig. 3. Horizon of approx. 2.5 m thick with (slightly) cemented silver sand (Beaujean West quarry).

Fig. 4. Well-cemented blocks of Nivelstein Sandstone have been used as building stone, as in this church at Gulpen (the Netherlands). The colour of the stones indicates the presence of iron (hydr)oxides, which may be an indication that the sands had become cemented before complete leaching had taken place.

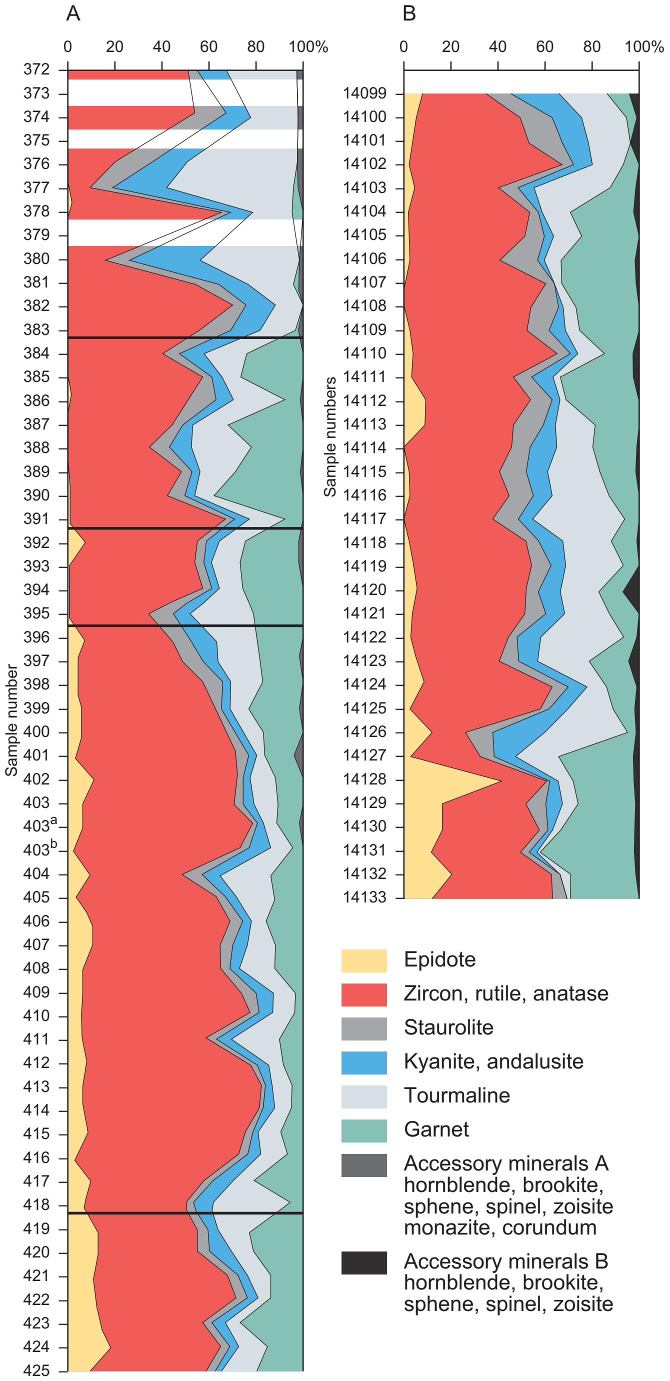


*Fig. 5. Heavy-mineral analysis of a section through the silver sands in (A) shaft II of the previous Dutch coal mine Hendrik and (B) shaft IV of the same mine. The shafts of the abandoned mine are no longer accessible. Adapted from Van Loon (1972/1973).*

Although the sands have an extremely low heavy-mineral content, the few remaining heavies diminish the applicability for several industrial purposes: monazite and rutile, for instance, may result in stains in cloths washed with washing powder containing the fine fraction of the silver sands.

In spite of the economic importance of the silver sands, relatively little is known about several aspects; in fact so little that they are still fairly enigmatic. This is remarkable, because detailed geological maps of the areas with silver sands exist in all three countries, and the deposits have been fitted in the local or national stratigraphic frameworks that, as a rule, have been analysed in detail with respect to both their horizontal and vertical extent and their genesis.

Van der Meulen et al. (2009) calculate that the total volume of sands in the Netherlands that they identified as potential silver-sand resources (of both low and high quality) can be calculated as 6 km$^3$, with an additional volume of 9 km$^3$ if an occurrence in the eastern parts of the provinces of Gelderland and Overijsel is included (but they are not sure whether this low-grade material can be sufficiently upgraded). The Dutch reserves of silver sands in the province of Limburg that can be exploited before 2050, amount to 70 × 10$^3$ m$^3$ (Maimone et al., 1985); a much larger body of silver sand is present (De Mulder & Ritsema, 2003), but cannot be exploited because of a location in either the Brunssumerheide natural reserve (Laban, 2007) or urban built areas. The annual production in the Netherlands is currently about 2.4 million tonnes.

## Geological context of the silver sands

Fossils that might indicate a specific depositional environment have not been found in the silver sands, but some ghost structures of shells occur. They have been interpreted by De Jong & Van der Waals (1971) as indicative of a shallow-marine, near-coast environment, but the precise conditions (beach, tidal flat, estuary, full-marine) could not be determined on the basis of these ghost structures. Almost all sand layers consist of grains that are well rounded and that are well sorted. This combination of extreme sorting and rounding is known from marine to coastal sediments only from a beach environment. This interpretation is supported by the (rare) occurrence of layers with well rounded flint pebbles (Fig. 6); the pebbles cannot have become so well rounded and sorted by transport under fluvial (braided, anastomosing or meandering) conditions, because no indications for such fluvial activity have been found in the silver-sand area itself or the nearby region. It is therefore most likely that the flint pebbles have been eroded by wave activity from Cretaceous rocks, which are present in the neighbourhood. They may have been transported by longshore currents, but if such currents were responsible, indeed, it is not clear why only so few layers (or strings) of flint pebbles are present, and why they have such limited extents within the otherwise uniform silver sands. The pebbles must have become well rounded in the surf zone along (and on) the beach. A beach environment is also indicated by the – scarce – occurrence of heavy-mineral laminae (Fig. 7). The occurrence of these laminae is, however, simultaneously enigmatic: why are concentrations of heavy minerals present in a sand unit that has been leached to such an exceptional degree? And why were most of the heavy minerals in these laminae less affected by weathering than their counterparts elsewhere in the silver sands?

*Fig. 6. Strings of well-rounded flint pebbles in the Nivelstein sandpit. Pen is ~14 cm long.*

*Fig. 7. Heavy-mineral laminae in incompletely leached silver sands of the now abandoned Oranje Nassau IV sandpit near Heerlen. Diameter of coin is 1 cm.*

It should be mentioned here that Gullentops (1972/1973) came to a somewhat different environmental analysis for the Opgrimbie Sands, which also include well rounded, well sorted flint pebbles. On the basis of grain-size analysis, he came to the conclusion that the sands had been deposited under tidal, estuarine conditions, but that the transport of the pebbles had essentially been fluviatile. Grain-size analysis is, however, nowadays no longer considered as a reliable tool to interpret transport or depositional environments. Moreover, if the flint pebbles would have undergone fluvial transport, the sands must also have undergone this type of transport and – as mentioned above – no unambiguous indications have been found for this; in contrast, indications for a near-coast environment abound (for instance in the form of herringbone structures indicating tidal conditions), which supports the interpretation that the pebbles owe their roundness and sorting to the presence for some time in a beach-like environment.

The majority of the sediments building the silver sands can, however, not have been deposited in a beach environment. Abundant small channels and ripple cross-lamination (sometimes forming sets of more than half a metre thick: Fig. 8) indicate strong and frequent current action; herringbone structures (Fig. 9) found at numerous places indicate tidal influence. The combination of beach and tidal deposits thus indicates a shallow coastal depositional environment. It should

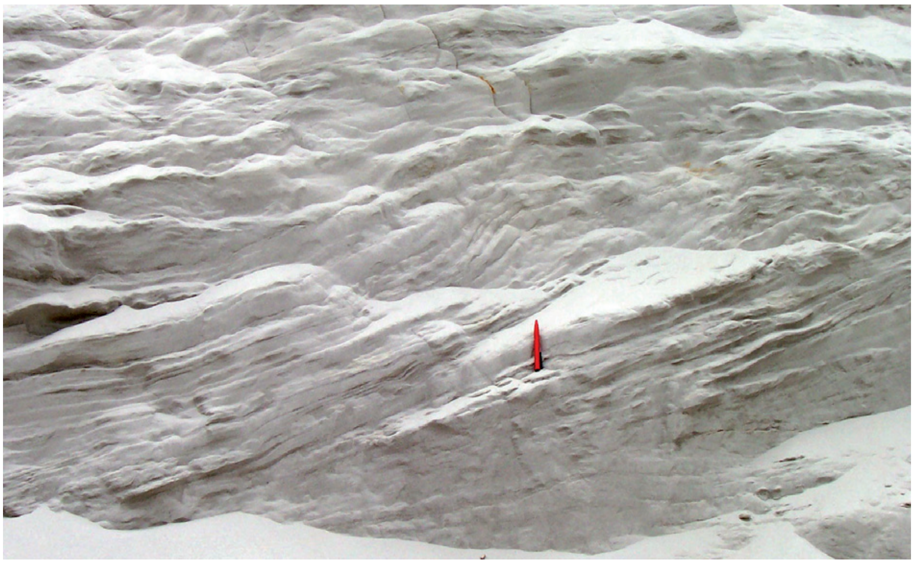


*Fig. 8. Thick cross-bedded layer in the Sibelco quarry, indicating a high-energy environment. Pen is ~11 cm long.*

*Fig. 9. Herringbone structure indicating tidal influence. Beaujean-West quarry. Pen is ~13 cm long.*

be mentioned in this context that the levels that abound in channels and ripple cross-lamination consist of sand that is equally well sorted and rounded as levels without such structures. It may therefore well be that the sorting and rounding have originated in a beach environment, but that a later deepening of the sea took place, so that the sands are, in whole or in part, in fact shallow-marine – at least partially tidal – sediments with grain characteristics that have been inherited from earlier presence along a pebbly beach.

A fairly detailed picture exists of the palaeogeographic development in the Dutch/German/Belgian border area for the Miocene. During the late Early and Middle Miocene, when the climate was subtropical and sea level reached a maximum, the shoreline ran more or less over this area, and thick sand units were deposited. The shoreline moved gradually to the NW and the area changed into a generally marshy environment with dense vegetation (amongst other plants, *Sequoia* trees). A succession of transgressions and regressions followed, resulting in a laterally extensive wedge-shaped contact between shallow-marine deposits and peat layers that form the westernmost parts of the Frimmersdorf browncoal seam (Fig. 2); the latter must have reached a total thickness of some 300 m. The peat became later compacted due to the weight of younger deposits and coalification started, turning the organic material eventually into browncoal (Fig. 10). Several browncoal layers (of up to 10 m thick), three of which occur in Southern Limburg, can be distinguished in the Dutch part of the area. In the German part, which was not reached by the transgressions, the browncoal reaches a thickness of about 100 m.

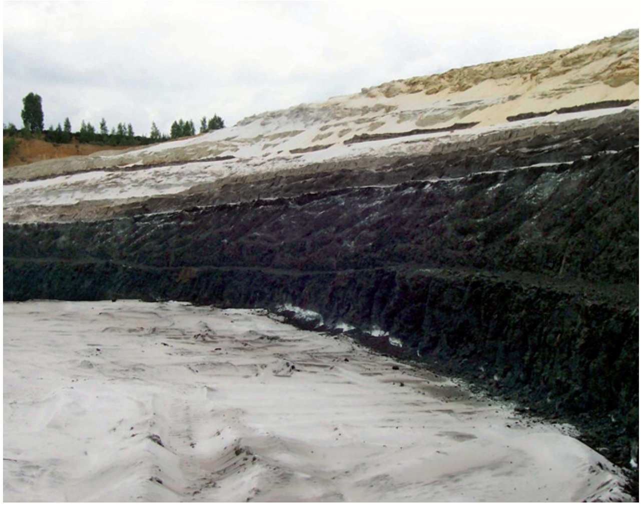
The sandy deposits under the browncoal layers must have become gradually leached by infiltrating rainwater full of humic acids, derived from the browncoal, as indicated by infiltrated organic material in the silver sands just under browncoal accumulations. The iron-oxide/hydroxide coatings around the sand grains became dissolved, and ongoing leaching resulted in the almost complete disappearance of the originally present minerals, except quartz.

## Major questions

Questions that have not yet been answered satisfactorily regard both theoretical and practical aspects of the silver sands. Theoretical questions are (1) whether the weathering-related characteristics of the heavy-mineral content (exceptionally low percentage; strongly weathered grains; species-poor assemblages) result from processes at the present depositional site or whether they have been inherited from an older (eroded) sedimentary unit, (2) what caused the extreme degree of leaching and (3) why the leaching reached such a considerable depth. Practical questions include both large-scale and small-scale aspects. Large-scale aspects include the question of (4) why leaching appears to have stopped suddenly at specific levels and (5) where the leached material has gone; a small-scale problem is (6) why the extreme chemical weathering did not affect all specimens of a specific mineral species equally.

### *Where did the weathering take place?*

The extreme quartz-rich nature of the silver sands cannot be explained as a result of supply from a normal source rock. It is, principally, possible however, that the source rock was a deeply weathered, old peneplain, so that the present-day characteristics of the sand should be considered as inherited. This hypothesis was put forward for the Opgrimbie Sands by



a.

b.

*Fig. 10. Browncoal layer in the Sibelco quarry. a. Position of the layer (approx. 1.90 m thick) intercalated between silver sands; b. Detail of the browncoal layer with a tree stem in living position. Pen is ~13 cm long.*

Gullentops (1963, 1972/1973), who also stated that perhaps part of the characteristics (weathering phenomena and uniform grain size) might have been obtained during fluvial transport. A third possibility is, obviously, severe leaching *in situ*.

Several reasons make it highly unlikely that the characteristics of the silver sand were inherited from a source area with material that shows comparable characteristics, although Van der Meulen et al. (2009) state that the material "probably originated from the well-weathered and therefore quartz-rich material that was available at the margins of the southern North Sea Basin in Belgium and adjacent areas". In fact, two types of sources might be considered, viz. (1) the above-mentioned deeply weathered peneplain of which the various sedimentary units originally had different characteristics, and (2) a sedimentary unit that was affected by weathering so severely that a deposit with silver-sand characteristics had formed.

If a peneplain or an older 'silver sand' was the source of the silver sands in the Dutch/German/Belgian border area, the source must have been eroded, and the eroded material must have been transported to its present-day depositional site. Both options are highly unlikely. A peneplain as a source is unlikely because no nearby peneplain is known that shows indications of deep weathering in the form of leached out sands. Moreover, hardly any erosion takes place on a peneplain, unless it becomes tilted or uplifted but no tectonics-affected peneplain with such characteristics is known from the neighbourhood either, and there is no indication that such a peneplain ever existed without leaving any trace. An older 'silver sand' deposit as a source area is equally unlikely, because it is difficult to imagine that it has completely disappeared without leaving a trace.

There are other, possibly even more convincing, arguments against a source rock from which the silver sands have inherited their characteristics. The present-day silver sands can have inherited the extreme characteristics from a 'parent' unit only if the material has been transported from the source area to the depositional area. Such a transport should have left traces, either in the form of a sediment deposited by the transporting medium, or in the form of an admixture in the present-day silver sands derived from erosion by the transporting medium of the sediments that were passed. No indications for a medium transporting severely weathered material have been found, however, nor do the sediments show admixtures of other sediments, for instance in the form of clay particles or coated grains.

The chance that the lack of both a possible source rock or source area and a transporting medium is a coincident, must be considered as practically nil, so that the option of inherited characteristics must be rejected.

The second option (severe weathering, rounding and sorting during transport) is not supported by geological evidence either. Admittedly, rounding and sorting increase as a rule with transport distance, but this is a gradual process, and no evidence of a gradually increasing degree of rounding or sorting in a specific direction has been found. Considering the fact that several indications exist, as detailed above, for deposition in a beach or other wave-affected near-shore environment, sorting and rounding must have occurred at (or nearby) the depositional site; there is no indication that the grains had already been sorted and rounded before arrival.

Weathering during transport, reflected in the form of a sediment that becomes compositionally more mature in a down-current direction, is well possible. The extreme maturity that is shown by the silver sands in the Dutch/German/Belgian border area can, however, be obtained only during a long-lasting stay at a location where severe weathering takes place. No examples are known of sediments that become so extremely weathered while covered by a marine, fluvial or lacustrine water mass; only a long stay at the bottom of an acid lake might result in this type of leaching, but truly acid lakes require hydrothermal or volcanic activity, and no such activity is known from this area during the Paleogene and Neogene. Extreme weathering would thus require deposition at a site where long-lasting, extreme leaching would affect the sediment before erosion would start again, transporting the leached sediments to their present site. Just like in the case of the peneplain hypothesized by Gullentops (1963, 1972/1973) there is, however, no geological evidence for a location where the sediments might have stayed for a long time, becoming severely leached, and removed again without leaving any trace.

Finally, it is difficult to imagine that the silver-sand deposits have been transported after weathering (in a source area or during an interruption of their transport history) because of the habit (shape) characteristics of the heavy minerals. It was found during a detailed investigation of heavy minerals from various sections that numerous heavy-mineral grains were severely affected by weathering, showing very fragile forms (Fig. 11) that would survive neither long transport by a current (the grains are too large for transport as suspended load), nor water movement in a wave-dominated environment.

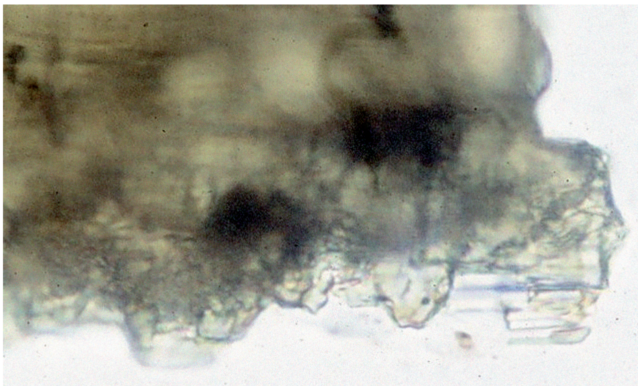


*Fig. 11. Detail of a strongly corroded tourmaline grain (Beaujean quarry) with fragile parts than cannot survive wave action or transport by flowing water. The part shown is ~60 µm long.*

On the basis of all the above arguments, it must be concluded that the silver sands cannot have come from elsewhere in the weathered state that they show now, but that the extreme weathering must have taken place *in situ* at their present site.

## What processes were responsible for the leaching?

Leaching of sands is common; extreme leaching removing almost everything but quartz is rare; the extreme leaching resulting in the Dutch/German/Belgian silver sands, bleaching them over a depth of many dozens of metres, has few equivalents. The two only processes that are known to be able to result in such an extreme weathering are long-lasting exposure to infiltration of rainwater rich in acids (e.g. humic acids), and long-lasting surficial exposure to weathering in a humid tropical or subtropical climate. Neither of these two options is, however, fully satisfactory in itself in the case of these silver sands.

The first option, i.e. leaching due to infiltrating rainwater that percolated though the overlying browncoal layers, must have played a role, indeed, as shown by humic material that has infiltrated the silver sands just below the contact with overlying browncoal (Fig. 12). It cannot explain the silver-sand architecture, however, because the thickness of the silver sands shows significant lateral variations. Moreover, silver sands occur regionally where browncoal layers are absent, and at some of these places browncoal most probably has not been present originally either. This implies that the leaching of the coastal sands that now constitute the silver sands cannot everywhere be ascribed to the downward infiltration of rainwater rich in humic acids. This conclusion is supported by the fact that the Dutch silver sands pass eastwards into sands that are intercalated between two browncoal seams, but that have nevertheless remained their original character, not having turned into silver sands. Moreover, no silver sands occur in

*Fig. 12. Infiltrated organic material in silver sand just below a browncoal layer in the Sigrano quarry. Knife is ~17 cm long.*

Germany under the Garzweiler browncoal seam (Manten, 1958), which seam is not present in Southern Limburg.

The alternative option, viz. leaching by (sub)tropical weathering, is not satisfactory either because no present-day equivalent is known of silver sands formed to such a depth in the tropics or subtropics. Leaching is a common process in these climate zones, and lateritic soils may reach great thicknesses in the tropics, but lateritization differs fundamentally from the transformation of coastal sands into practically pure quartz sands. A phenomenon that points at other processes than mere weathering is that the lower boundary of the silver sands tends to be sharp. This is contradictory to the practice of (pedogenesis-related) chemical weathering, in particular in the case of deep weathering.

It must thus be deduced that an answer to the question of what process was (or which processes were) responsible for the deep leaching cannot be answered on the basis of 'normal' leaching under 'normal' conditions.

The extreme leaching – that must have occurred *in situ*, as shown in the previous section – can have taken place only if the sediments were exposed sufficiently long to conditions that allowed severe chemical weathering. No indications for deep pedogenesis have been found in these shallow-marine to coastal sediments, although the botanical characteristics of the browncoal seams that are regionally found on top of the silver sands prove that the marine environment changed into a coastal marsh environment. The regression apparently occurred so slowly that the continuous subsidence of the North Sea Basin kept more or less pace with the accumulation of organic material, so that thick successions (of up to 300 m peat in Germany, later compacted to some 100 m browncoal) of organic material could accumulate without significant changes of the sedimentary surface with respect to the sea level.

### *Why was such a thick succession affected by leaching?*

The accumulation of hundreds of metres of organic material must have taken considerable time. Considering the humid tropical climate, huge amounts of rain water must have infiltrated the succession that initially must have formed a peat layer. During the infiltration, large amounts of humic acids from the peat must have been transported downwards by the meteoric water. As this process must have gone on for a long time, this led eventually to the severe leaching of the sands to a depth of, locally, up to 90 m.

### *Why is the lower boundary of the silver sands sometimes sharp?*

Downward movement of the meteoric water rich in humic acids will, in principle, have continued until the real groundwater table or a hanging groundwater table (due to an impermeable layer) was reached. No evidence in favour of either possibility has been found, but in the Netherlands a commonly sharp boundary with the underlying Kakert Member (Breda Formation) exists. This member consist of fine-grained, slightly glauconitic – and therefore yellowish-greenish; in weathered condition greyish-brownish because of grains coated by iron (hydr)oxides – shallow-marine sands: Westerhoff et al., 2003). Apparently the boundary between the two members prohibited further downward infiltration of the acids-rich meteoric water.

The contact between the severely bleached and the underlying unbleached sands is remarkable for two reasons: (1) the boundary is commonly sharp but sometimes gradual, and (2) where the boundary is sharp, no features occur that explain why infiltration towards deeper levels was impossible. Downward infiltration of rain water full of dissolved material commonly results in the precipitation of, among others, iron oxides and/or hydroxides – however limited in quantity – if an impermeable level is met, but no such precipitates have thus far been found in the Netherlands. This implies that either the water must have penetrated the underlying sands (but did not – for some enigmatic reason – continue its leaching activity) or has bent off into a more or less horizontal direction. The lower part of the silver sand does, however, not act as the basis of an aquifer nowadays, nor does the granulometry or permeability of the underlying sediment suggest that the sediments of the Kakert Member are impermeable.

It must be mentioned in this context, however, that an impermeable, hard layer is said to be present underneath the Opgrimbie sands in the Belgian Sibelco quarry. This layer is – for a reason that has not been explained as yet – said to be rich in marcasite, $FeS_2$ (pers. comm. Mr Hilven, plant manager, 2009). According to the present-day geologist of the quarry (P. Demechelaar), several zones with marcasite agglomerates have been present in the past, but without a clear stratigraphic distribution; one of these zones is now exposed in a corner of the quarry. This has remained unnoticed thus far in the literature; even in the explanatory text to the geological map that includes the Opgrimbie Sands (Buffel et al., 2001). The apparently random stratigraphic distribution of the marcasite-rich zones can, obviously, not explain the commonly sharp boundary between the silver sands and the underlying sands.

It must consequently be concluded that the boundary between these units is still enigmatic. A possible solution for this enigma is presented in the 'Discussion' section.

### *Where did the leached-out material go?*

The total volume of the silver sands, only in the Dutch part of the border area, amounts to several hundreds of millions of cubic metres at least. This implies that probably tens of millions of cubic metres – but at least many millions of cubic metres – of material (almost all mineral grains except quartz, but also mineral coatings, and probably part of the quartz grains, too) must have been removed from the sediments that now form the silver sands. Considering the thickness of the silver sands, it is not likely that the leached-out material was removed by a more or less horizontal groundwater flow that was active throughout the succession. In addition, the uniform granulometry of the sands (in most levels, the great majority of the grains are in the 150-260 µm range), resulting in a comparatively low porosity, makes behaviour as a thick aquifer unlikely. It must therefore be assumed that the dissolved material moved predominantly downwards, in the infiltrating meteoric water.

It is obvious from the unweathered state of the underlying sediments, however, that the water that infiltrated what are now the silver sands, did not proceed downwards into the underlying material. As infiltration will have gone on in the then humid climate, the downwards infiltrating water must have chosen another direction as soon as the boundary with the underlying material was reached. This can be explained only by a diversion of the pathway of the meteoric water from roughly vertical to roughly horizontal at the contact plane between the two units.

An (almost) horizontal flow direction over a then existing (but now no longer present: see the 'Discussion' section) impermeable horizon is not unlikely, considering the fact that the silver-sand belt is situated in a tectonically active region, and some tilting has occurred. The precise tectonic architecture during the Miocene is not known, but it is most likely that drainage then occurred either to the river valley in the East or the sea in the West. This implies that the dissolved constituents were ultimately drained into the river or the sea, which explains the lack of precipitates (particularly iron hydroxides and/or oxides) at the contact between the silver sands and the underlying sands.

## *Why did not all minerals suffer equally from the weathering?*

The silver sands consist almost exclusively of well rounded quartz grains. This mineral is hardly susceptible to leaching by humic acids, and the almost perfect rounding also decreases the susceptibility. It cannot be fully excluded, however, that some part (it cannot be estimated which percentage) of the quartz grains has also been dissolved, as many of the grains of other mineral species show features that evidence strong chemical weathering.

It has been mentioned already that the heavy-mineral content of the silver sands is exceptionally low, but heavy-mineral analyses have nevertheless been carried out. This was done for Dutch silver sands by Muller (1943), De Jong & Van der Waals (1971), Van Loon (1972/1973) and Van Loon & Mange (2007), who all found very poor assemblages (see Fig. 5). The heavy-mineral content of the Belgian Opgrimbie Sands was investigated by Gullentops (1972/1973). He also found a very poor assemblage, with some 70% of zircon and some 20% of tourmaline.

Heavy-mineral analyses used for a long time to be restricted to the counting of percentages of the heavy-mineral species. Van Loon (1972/1973), however, also investigated their shape, which was, obviously, determined by two processes: transport and *in situ* weathering. Only few mineral species occur in sufficient quantities to allow the latter type of research in a statistically reliable way, but garnet, staurolite and tourmaline are present in sufficient quantity to perform such an analysis, and they also appear to be sufficiently susceptible to extreme leaching conditions to show different degrees of chemical weathering.

It is well known that the resistance of the various mineral species against chemical weathering depends on a number of factors (which are still badly known). This is due to the fact that the conditions under which minerals have been chemically affected, are rarely known precisely (cf. Velbel, 2007; Bateman & Catt, 2007; Morton & Halsworth, 2007). In addition, the relative resistance of heavy minerals against leaching has commonly been tested under laboratory conditions, which may differ greatly from nature. Wherever resistance to chemical weathering was determined for mineral species from field samples, this commonly occurred by counting the relative percentages of the individual species, and comparing the results with those from non-affected samples from (presumably!) the same source area.

Van Loon was probably the first to investigate the effects of leaching on different heavy-mineral species within field samples by means of studying the characteristics of individual grains. He did so by establishing two or more classes of affected grains (e.g., not affected, slightly affected, strongly affected) on the basis of the grains' shapes. When he applied this method to the silver sands of the Dutch/German/Belgian border area (Van Loon, 1972/1973), he found that entirely fresh grains coexist in one single sample with extremely weathered grains of the same heavy-mineral species (Fig. 13). Van Loon (1972/1973) and Van Loon & Mange (2007) concluded that the degree of chemical weathering is a statistical value rather than a property shared by all similar grains. Why entirely fresh mineral grains of species that are considered to have a relatively low resistance to chemical weathering (such as garnet and staurolite) can be present in extremely leached-out sediments such as silver sands, has remained an enigma for a long time. The unquestionable severe dissolution makes it only more enigmatic that many samples contain heavy-mineral specimens, also of species that are relatively easily weathered, that are still completely fresh. Relatively fresh grains are found also in samples that contain strongly affected specimens as well. In some cases, such as tourmaline varieties, this might be explained by differences in the susceptibility to chemical dissolution under the influence of humic acids, but this explanation cannot explain the considerable differences in weathering of, for instance, the staurolite and garnet grains (see Van Loon, 1972/1973). The apparent 'statistical' value of the weathering (Van Loon & Mange, 2007) can be explained only if some differential weathering took place.

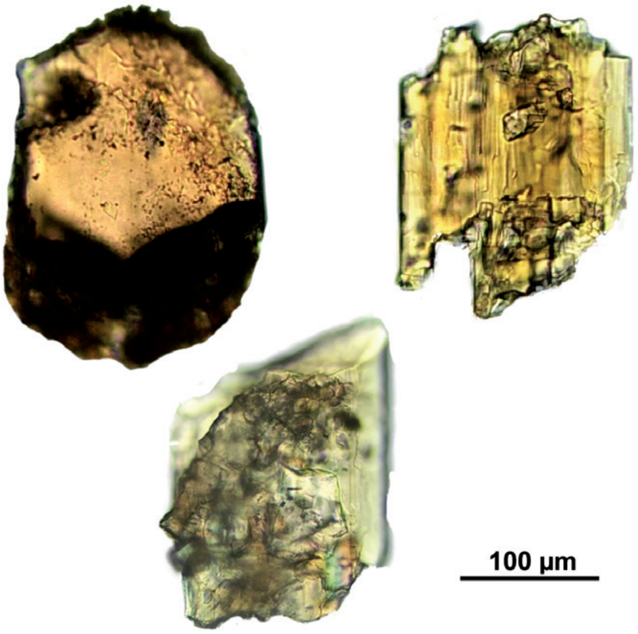


*Fig. 13. Tourmaline grains (from a single field sample) showing slight, intermediate and severe chemical weathering.*

It is in this context of great importance to mention that the extremely low percentage of heavy minerals in the silver sands made it necessary to take large field samples. Van Loon & Mange (2007) calculated that a reliable counting of the heavy minerals required field samples of about 3 kg, and that – if specific characteristics were to be analysed (e.g. the degree of weathering of a specific mineral species) – field samples of 10-30 kg were required. This implies that, as a rule, a single field sample contained grains from a vertical interval of certainly 10 cm. The importance of this uncommon sampling method may help to explain the differential weathering, as will be detailed in the 'Discussion' section.

## Discussion

The silver sands under study show some uncommon characteristics that inevitably lead to uncommon explanations. In the above sections, for instance, it was argued that the extreme leaching of coastal sands by humic acids from overlying browncoal layers, in combination with the locally sharp boundaries between these leached sands and the underlying sands, can be explained satisfactorily only if the direction of the initially downward infiltration of acid-rich rain water in the sands under study was bent into a more or less horizontal direction at the contact plane with the underlying sands. Such a change from vertical to horizontal might also explain the enigma why silver sands are found at places where no browncoal cover is present, and where it has probably not been present in the past either: the acids-containing groundwater bleached the sands that it passed during the more or less horizontal flow, irrespective of the occurrence of browncoal at a higher level. The bleaching could continue during the flow until the acids that were carried along were no longer sufficient to bring iron (hydr)oxide coatings into solution; with a diminishing acid character of the flow, the weathering of the heavy minerals, obviously, also gradually stopped.

Another enigma can also be solved by considering the water movement: the boundary between the silver sands and the underlying sands is not sharp everywhere; in several places it is irregular and gradual, and it may be even impossible to find in the field a true boundary between the coastal silver sands and the shallow-marine sands underneath in which many grains are coated with iron (hydr)oxides, thus being responsible for the colour of these sediments. This is exactly the type of contact that is to be expected in the case of deep weathering. Apparently the infiltrating rain water was not forced everywhere to bend into a horizontal direction at the contact plane between the two units. This implies that the contact plane must have had laterally different characteristics, even though such differences are not found any more.

It is likely that the two units gradually pass into each other with regard to their depositional environment (probably from full – though shallow – marine to coastal) as, apart from the post-depositional bleaching, only a small change in sorting can be found: the underlying sands are somewhat less sorted (and therefore a bit less permeable). It is therefore difficult to imagine that the boundary between these two units acted as a sedimentologically and hydrologically significant boundary from the very beginning. Apart from the difference in leaching and, to a much lesser degree, sorting it is not a significant boundary now either. This implies that a process must have acted that changed the characteristics of the boundary temporarily (and not everywhere). Few early-diagenetic processes are capable of doing so, and the only one that may provide a feasible explanation for this feature is temporary and local cementation.

The stratigraphic unit of the silver sands has undergone cementation, indeed, as shown by the use of cemented blocks as building stones. Although the cementation in the German part of the area is locally strong (Nivelstein Sandstone), the cementation is only slight in the Dutch and Belgian area: the cemented material can commonly be easily fragmented between the fingers in the case of thin cemented horizons, and it can easily be fragmented with a hammer (or even bare hands) in the case of thick cemented horizons. The fact that the Heksenberg Member now regionally (and locally) consists of sandstone, particularly between the Morken and Frimmersdorf browncoal seams, whereas the sands are not cemented elsewhere, is proof of differential cementation (Fig. 3). The partial cementation may be a primary diagenetic feature, but it might just as well be a result of partial loss of the cement after an earlier, more complete degree of cementation. It is well known that changing conditions in pH may cause alternating phases of cementation and dissolution of cement, although the latter is not so common as the former.

Differential cementation, in combination with later local dissolution of cement, can well explain most of the features that made the silver sands so enigmatic with regard to their properties such as the locally sharp boundary with the underlying Kakert sands, and the joint occurrence of heavily weathered and 'fresh' heavy minerals.

The sharp boundary between the silver sands (Heksenberg Member) and the underlying brownish sands (Kakert Member) can be explained by a locally cemented level (however slight the cementation may have been) that thus had become impermeable. It is not remarkable that cementation started just at this level, as the lower permeability of the Kakert Member must have reduced the flow velocity of the infiltrating water, which facilitated the preservation of gradually forming precipitates. The impermeable boundary forced – as argued above – the flow of infiltrated rainwater to change its direction from roughly vertical to roughly horizontal, parallel to the contact plane. The fact that this contact plane, as far as accessible, is not cemented anymore nowadays, does not contradict this hypothesis, as the cementation must have been only slight, so that the cement may have disappeared later (possibly due to ongoing supply of rainwater rich in humic acids, but under changed pH conditions), particularly if the cement was still incomplete and composed of small crystals that were more susceptible to weathering than the primary (sedimentary) quartz grains.

The local cementation must have taken place not only at a relatively large scale (causing the impermeable boundary layer between the Heksenberg and Kakert Members of the Breda Formation), but also at a small scale. This is still visible in the various present-day outcrops, where thin, discontinuous bands of slightly cemented silver sand occur (Fig. 14). These cemented bands have often a thickness of only one to a few millimetres, and they have lateral extents of a few centimetres to a few metres. If such slightly cemented bands existed during the time that the infiltrated rainwater was forced to bend off to a more or less horizontal direction, the flow will have taken pathways under and above the cemented horizons. The heavy minerals in these cemented horizons thus were protected against the acids, and were affected only slightly or not at all, in contrast to their counterparts in the non-cemented parts. This explains why samples from the silver sands, particularly if they were large in order to contain sufficient heavy-mineral grains for analysis of their characteristics, can contain both severely weathered, slightly weathered and non-weathered specimens of the same mineral species, thus resulting in a 'statistical' value for the degree of weathering. This 'statistical' value thus represents a proxy for the degree of cementation rather than for the amount of infiltrating humic acids.

## Conclusions

The silver sands in the Dutch/German/Belgian border area show features that have thus far been difficult to explain, and that also seemed unrelated to each other. It turns out, however, that all these features can be explained by two processes: (1) severe leaching by infiltrating rain water that was rich in humic acids from the overlying browncoal, and that was locally forced to change its mainly vertical direction into a roughly horizontal direction because of an impermeable level, and (2) changing pH conditions that resulted in local and temporary cementation and dissolution of cement, thus producing local and partly temporal levels that controlled the flow of the infiltrated water and that also locally protected sediments from the leaching that affected the great majority of the grains.

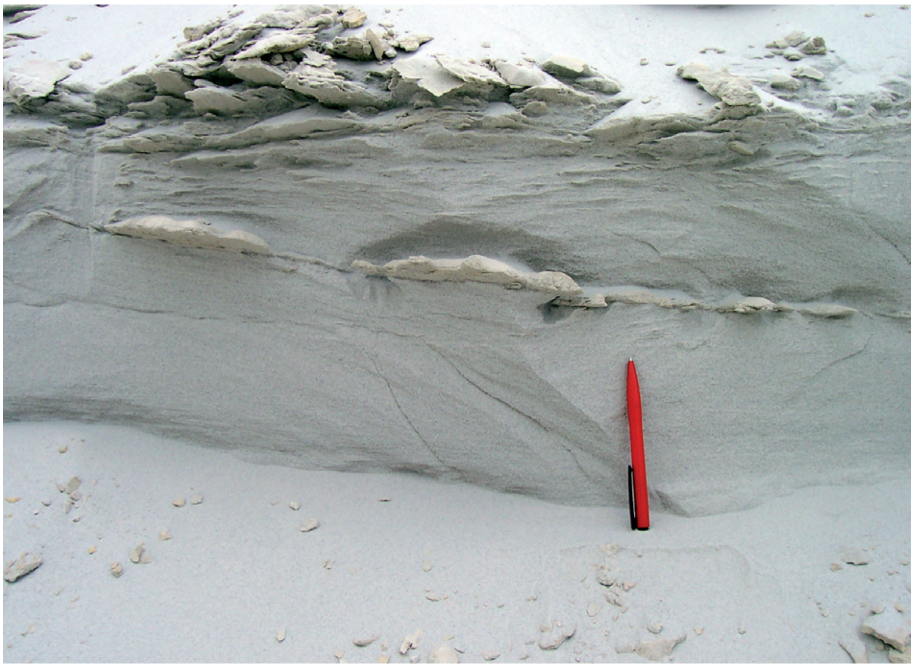


*Fig. 14. Differentially cemented silver sands, probably due to local dissolution of cement, resulting in thin, discontinuous impermeable levels that influence the flow pattern of groundwater. Pen is ~13 cm long.*

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