Assessment of the state of conservation of a Middle Neolithic flint mine in Maastricht limestone

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Upper Cretaceous Maastricht limestone ('mergel') outcrops in the provinces of Dutch and Belgian Limburg. The Upper Cretaceous in the Netherlands consists of the geological Maastricht Formation and the upper part of the Gulpen Formation. Limestones from the Maastricht Formation represent one of the few native Dutch natural stones used for building and construction. Locally, limestone from both formations contains considerable amounts of flint. This flint has been mined in Neolithic times, both from the Lanaye limestone in the Gulpen Formation and the Emael Limestone in the Maastricht Formation. Around the village of Valkenburg aan de Geul, flint was mined from the latter. In the current study, the state of conservation of a Middle Neolithic flint mine situated at the Plenkertweg in Valkenburg aan de Geul is assessed, 8 years after the site was discovered and exposed.

Key words: Maastricht limestone, mergel, flint mining, ancient mine, Neolithic, conservation, Valkenburg

1 Introduction

Maastricht limestone, a soft, very porous limestone outcropping in the Dutch and Belgian provinces of Limburg, has regionally been used as a dimension stone since at least Roman times. Even earlier, since Neolithic times, flint has been mined from this limestone, amongst others around the village of Valkenburg aan de Geul, Dutch Limburg. In 1992, a Neolithic flint mine was discovered along the Plenkertweg in the village of Valkenburg itself (Brounen et al. 1993; Fig. 1). The current paper relates the assessment of the state of conservation of this flint mine by a combination of determination of hygric characteristics and a polarized light microscopy (PFM) and electron microscopy study of the limestone.

Figure 1. Overview of the site of the Neolithic flint mine at the Plenkertweg, Valkenburg (Apr. 2001). Numbers (1 to 4) indicate sampling localities.

2 Maastricht limestone: geology, use as building stone, flint mining

Surface geology of the southernmost part of the southern Dutch province of Limburg and adjacent parts of Belgian Limburg is dominated by Upper Cretaceous deposits, in particular limestones of the Maastricht Formation, deposited in the geological epoch of the same name, Maastrichtian (65.5 – 70.6 Ma). This formation, which overlies limestones of the Gulpen Formation resting on sands of the Vaals Formation, comprises six limestone series, the Meerssen and Nekum limestones in the upper Maastricht Formation and the Emael, Schiepersberg, Gronsveld and Valkenburg limestones in the lower Maastricht Formation (Felder 1975, Felder & Bosch 2000; table 1). Total thickness varies from c. 45 to c. 90 meters. All limestones have been developed in two depositional facies, viz. The Maastricht and the Kunrade facies. The Maastricht facies comprises soft, fine to (very) coarse yellow white limestones, the Kunrader facies light grey limestones that are generally harder (Felder & Bosch 2000). Limestones from the Maastricht facies are locally known as ’mergel’ (litterally but erroneously: marl), ‘tuf’, ‘tufkrijt’ or ‘tuffeau de Maestricht’. More appropriate, they can be denoted as Maastricht limestone. They have been used as building stone since Roman times in both Dutch and Belgian Limburg (Keuller et al. 1910, Engelen 1972, 1975, Bosch 1989, Felder & Bosch 2000, Dreesen et al. 2001, Dreesen & Dusar 2004, Dubelaar et al. 2006; Fig. 2), as well as in the Belgian city of Liège (Fig. 3). Traditionally, various Maastricht limestones used as dimension stone have been denominated as Sibbe, Roosburg, Zichen (Sichen) and Kanne block (Keuller et al. 1910, Dreesen et al. 2001, Dubelaar et al. 2006). Though the extent of Roman use is disputed and considered to have been rather limited (Silvertant 2002), Romans already exported the stone to other areas, for example to be used in the construction of the castellum walls of Utrecht, in the centre of the Netherlands (De Groot 1994, Rijntjes 1994). In medieval times, apart from its regional importance, small amounts of Maastricht limestone were used in several towns in the central Netherlands (Slinger et al. 1980, Nijland et al. 2007).

Figure 2. Ruin of Valkenburg castle (12th-17th century), with the remains of the surrounding wall in Maastricht limestone on top of that limestone itself. Decay of both the wall and the rock is clearly visible (2001)

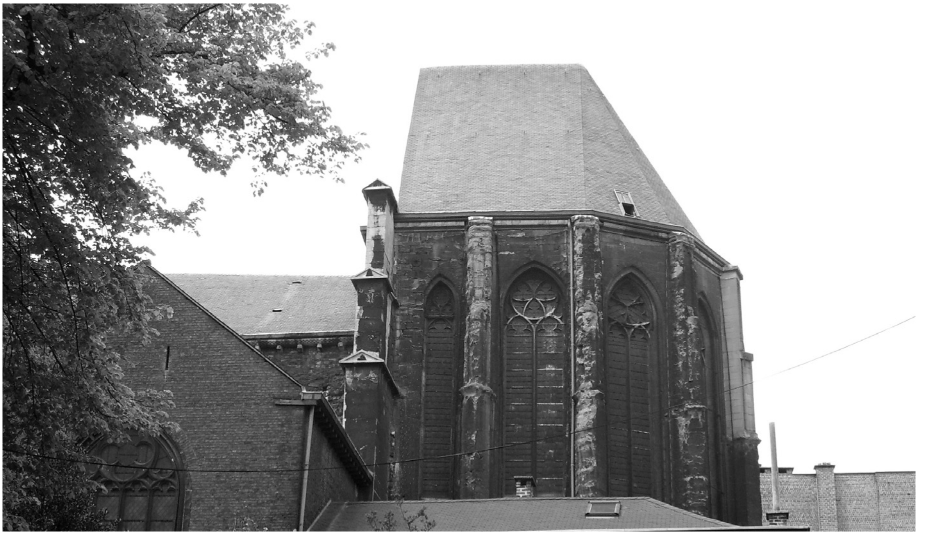

Figure 3. Choir of St. Denis church in Liège, constructed in Maastricht limestone (2008)

Maastricht limestones are soft, very porous (and hence light), very pure fossiliferous limestones, with only very small Mg and Si contents (Table 1). Porosity may reach over 40 vol.%, mainly intergranular, with intragranular porosity in bioclasts and fossil fragments. Grains are poorly cemented only, without siginificant compaction or pressure solution effects. Compressive strengths are generally low (3 – 5 N mm-2), though harder varieties may have compressive strengths up to c. 35 N mm-2 (Dubelaar et al. 2006)

Limestones, both of the Maastricht and Gulpen formations, may contain significant amounts of chert (flint) (Table 1). Locally, chert (flint) has been used as a building stone on a limited scale for farmhouses, etc., both in Dutch Limburg and the Belgian Voerstreek. Flint from the Cretaceous limestones in Limburg has been mined in Neolithic times for tool production. An overview of known prehistoric flint mining sites in southern Limburg and adjacent areas is given by Felder (1998). Two important production centres existed at Rijckholt – St. Geertruid and around the current village of Valkenburg aan de Geul. At Rijckholt – St. Geertruid tens of ancient underground mines have been excavated, results of which are documented in Rademakers (1998a). 14C dating of charcoal and bone artefacts show mining at 3700 – 4000 BC (Rademakers 1998b). The flint layer exploited is situated in the Lanaye limestone of the Gulpen formation (Felder & Bosch 2000). The Valkenburg production centre comprises mines at Valkenburg itself and surrounding villages, with 14C ages in the range of 2500 – 3630 BC (Rademakers 1998b). Products of Valkenburg flint have been traced at least 50 km into Germany (Löhr 1976). Flint was mined from the Emael limestone of the Maastricht formation (Felder 1976, 1998). In more recent times, flint has, on a very limited scale, been used as a dimension stone, generally as isolated blocks, but locally for walls of farms and an isolated chapel (Nijland et al. 2006).

The flint mine investigated in the current paper is situated along the Plenkertweg in the village of Valkenburg aan de Geul, and was discovered in 1992 (Brounen et al. 1993; Fig. 1). Mining was performed along shafts (Fig. 4); traces of stone cutting are clearly recognizable in the mine (Fig. 5). Mining was done from shafts. 14C dating of charcoal from two mine shafts shows ages in the range 3050 – 3632 BC (Rademakers 1998b). Flint was obtained from the Emael limestone. This limestone from the Maastricht formation is a typical soft, light yellow Maastricht limestone with large light grey flint nodules in its lower part. The Emael limestone is separated from the Schiepersberg limestone at its base by a hardground called Romontbos horizon, and from the overlying Nekum limestone by the Laumont horizon. Flint nodules often form well developed pipes and plates, that may be easily be separated from the limestone (Felder & Bosch 2000).

Figure 4. Impression of one of the ancient corridors at the Plenkertweg flint mine (June 2001); this part of the outcrop corresponds to sampling locality 3 on figure 1.

Table 1. Lithostratigraphy and lithological characteristics of the Upper Cretaceous in Limburg

Chrono-stratigraphy		Lithostratigraphy		Thickness m		
CRETACEOUS	Maastrichtian	Maastricht Formation	Meerssen limestone		ca.	18
			Nekum limestone	9	-	15
			Emael limestone	2	-	10
			Schiepersberg limestone	4	-	6
			Gronsveld limestone	4	-	10
			Valkenburg limestone	3	-	45
		Gulpen Formation	Lanaye limestone	15	-	18
			Lixhe 3 limestone	9	-	11
			Lixhe 2 limestone	8	-	12
			Lixhe 1 limestone	5	-	10
	Campanian		Vylen limestone	0	-	>100
			Beutenaken limestone	0	-	12
			Zeven Wegen limestone	0	-	30

Notes on Table 1:

- For thickness, density and flint contents ranges I composition (minimum – maximum) are given, in case of density excluding any flint (i.e. the limestone itself only).
- The Meersum and Lanaye limestones contain several harder banks with higher densities, 1.50 – 2.40 g/cm³ and 1.80 – 2.50 g/cm³, respectively.
- Density of the Gronsveld limestone shows a distinct regional east-west variation, with 1.21 – 1.40 g/cm³ in the west and 1.30 – 1.80 g/cm³ in the east.
- The Zeven Wegen limestone shows a distinct variation in chemical composition between its eastern and western parts (as illustrated by the different $CaCO_3$ and SiO_2 contents in the table).
- Vylen limestone is extremely variable in composition (50 – 98 wt.% $CaCO_3$).

Table 1. continued (Felder & Bosch 2000)

Average composition			Density			Flint content		
$CaCO_3$	$MgCO_3$	SiO_2						
	wt.%			g/cm³			vol.%	
97.8	1.6	0.2	1.28	-	1.47		None	
97.5	1.7	0.3	1.28	-	1.49		<	1
97.4	1.6	0.7	1.28	-	1.37			9
96.0	1.8	1.3	1.21	-	1.40			7.5
95.2	2.5	1.7	1.21	-	1.40	5	-	10
78.7	2.3	15.2	1.30	-	1.80	5	-	8
96.4	1.2	1.3	1.30	-	1.60	18	-	20
							ca	20
91.3	0.8	6.1	1.51	-	1.64		ca	15
							ca	15
70.8	0.8	24.7	1.48	-	1.75		ca	10
66.4	None	30.0	1.50	-	1.57		ca	5
97.1	0.5	2.3	1.55	-	1.57		<	1
80.4		16.5						

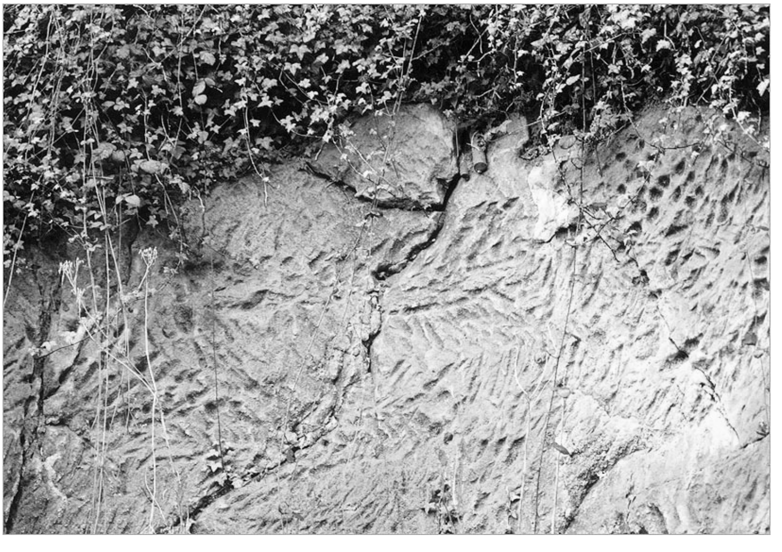

Figure 5. Traces of stone cutting in one of the ancient shafts at the Plenkertweg flint mine (Apr. 2001); this part of the outcrop corresponds to sampling locality 2 on figure 1.

3 Sample description

In order to assess the state of conservation of the Plenkertweg flint mine, samples have been collected from four parts of the site, following a visual inspection. Localities are shown on figure 1. Sampling consisted of powder samples, used to determine actual and hygroscopic moisture contents and drill cores, used for microscopic investigation to determine nature and depth of deterioration. Results are given in chapters 4 and 5, respectively. The sampling localities and drill cores are described below.

Locality 1

The locality itself is shown in figure 6. Details of places from which cores have been obtained are given in figure 7. Sample 1 is a drill core, showing a black to green surface layer, with some loss of matrix up to a depth of 5 mm below the surface. Sample 2 is a drill core, showing a hard, black to grey surface layer. Part of this black crust has come off from the wall. Sample 3 is a drill core with a thick black – green biogenic surface layer, from which water flows upon pressing the surface.

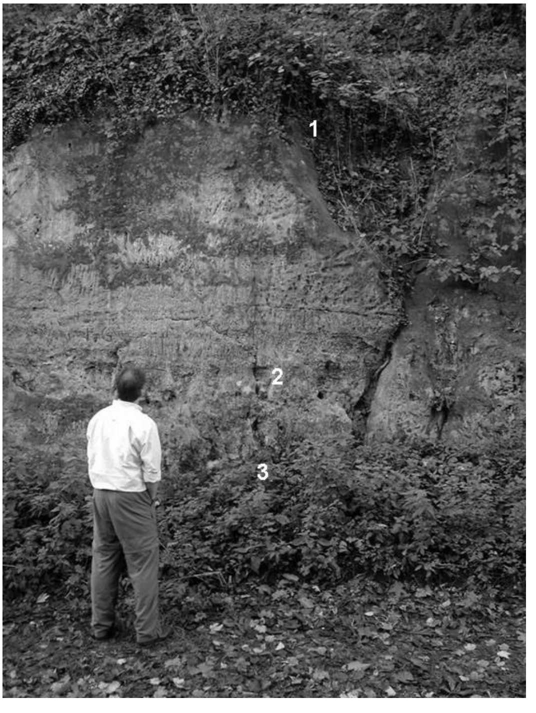

Figure 6. Sampling locality 1 (Oct. 2001)

Locality 2

The locality itself is shown in figure 8. Sample 4 is a shard loosely attached to the wall; the surface is covered with a greenish layer containing hyphae. The matrix appears to be not deteriorated. Samples 5 and 6 are drill cores collected from the boundary between two limestone layers. Sample 6 has been taken from a layer with less integrity, sample 5 just above that layer. The layer from which sample 6 has been derived has back weathered deeper compared to the under- and overlying layers. The surface of sample 5 is covered with a thin layer of algae; below this, some loss of matrix has occurred up to 15 mm depth. Sample 6 shows brownish red and green algae on its surface. Grainsize is coarser, with relatively transparent appearance of the calcite. Down to 15 mm depth, the limestone has partly been disintegrated.

Locality 3

An overview of locality 3 is given in figure 9. Traces of ancient stone cutting at this locality are present in limestone with a disintegrated surface layer. Samples 7 and 8 have been drilled from the back wall of the ancient mine corridor present at locality 3. Both are

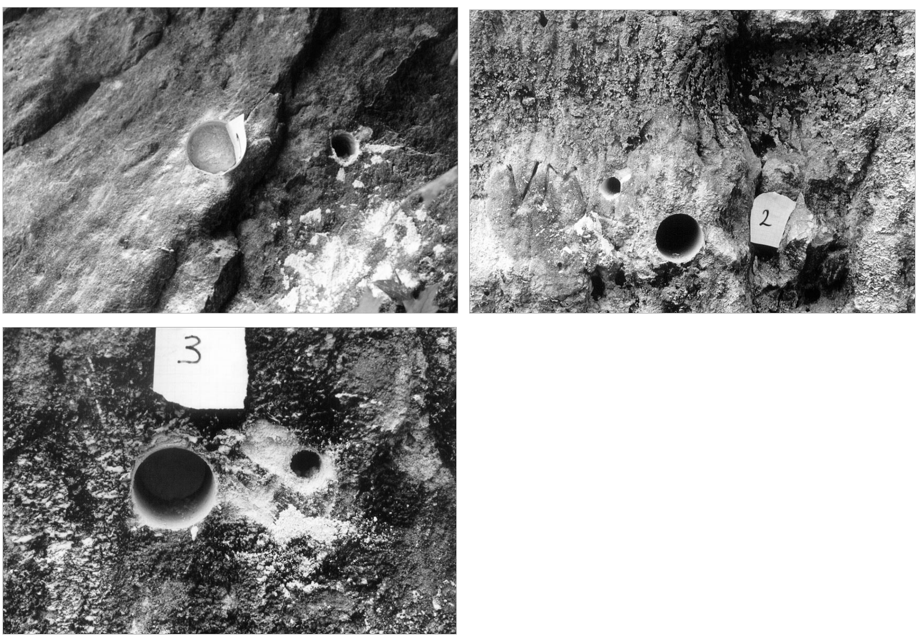

Figure 7. Localities of samples 1 (upper left), 2 (upper right) and 3 (below) from locality 1 (June 2001; see also Fig. 6)

Figure 8. Sampling locality 2 (June 2001)

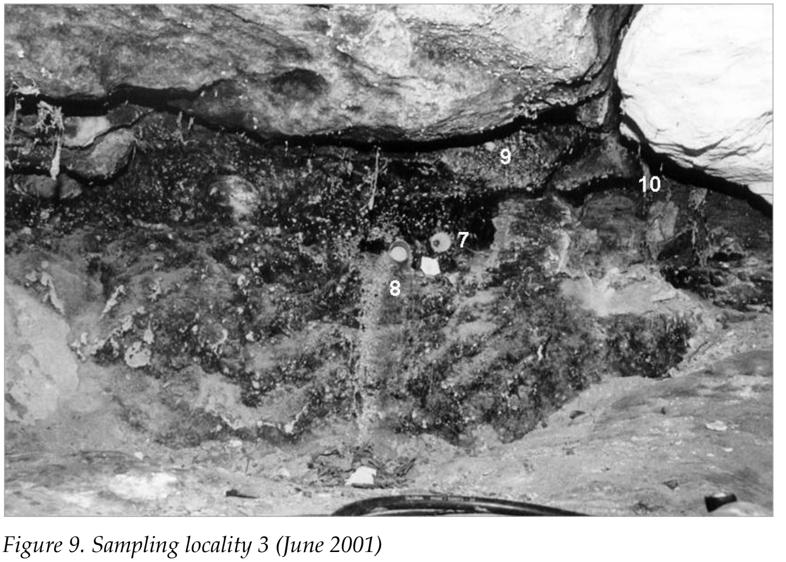

Figure 9. Sampling locality 3 (June 2001)

covered with a thick, moist biogenic layer, composed of algae and mosses. Down to 8 mm below the surface, the limestone has been discoloured and has almost completely been disintegrated (Fig. 10). Deeper, up to 30 mm below the surface, a loss of matrix is clearly visible, whereas locally loss of matrix occurs down to 60 mm deep, i.e. the entire length of the cores. For comparison, the depth of preserved traces of ancient stone cutting in this

Figure 10. Details of biogenic layer on sample 8 and disintegration of the limestone (right picture)

Figure 11. Overview of locality 4 (June 2001)

part of the mine varies from several mm up to about 30 mm. Samples 9 and 10 are powder samples, collected from the roof of the mine shaft; sample 10 has been collected from a partly detached block on the roof.

Locality 4

Locality 4 is the area of a sub vertical fracture in the outcrop (Fig. 11). Sample 11 is a drill core, taken over the fracture. The surface of the limestone is grayish to black and rather hard. The grayish to black colour is due to the presence of a gypsum crust (Fig. 12), which has partly detached. The fracture is partly filled by soil from above.

4 Hygric characteristics

Apparent density and porosity of the Maastricht limestone from the four sampling localities at the Plenkertweg flint mine, as determined according to RILEM CPC 11.3, are given in table 2. Porosities have been determined from the deeper, not deteriorated, parts of the drill cores. Actual and hygroscopic moisture contents (the latter for sample 2 only) are shown in figure 13. Actual moisture contents may amount up to about 13-15 wt.% in sample 9. Hygroscopic moisture contents are low (for all samples, except sample 2 lower

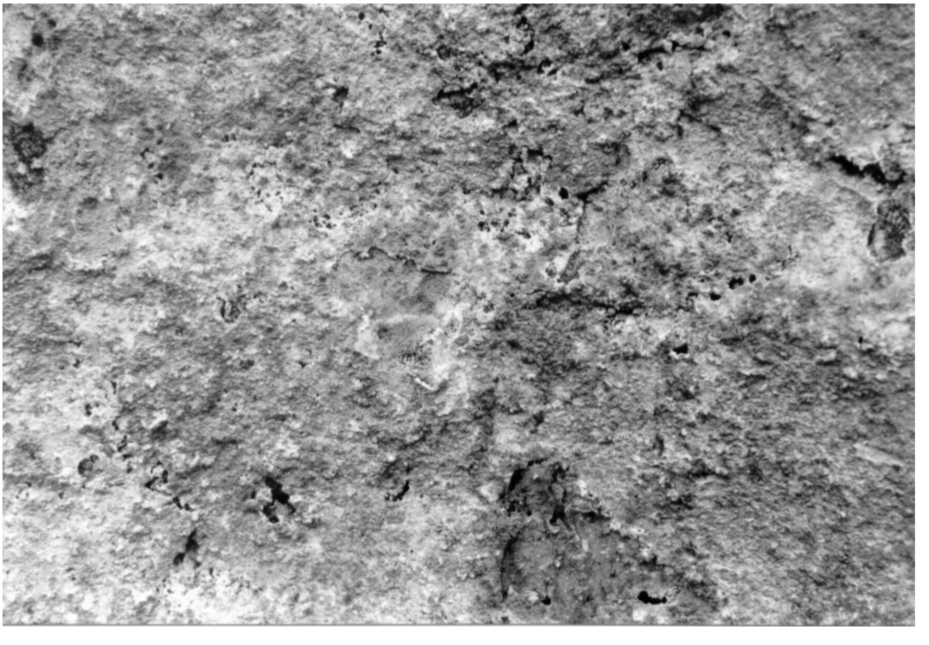

Figure 12. Detail of black gypsum crust at locality 4

than 1 wt.%), i.e. almost no hygroscopic salts are present in the limestone, except for sample 2 from locality 1, in which a minor amount of hygroscopic salts is present at depth.

5 Petrography

Sample 5 (locality 2) has been investigated by polarizing and fluorescence microscopy (PFM). The sample represents a pure, porous bioclastic limestone, with a microstructure

Table 2. Apparent density and porosity of Maastricht limestone from the Plenkertweg flint mine

Sample	Apparent density	Apparent porosity
	kg m-3	vol.%
1	1376	48.1
2	1696	36.0
3	1325	50.0
6	1369	48.3
8	1324	50.3

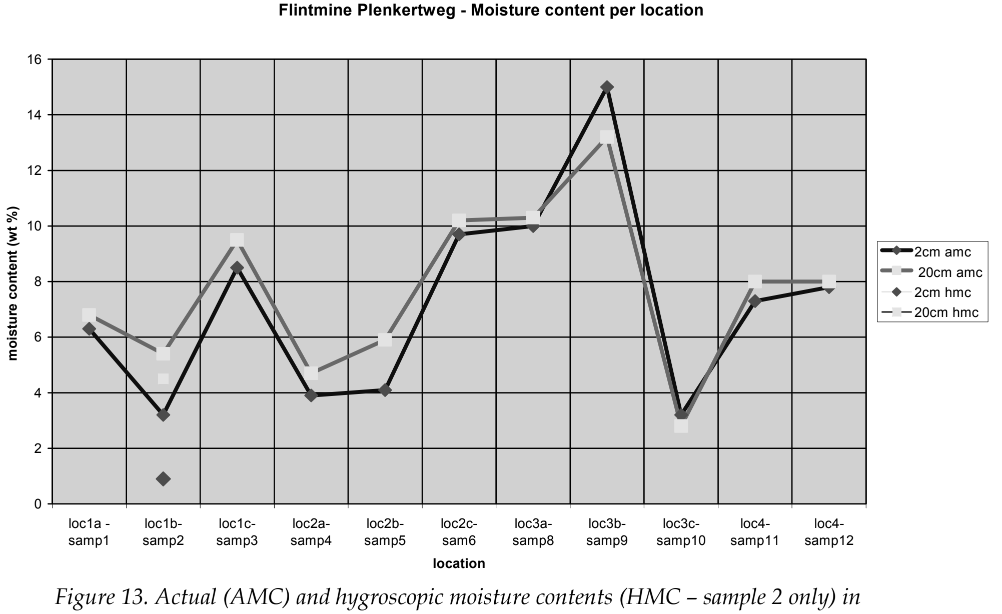

Figure 13. Actual (AMC) and hygroscopic moisture contents (HMC – sample 2 only) in Maastricht limestone from the Plenkertweg flint mine. Moisture contents have been determined at two depths for each sample place.

typical of Maastricht limestone (cf. Dubelaar et al. 2006). No difference in microstructure is observed in the limestone directly below the surface (Fig. 14) compared to deeper levels (Fig. 15). Glauconite nor any other minerals than primary and secondary calcite have been observed. Bioclast fragments are cemented by secondary calcite cement (sparite) to a very limited extent (Fig. 16). Clear homogeneous sparite crystals, occurring as overgrowths on bioclasts, amount 2 – 3 vol.%, and occasionally developing rhombohedral crystal faces. Coarse, intergranular porosity is about 50 vol.% (Fig. 14, 15), in addition to intragranular porosity within the bioclasts. A clear penetration of microorganisms into the surface of the limestone is visible microscopically (Fig. 17).

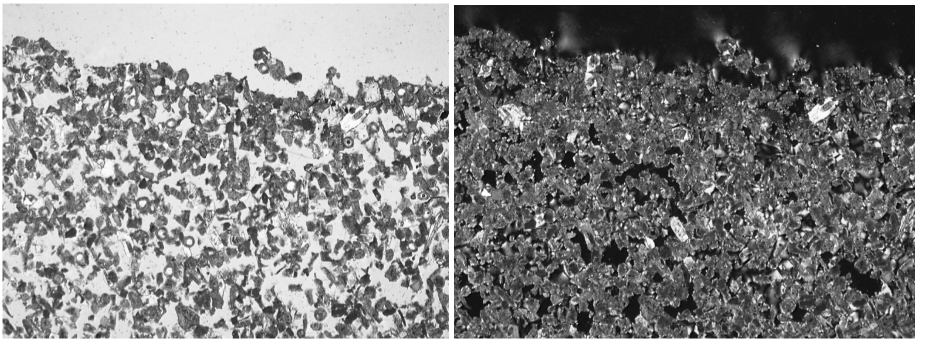

Figure 14. Microphotograph showing an overview of the microstructure of Maastricht limestone in sample 5, directly below the surface (left // polarized light, right + polarized light, view 5.4 x 3.5 mm).

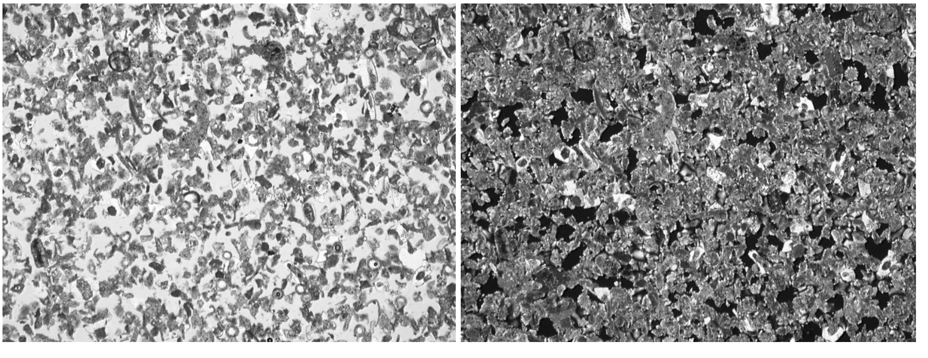

Figure 15. Microphotograph showing an overview of the microstructure of Maastricht limestone in sample 5 at c. 45 below the surface (left // polarized light, right + polarized light, view 5.4 x 3.5 mm).

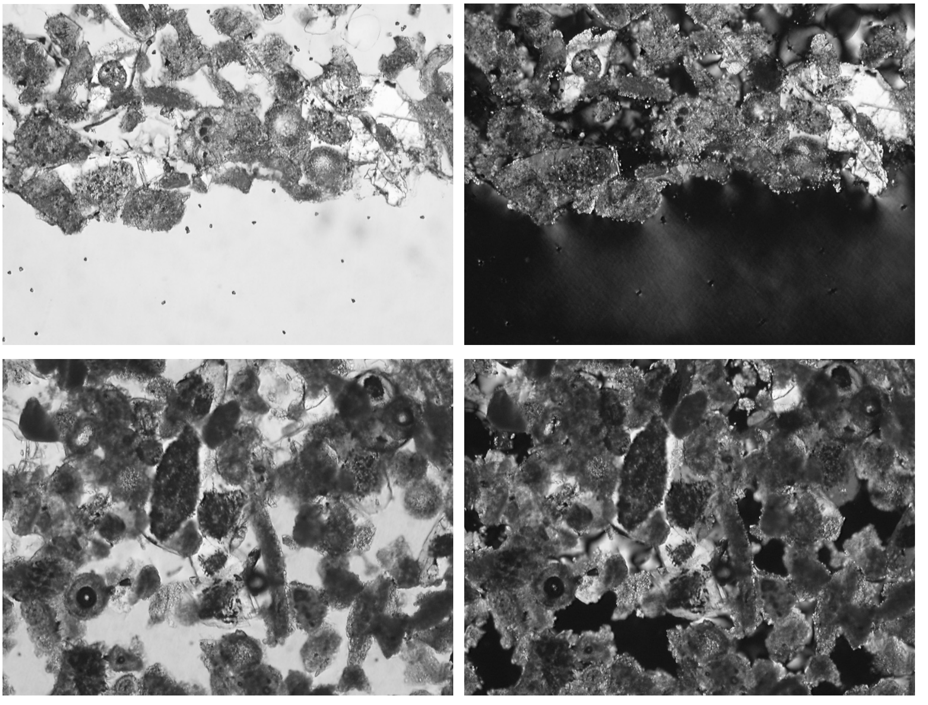

Figure 16. Microphotograph showing secondary calcite (sparite) overgrowths on bioclasts, cementing the latter in the surface of sample 5; upper pictures at the surface, lower ones at c. 20 mm depth (left // polarized light, right + polarized light, view 0.7 x 0.4 mm)

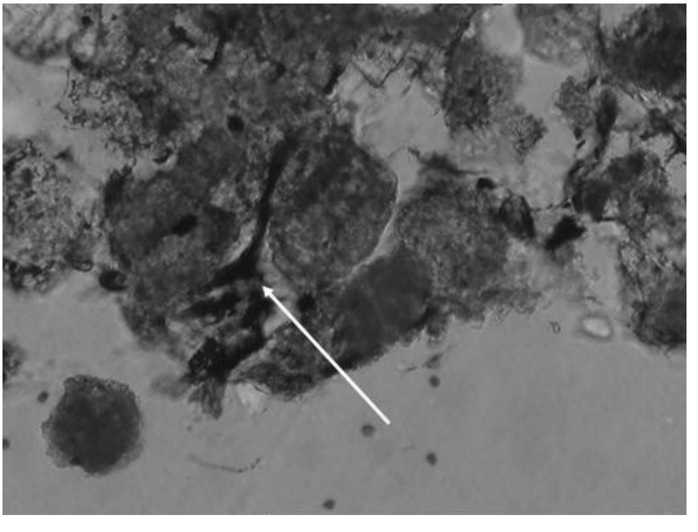

Figure 17. Microphotographs showing penetration of microorganisms into the surface layer of Maastricht limestone in sample 5 (// polarized light, view 0.7 x 0.4 mm)

SEM-EDS investigation of sample 2 (locality 1), with a hard, black crust on the surface, shows this c. 0.2 mm thick crust to be composed of gypsum (Fig. 18-20). In addition to gypsum, the surface layer contains some aluminosilicates, possibly clay minerals. Though rare nodules of clay minerals may occur locally in the Maastricht limestone (Nijland et al. 2007), clay minerals are generally absent. Given the fact that they also occur abundantly on top of rounded, partly dissolved calcite crystals in sample 6 (see below), it is supposed they have been derived from the soil above the outcrop. 1-2 mm below the gypsum crust, the limestone is composed of well defined calcite crystals, also covered with tiny aluminosilicate particles (Fig. 21-22).

Sample 6 (locality 2) is a soft, high porosity, partly disintegrated limestone. The greenish – brown surface layer is shown by SEM to represent a biofilm, in which hyphae of fungi and algae are present (Fig. 23). Below the biofilm, calcite grains have rounded edges and grain boundaries are diffuse, possibly due to dissolution (Fig. 24, 25). Calcite grains are covered by tiny Mg-bearing aluminosilicate grains (Fig. 25, 26).

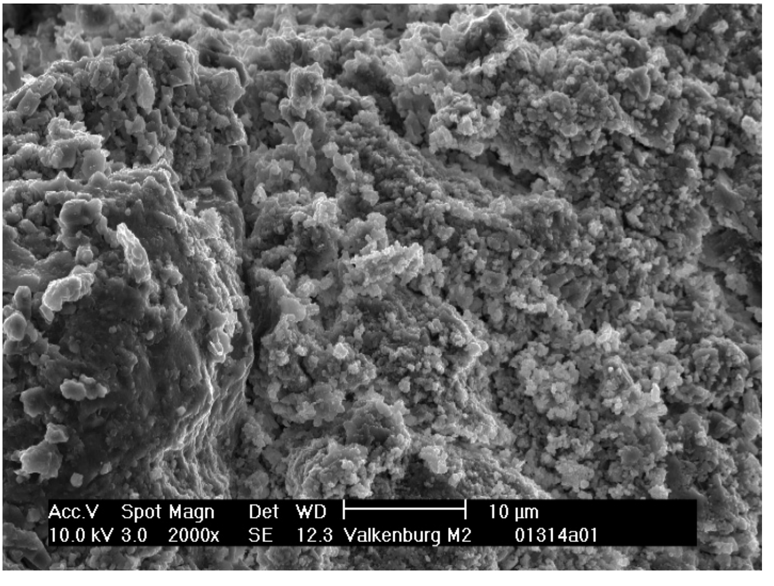

Figure 18. SEM microphotograph showing an overview of the gypsum crust on Maastricht limestone of sample 2 (locality 1)

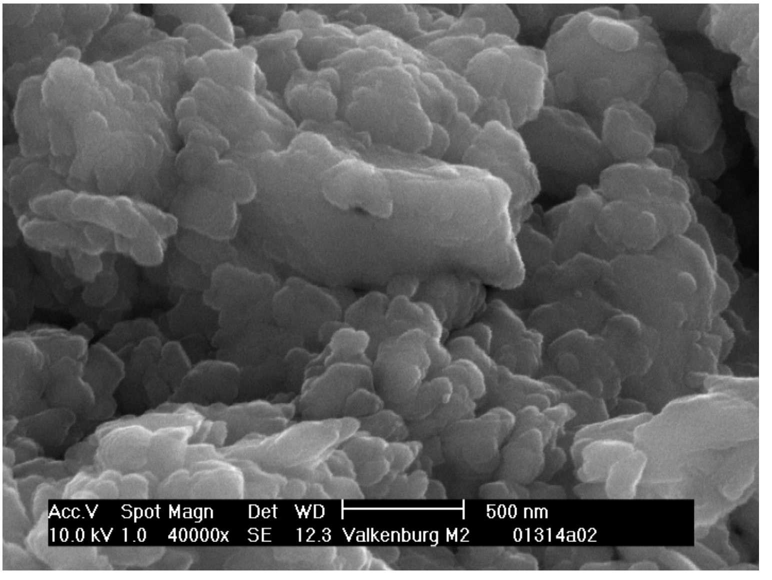

Figure 19. SEM microphotograph showing a detail of the gypsum crust on Maastricht limestone of sample 2

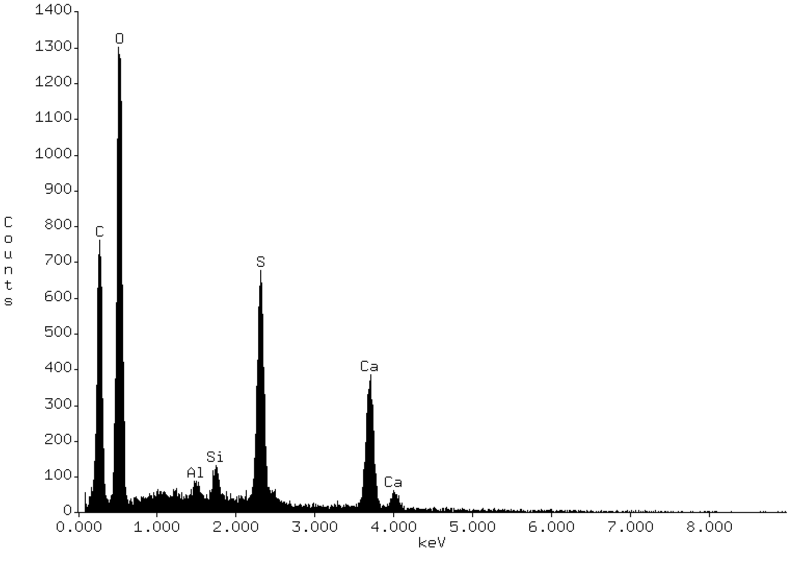

Figure 20. EDS spectrum of the surface layer on sample 2

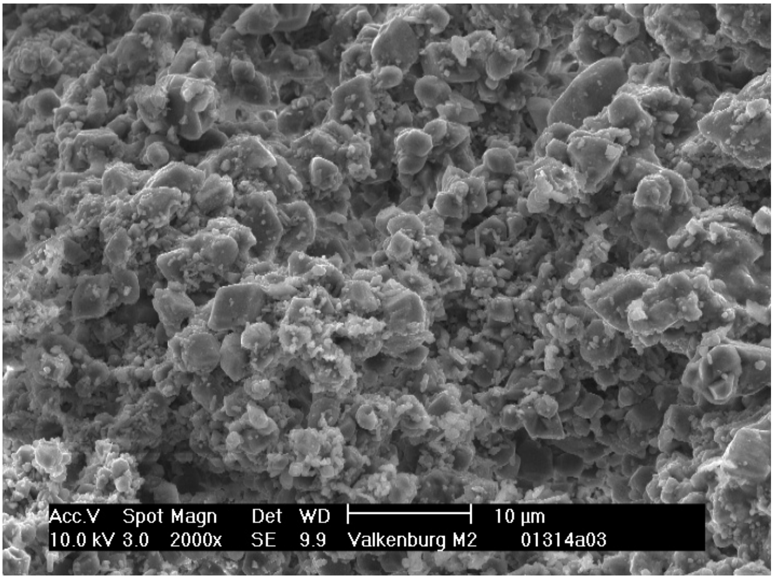

Figure 21. SEM microphotograph showing an overview of the microstructure of Maastricht limestone in sample 2 at 1-2 mm depth

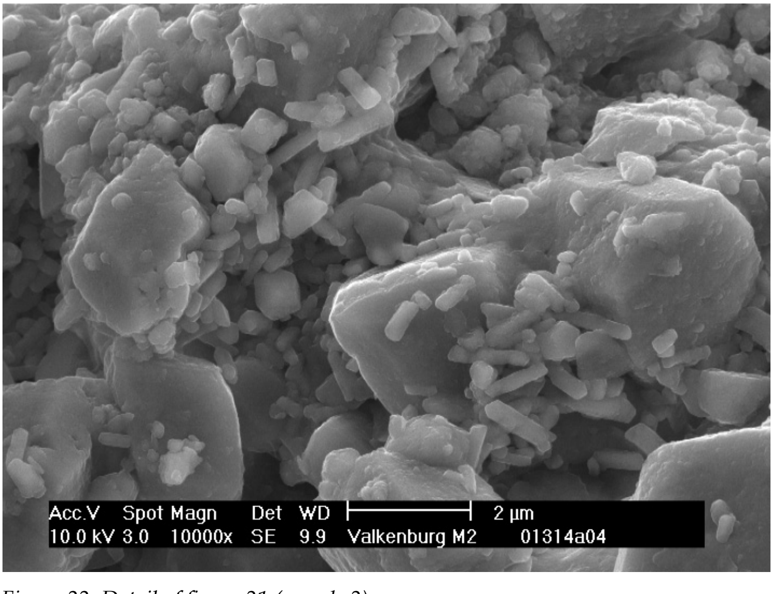

Figure 22. Detail of figure 21 (sample 2)

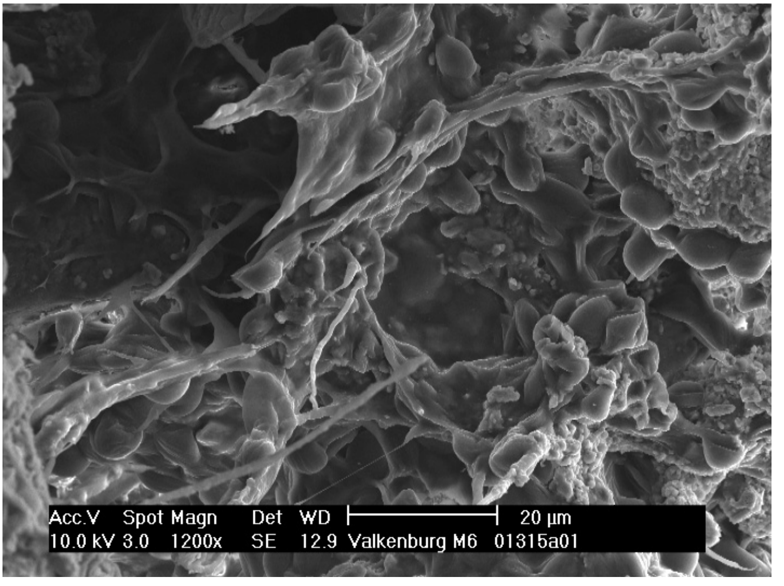

Figure 23. SEM microphotograph of the biofilm on top of Maastricht limestone in sample 6 (locality 2)

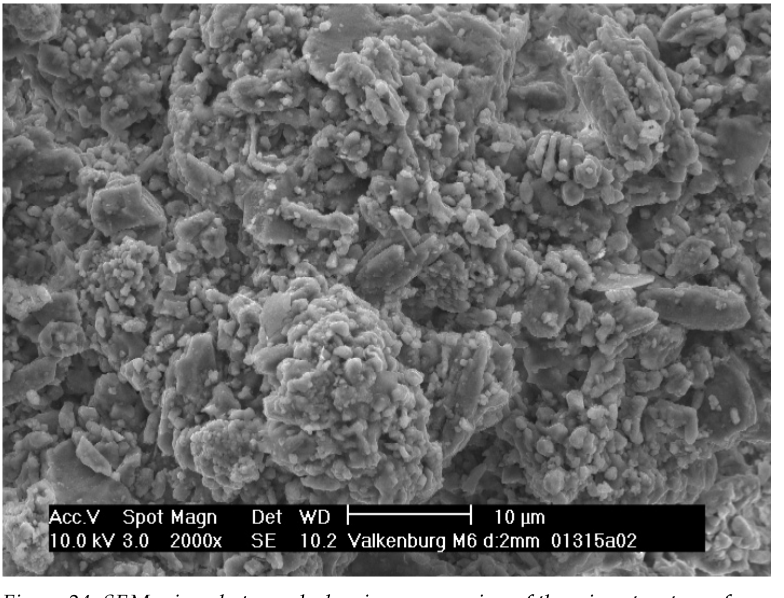

Figure 24. SEM microphotograph showing an overview of the microstructure of Maastricht limestone in sample 6 at 1-2 mm depth

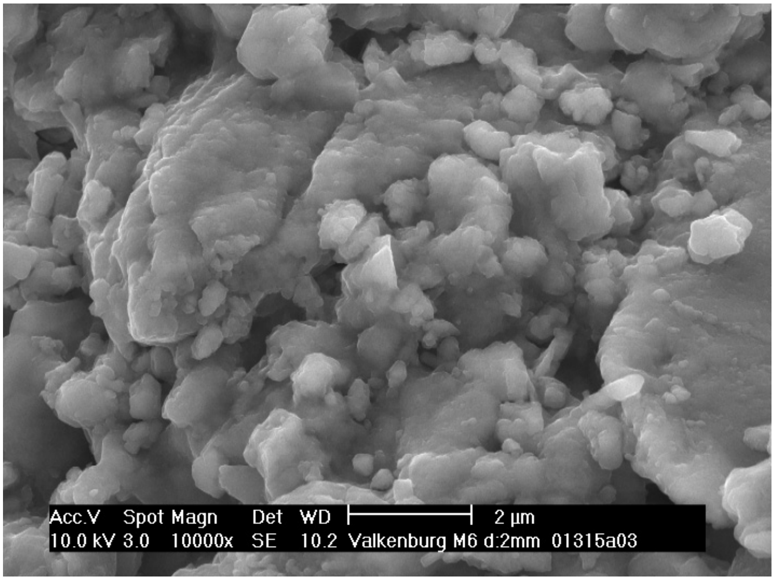

Figure 25. Detail of Figure 24 (Sample 6). Note the tiny Mg-bearing aluminosilicate grains

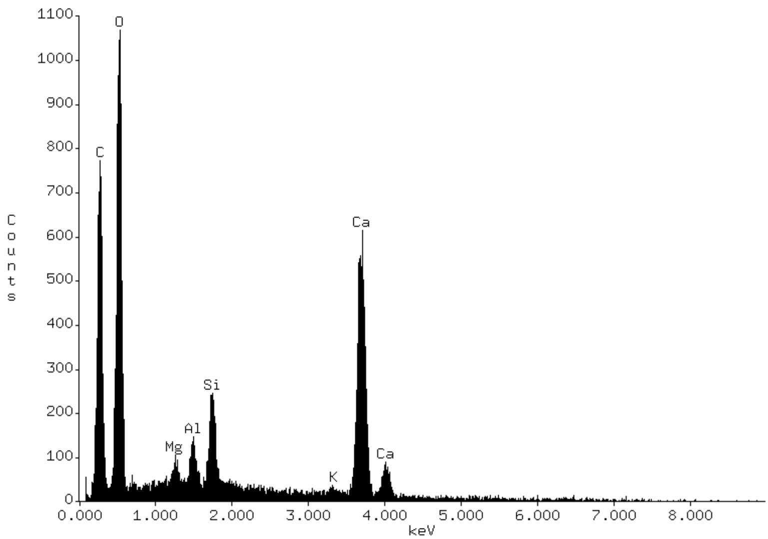

Figure 26. EDS spectrum of Mg-bearing aluminosilicates covering calcite grains in Figure 25 (Sample 6)

6 Discussion and conclusion

At the Plenkertweg flint mine, flint has been mined from the Emael limestone of the Upper Cretaceous Maastricht Formation, which comprises a series of soft, poorly cemented, very porous bioclastic limestones together denominated as Maastricht limestone, or in Dutch: mergel. At the outcrop, Middle Neolithic shafts, corridors and traces of stone cutting have been preserved. Since discovery and exposure, Maastricht limestone in the outcrop shows several damage features. Besides geological fractures and joints in the outcrop, damage patterns are:

- Biocolonization, resulting in thin biofilms composed by algae and fungi as well as thicker biological layers composed by mosses
- Formation of gypsum crusts and partial detachment of these crusts
- Disintegration of the matrix by dissolution of the carbonate cement

As will be evident, the availability of moisture is a controlling factor in all three. Below, the role of moisture is evaluated for localities 1 and 3.

At locality 1, moisture affecting the Maastricht limestone may be derived from several sources:

- Percolation of meteoric water from the overlying soils
- Infiltration of rain water through the surface
- Penetration of water splashing from field and road
- Rising damp

Simultaneously, growth of higher plants in front of the outcrop hampers evaporation, creating a local microclimate. Combined, three zones with different damage patterns developed:

- Along the top of the outcrop, a fairly moist zone stimulates development of algae, accompanied by minor dissolution of the limestone.
- Halfway the limestone outcrop, moderately moist conditions allowed the development of gypsum crusts, protecting the limestone from further damage due to dissolution.

- At lower levels, wet conditions stimulated the development of rather thick biofilms, hampering drying and promoting dissolution of the limestones deeper below the surface.

At locality 3, percolation of meteoric water from overlying soils is the sole source of moisture. However, evaporation is significantly hampered by the very limited air circulation within the ancient mine corridor and high relative humidity in the corridor in the summer, due to lower temperature in the corridor compared with that of the surrounding air, stimulating biocolonization. Subsequently, increasing biocolonization causes a progressive decrease of evaporation.

As will be evident, preventive conservation of the site would primarily be directed at limiting the amount of available moisture whilst simultaneously promoting evaporation. Measures in this respect might include prevention of rising damp by providing drainage channels at the foot of the wall, prevention of splash up water and prevention of biological growth in front of the wall and in fractures. However, more insight in deterioration rate and mechanism is considered necessary.

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