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PRESENCE OF BENTONITE BEDS IN THE EARLIEST EOCENE TIENEN FORMATION IN BELGIUM AS EVIDENCED BY CLAY MINERALOGICAL ANALYSES

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ABSTRACT

In the Tienen Formation of earliest Eocene age in Belgium, at the southern rim of the North Sea Basin, the presence of thin bentonite and bentonitic layers has been identified and confirmed by XRD, ESEM and major element chemical analyses. For the first time in Belgium, bentonites have been described at that stratigraphic level, linking it to the well known earliest Eocene North Sea Basin volcanism related to the onset of the NE Atlantic Ocean opening between Greenland and Europe.

1. INTRODUCTION

The presence of pyroclastic volcanism in the North Sea Basin during the late Paleocene and earliest Eocene is well known (Knox and Morton, 1983, 1988). This igneous activity is related to the initiation of the sea-floor spreading between Greenland and Europe at the start of the formation of the NE Atlantic Ocean.

Pyroclastic tephra layers typically characterise the Balder Formation of earliest Eocene in the central North Sea. In the southern part of the North Sea ash layers are less common (Knox, 1993) (Fig. 1). A few volcanic ash horizons are reported in the Upper Paleocene Ormesby Clay Formation and in the lowermost Eocene Reading and Harwich formations of southeast England (Huggett and Knox, 2006). Also, in the Sparnacien facies of the earliest Ypresian at Varengeville in Northwest France, discrete tephra layers have been identified (Knox, 1993).

In Belgium, only sporadic indirect indications of volcanic activity at the Paleocene-Eocene transition (Table 1) have been suggested based on core descriptions of wells in North Belgium, i.e. the Knokke well (location indicated on Figure 1): a diatom bloom in the top of the Tienen Formation (Oosthoek Sand Member); glass shards in a marine clay layer (the Zoute Silt Member), just above the Tienen Formation and underlying the clays of the classical Ieper Clay Group (King, 1990); pyroxenes in the base of the Ypresian sediments (Geets, 1993) and finally zeolites of the clinoptilolite-heulandite group higher up in the Ypresian clays (Fobe, 1989).

In this paper for the first time bentonite layers are described from the lowermost Eocene of central Belgium, an occurrence at a large south-eastern distance from the volcanic centre. These bentonites were recognized in a temporary excavation and studied in the framework of a large scale study of bulk and clay mineralogical variations in the Eocene-Oligocene interval of the Southern North Sea Basin (Zeelmaekers, 2011). The current paper therefore is strictly focused on mineralogical and petrographic criteria in recognizing bentonites.

2. GEOLOGICAL AND STRATIGRAPHICAL SETTING

At the Paleocene-Eocene transition, the sea level was low and consequently the Southern North Sea area, including the Paris and Belgian basins, was dominated by continental and transitional coastal deposits as discussed and reconstructed in a paleogeographic map by Dupuis and Thiry (1998) (Fig. 1). These deposits are the first sediments to fill a previously incised topography and therefore represent the start of a new transgression; the lowest sea-level position during which the topography below the Tienen Formation deposits was incised, is interpreted as an important regional sea-level low (Vandenberghe et al., 2004) at the start of the Ypresian.

In Belgium these deposits filling the incised topography are grouped into the Tienen Formation (Table 1). In Central Belgium and in the Hainaut province the sediments are dominantly fluviatile while in north and

Chronostratigraphy		Lithostratigraphy			Facies
Eocene	Ypresian	Ieper Group	Kortrijk Fm		marine clays
				Zoute Member	marine silty clays with glass shards
		Landen Group	Tienen Fm		Continental lagoonal sand, clay, lignite, silicified horizons
Paleocene	Thanetian		Hannut Fm		Marine sand, silt, calcareous silt

TABLE 1: Stratigraphic table displaying the units at the Paleocene-Eocene transition discussed in the text.

northeast Belgium (Knokke, Kallo) and northwest France (Cap d'Ailly) the sediments are dominantly brackish lagoonal.

Details of the stratigraphic context of the Tienen Formation and under- and overlying strata can be found in Steurbaut (1998) and Steurbaut et al. (2003). The δ^{13}C shift indicating the onset of the CIE and consequently the Paleocene-Eocene almost coincides with the erosive level underlying the Tienen Formation (Steurbaut et al.,1999; Steurbaut et al., 2003; Magioncalda et al., 2004). In the Knokke well the Tienen Formation is overlain by the Zoute Silt Member from which it is separated by some flint pebbles; the Zoute Silt Member forms the base of the Ieper Group clays in this well. The Zoute Silt Member corresponds to the *Deflandrea oebisfeldensis* acme and hence the upper part of the *Apectodinium hyperacanthum* zone. The appearance of *Wetzeliella astra* occurs in the clays above the Zoute Silt Member. The boundary between the Tienen Formation and the Zoute Silt Member is interpreted as the NP9/NP10 boundary (Steurbaut, 1998). The marine Zoute silty clays are the transgressive system tract above the Tienen Formation lowstand deposits; the transgressive sequence however is eroded and this lowest Ypresian sequence misses a highstand tract because of regional uplift, before renewed subsidence formed the main mass of Ypresian clays (Vandenberghe et al., 1998).

The Goudberg section, in which the bentonite layers were discovered, consists of the upper part of the Tienen Formation in central Belgium. The maximum burial depth of these sediments has never exceeded about 100 to 200 m, as can be derived from the regional geologic history.

2.1 THE GOUDBERG SECTION

The Tienen Formation was exposed in a temporary excavations for the high speed railway construction in the late 90's between Leuven and Tienen in central Belgium (Fig. 2). At the Goudberg section the top of the Tienen Formation exposed about 150 cm of locally occurring variably coloured layered clays and silts (Fig.3) between the level with silicified conifer stumps of the Overlaar petrified forest (Fairon-Demaret et al., 2003) and the classical Ieper Group clays. The lowest black lignitic horizon of this zone (sample EZT52 on Fig. 3) is the level containing the silicified tree stumps that developed during the initial Eocene thermal maximum. The grey, more homogeneous clays above the layered and coloured clayey zone, starting with sample EZT 58 on Figure 3, are the classical Ieper Group clays. The white leached quartz sands at the base of the section (Fig.3) have a widespread occurrence in the region and contain many complex syndepositional deformation structures resulting from the vertical tectonic movements around that time (Sintubin et al., 2000).

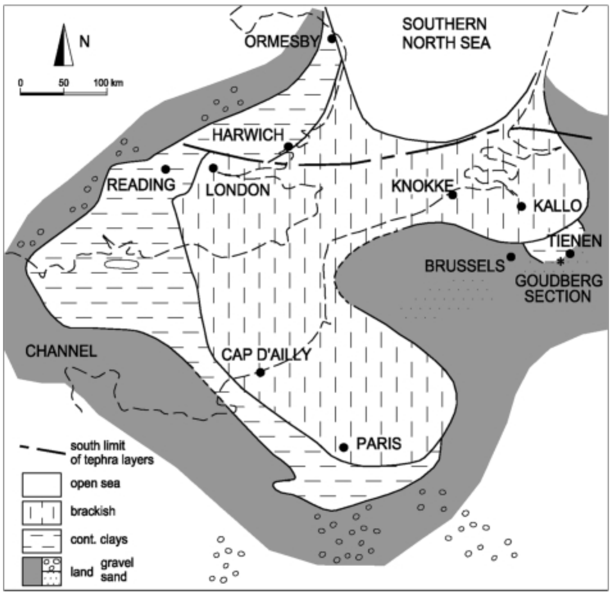

FIGURE 1: Localisation map and paleogeographic sketch map of the earliest Eocene in the southern North Sea Basin (adapted from Dupuis &Thiry (1998) and Knox (1993)). The Goudberg Section is indicated by an asterix.

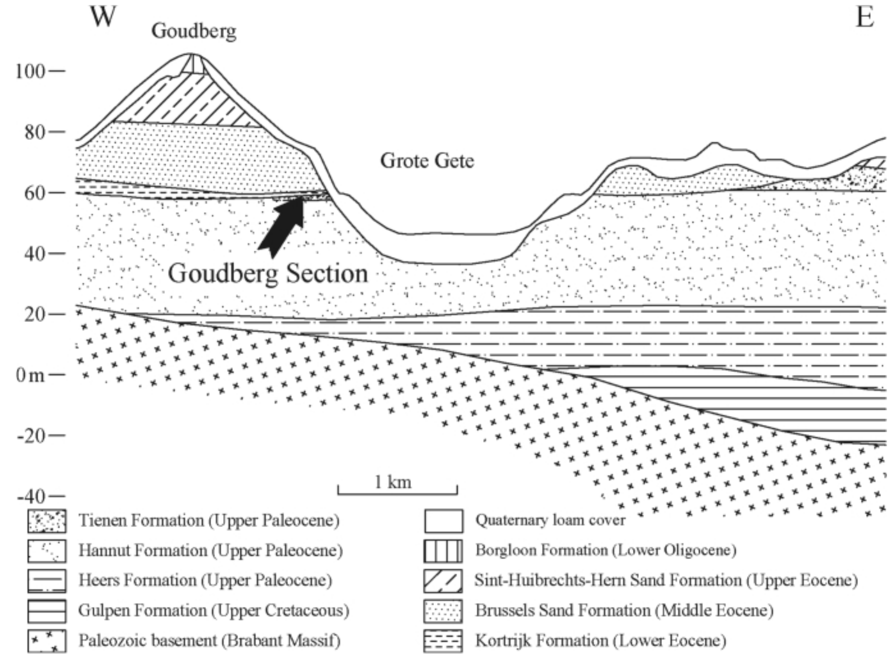

FIGURE 2: Geological section of the Goudberg (modified after Sintubin et al., 2000).

The unusual pale yellowish (EZT51) and pale grey (EZT54) colours of two thin layers in the section raised suspicion for a potential volcanic origin. To test this hypothesis eight samples (Fig. 3) of this section were analysed in detail for clay minerals.

Being part of a larger scale mineralogical study (Zeelmaekers, 2011), the main analytical methods used for the presented study are based on X-ray diffraction (XRD), supplemented by Environmental Scanning Electron Microscopy (ESEM) and major element chemistry.

3. Methodology

Samples were analysed for both qualitative and quantitative bulk and clay mineral composition. Bulk samples were milled, side-loaded and recorded on a Philips PW1830 X-ray diffractometer. After phase identification their quantitative composition was determined using a set of prerecorded standards using the Quanta software, which is based on the method of Środoń et al. (2001) and is generally accepted to be a highly accurate and repeatable method (e.g. Kleeberg, 2005; Omotoso et al., 2006). In parallel, splits of the samples were treated using the Jackson procedures (Jackson, 1975) to be able to extract the <2µm and <0.2µm clay sized fractions. The qualitative and quantitative composition of these fractions was determined by modelling of the oriented slides diffraction patterns using the Sybilla software. Details of the procedures and the software can be found in Zeelmaekers (2011). Quanta and Sybilla are Chevron proprietary software programs. Major element chemistry was determined by a lithium metaborate/tetraborate fusion ICP method at Activation Laboratories (Ontario, Canada).

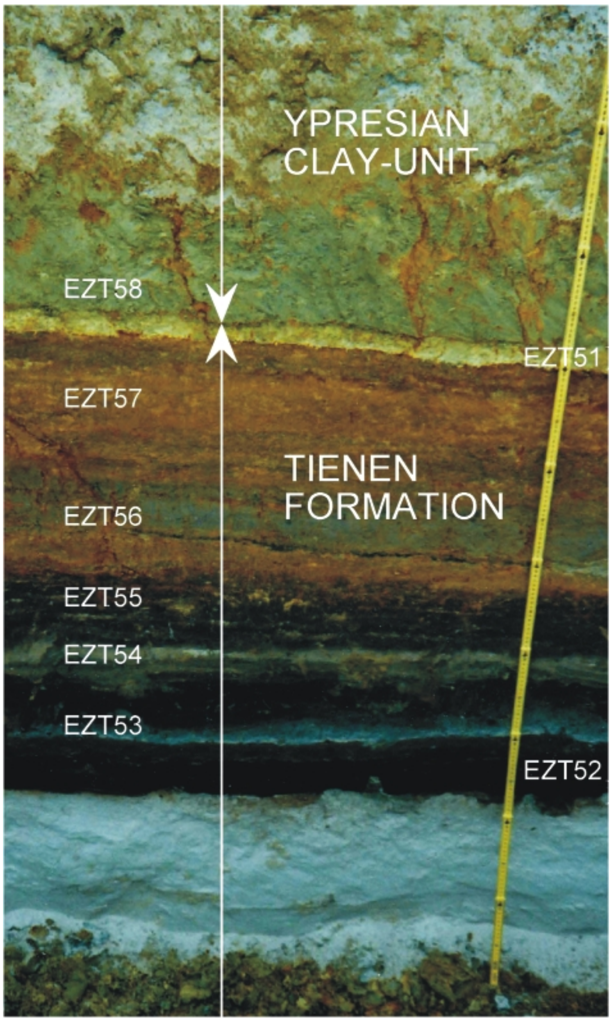

Figure 3: Photograph of the sampled part of the Goudberg section, with numbers and positions of the samples.

4. Analytical results

4.1 Bulk analysis

The most striking observation is that 3 of the samples are extremely and unusually clay-rich (Table 2); the yellowish sample (EZT 51) and the pale grey sample (EZT 54) consist of respectively 93% and 83% of “2:1” clay minerals. The sample from the silicified forest level (EZT 52) contains 92wt% of “2:1” clays. In this paper, and in this methodology, the “2:1” clay minerals are defined as the sum of illite + smectite + mixed-layered illite-smectite.

These are extremely high clay contents and much higher than the other samples of the clayey top of the Tienen Formation, for which the “2:1” clay content varies between 16 and 28 wt%. The 3 clay-rich samples are comparable only to the clay contents of some of the most pure Clay Mineral Society Source Clays, which are known bentonites (see Moll, 2001; Chipera and Bish, 2001). The yellowish sample (EZT51) has an even more unusual assemblage as it contains besides 93wt% of “2:1 clays”, also 6wt% of anatase. The pale grey sample (EZT54) contains 1wt% anatase. The XRD traces of powder recordings of the bulk rock and the extracted <0.2µm fraction are compared in Figure 4, showing that even in the finest fraction still considerable amounts of anatase occur; this is also confirmed by the chemical analysis of 4.2% TiO_2 in the <2µm fraction. The quartz content in the yellowish sample (EZT51) is 0.6wt%, in the pale grey sample (EZT54) 12wt% and in the silicified forest level 5wt% (EZT 52). No other minerals are present in the yellowish sample; in the pale grey sample also 2% K-feldspar and 2% kaolinite are present and in the silicified forest level 0.9% kaolinite and 2% chlorite.

Sample	Qtz	Kspar	Plag	Anat	ΣNon-Clays	Kaol	Chl	Σ2:1	ΣClays	Total
EZT58	60	3	0.4	0.2	**64**	3	0	33	**36**	**100**
EZT51	0.6	0	0	6	**7**	0	0	93	**93**	**100**
EZT57	83	0	0.1	0	**84**	0.5	0	16	**16**	**100**
EZT56	68	0.5	0	0.5	**69**	9	0	23	**31**	**100**
EZT55	59	3	0	0.5	**62**	10	0	28	**38**	**100**
EZT54	12	2	0	1	**15**	2	0	83	**85**	**100**
EZT53	72	2	0	0.9	**75**	6	0	19	**25**	**100**
EZT52	5	0	0	0	**5**	0.9	2	92	**95**	**100**

Table 2: Quantitative bulk rock composition of samples EZT51-58 (all in wt%). Qtz = Quartz; Kspar = K-feldspar; Plag = Plagioclase; Anat = Anatase; Kaol = Kaolinite; Chl = Chlorite; Σ2:1 = sum of 2:1 clays (=illite+smectite+ mixed-layered illite-smectite).

All other samples in the clayey top of the Tienen Formation in the section have much higher quartz contents, ranging from 59 to 83% and show kaolinite contents up to 10%, a composition clearly different from the 3 samples discussed above and more in line with regular clastic sediments.

4.2 CLAY MINERALS

The analysis of the extracted <2µm clay fraction (Table 3) shows the pale yellowish (EZT51) and pale grey (EZT54) horizons to consist of respectively 95 and 93% of smectite, 5 and 6% randomly interstratified mixed–layered illite-smectite ("ISR0"), no discrete illite and 1% kaolinite in the pale grey layer (EZT51). The silicified forest level (EZT52) contains 79% of smectite, 13% of ISR0, 7% of kaolinite and 1% of illite. The other samples of the Tienen Formation in the studied section are also smectite-rich (up to 67%) but contain higher proportions of other clay minerals with up to 24% of kaolinite, up to 8% of illite and up to 13% of ISR0. The ISR0 component in all samples has the same swelling percentage of 34 to 39 % in the mixed layer phase. Additional clay mineral analyses carried out on the <0.2µm fraction show similar results taking into account the smaller grain size (Table 4).

4.3 ESEM

As demonstrated by the presented XRD analyses, the pale yellowish layer (EZT51) and the pale gray layer (EZT54) consist almost entirely of smectite. Such a composition is highly unusual for sedimentary rocks and is indicative of bentonites. To confirm this conclusion the clay fabric of sample EZT51 was examined by an Environmental Scanning Electron Microscope (ESEM) equipped with an EDAX EDS system. Contrary to known layered sedimentary fabrics, the sample shows massive well crystallized clay particles without any apparent depositional features (Figs. 5 and 6). Such a texture is consistent with a bentonite deposit, in which the smectite is an in situ alteration product of the unstable glassy phases originally present in the volcanic ashes. The aforementioned anatase phase is not present as small well-shaped crystals but very finely dispersed, showing a "sponge-like" texture, clearly visible as brighter spots on the dual backscatter image shown in Figure 6.

4.4 MAJOR ELEMENT CHEMISTRY

The <0.2µm fraction of the samples EZT51, 52, 53 and 54 is sufficiently rich in smectite (Table 4) to permit a correction of the chemistry of such samples for the other present minor components (kaolinite and ISR0). The major element composition of these samples is given in Table 5. To correct for the kaolinite, its fixed chemical composition ($Al_4Si_40_{10}(OH)_8$) is used. To correct for the illitic components in

	smectite	illite	ISR0		kaolinite	Total
Sample	**wt%**	**wt%**	**wt%**	*%S*	**wt%**	**wt%**
EZT58	66	11	23	*34*	0	**100**
EZT51	95	0	5	*39*	0	**100**
EZT57	90	2	8	*34*	0	**100**
EZT56	67	3	6	*34*	24	**100**
EZT55	83	1	3	*34*	13	**100**
EZT54	93	0	6	*34*	1	**100**
EZT53	69	8	9	*34*	14	**100**
EZT52	79	1	13	*37*	7	**100**

TABLE 3: Quantitative composition of the extracted <2µm fraction of samples EZT51-58. ISR0 = randomly interstratified mixed-layered illite-smectite; %S = percentage of smectite layers in the mixed-layered illite-smectite phase.

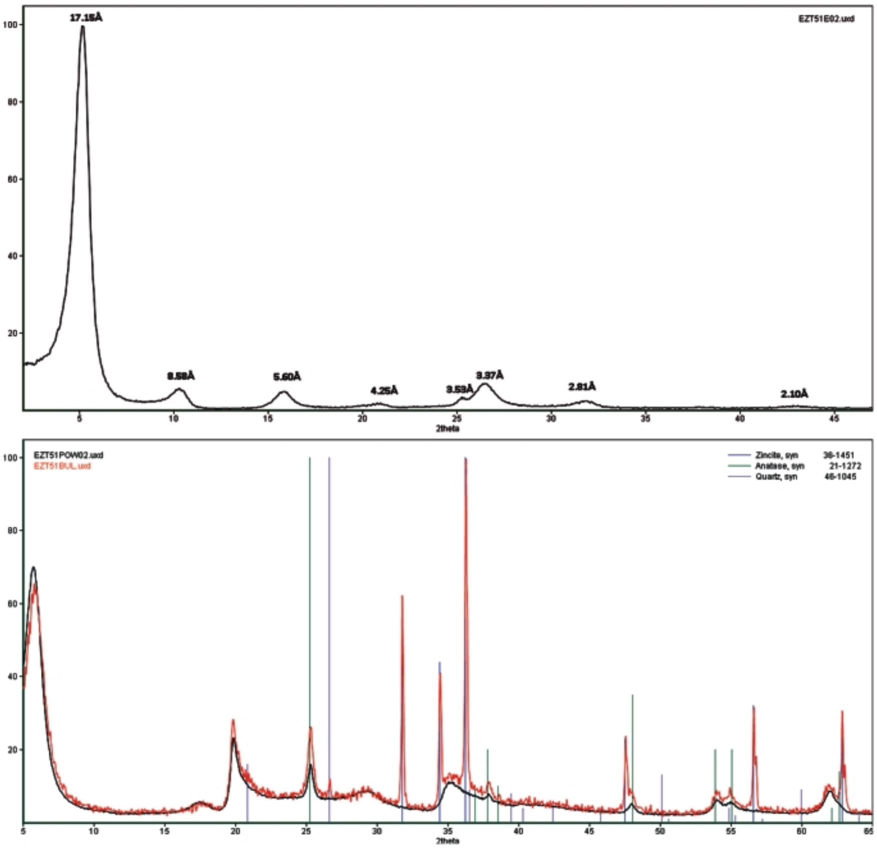

FIGURE 4: XRD traces of sample ETZ51. BUL is the powder bulk XRD trace showing considerable amounts of anatase and some quartz (zincite is the reference additive); POW02 is the powder <0.2 µm trace showing considerable anatase without quartz; E02 is the oriented and ethylene glycolated <0.2 µm trace showing an almost pure montmorillonite.

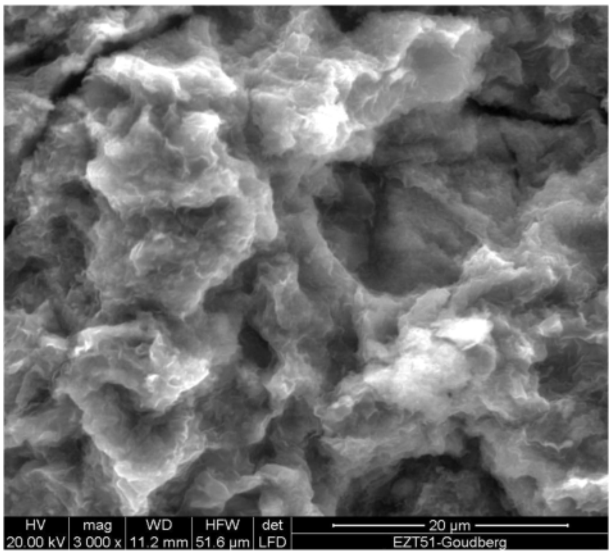

Figure 5: ESEM image of sample EZT51 (magn .3000x) showing an authigenic texture of the smectite phase (courtesy of Chevron, Houston).

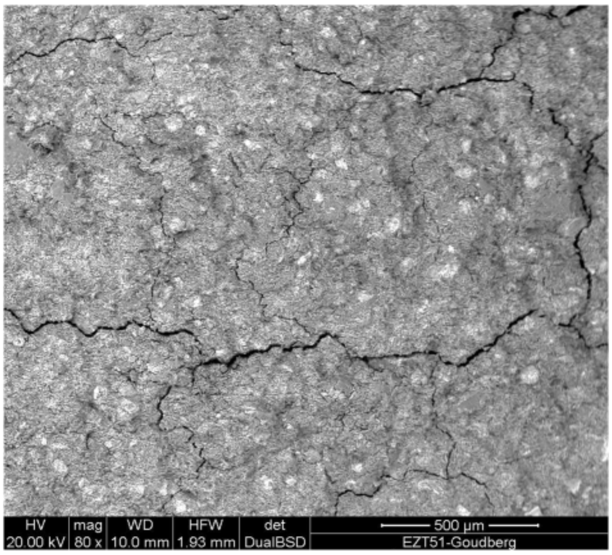

Figure 6: Dual backscatter ESEM image of sample EZT51 (magnification 80x) showing finely dispersed “spongy” anatase occurring as bright spots (courtesy of Chevron, Houston).

the ISR0, the end-member illite composition proposed by Środoń et al. (2009) as $K_{0.95}(Si_{3.25}, Al_{0.75})(Al_{1.81}, Fe_{0.01}, Mg_{0.19})O_{10}(OH)_2$ was used to determine the ratios of each oxide versus the analysed K_2O and thus subtract the correct proportions from the total chemistry. This leads to the following structural formulas for the smectite phase:
EZT 51 (pale yellow sample):
$(Si_{3,933}\ Al_{0,067})(Al_{1,361}\ Mg_{0,320}\ Fe^{3+}_{0,255}\ Fe^{2+}_{0,038})\ O_{10}(OH)_2.EXCH^+_{0,502}$
EZT 54 (pale grey sample):
$(Si_{3,845}\ Al_{0,155})(Al_{1,273}\ Mg_{0,242}\ Fe^{3+}_{0,456})\ O_{10}(OH)_2.EXCH^+_{0,485}$
EZT 53
$(Si_{3,848}\ Al_{0,152})(Al_{1,421}\ Mg_{0,263}\ Fe^{3+}_{0,293})\ O_{10}(OH)_2.EXCH^+_{0.483}$
EZT 52 (silicified forest level):
$(Si_{3,617}\ Al_{0,383})(Al_{1,014}\ Mg_{0,127}\ Fe^{3+}_{0,814})\ O_{10}(OH)_2.EXCH^+_{0.643}$
with EXCH+ referring to the total amount of exchangeable cations.

	smectite	illite	ISR0		kaolinite	Total
Sample	**wt%**	**wt%**	**wt%**	*%S*	**wt%**	**wt%**
EZT51	98	0	2	*39*	0	**100**
EZT54	93	0	7	*34*	0	**100**
EZT53	83	0	11	*34*	6	**100**
EZT52	81	0	11	*33*	8	**100**

Table 4: Quantitative composition of the extracted <0.2µm fraction of samples EZT51-54. ISR0 = randomly interstratified mixed-layered illite-smectite; %S = percentage of smectite layers in the mixed-layered illite-smectite phase.

In the ternary diagrams proposed by Güven (1988), samples EZT51 and 53 plot in the “Wyoming montmorillonite” field, which are known alteration clays originating from volcanic ash. The pale gray sample EZT54 plots in the “Fe-beidellite” field. The sample from the silicified forest level (EZT52) plots clearly outside all fields.

The total layer charge, which is compensated by the exchangeable cations (and thus equal in absolute charge), of all samples is well above the smectite average of 0.41 as proposed by Środoń and McCarty (2008). The 4 samples are also characterized by a high TiO_2 content (1.2 to 4.2%) of the 0.2µm fraction. An above average smectite layer charge and a high TiO_2 content are considered to be indicative for alteration and weathering of basic volcanic rocks as Środoń et al. (1992) observed exactly these features for weathered basaltic

Unit Symbol Detection Limit	SiO2 % 0.01	Al2O3 % 0.01	Fe2O3(T) % 0.01	FeO % 0.01	Fe2O3 % 0.01	MnO % 0.001	MgO % 0.01	CaO % 0.01	Na2O % 0.01	K2O % 0.01	TiO2 % 0.001	P2O5 % 0.01	Total % 0.01
EZT51	44.51	13.93		0.51	3.78	0.011	2.42	2.61	< 0.01	0.14	4.211	0.05	72.26
EZT54	46.41	15.06	7.12			0.02	1.96	2.65	0.01	0.28	2.478	0.05	76.03
EZT53	47.73	18.48	4.39			0.012	2.05	2.5	0.04	0.37	1.508	0.05	77.13
EZT52	42.32	16.3	11.12			0.007	0.93	3.03	0.06	0.33	1.158	0.08	75.34

Table 5: Major element composition for the extracted <0.2µm fraction of samples EZT51-54 as determined by fusion ICP. For sample EZT51, Fe2+ and Fe3+ were determined separately, for the other samples Fe2O3(T) includes both. Loss On Ignition (LOI) analyses were not performed due to small sample sizes, but their values can be assumed as " = 100% - Total ".

tuffs in a detailed study of illite and smectite layer charge on a large set of samples.

5. CONCLUSION

The Tienen Formation comprises a lowstand deposit formed precisely after the start of the Eocene. In its top thin bedded coloured clay layers occur, formed just before the classical marine and deeper water Ypresian Ieper Group clays are deposited in the North Sea Basin.

A mineralogical analysis of these clays demonstrated that a peculiarly coloured thin bed at its top (EZT 51) is an almost pure bentonite. The identification is based on a composition of over 90% Wyoming-type montmorillonite smectite, with only very small amounts of randomly interstratified mixed-layered illite-smectite (ISR0) only traces of quartz, 6% of anatase, and a non-depositional fabric in ESEM. The ISR0 component has to be detrital because only smectite is formed from volcanoclastic glass and the sediments of the Tienen Formation were not buried deep enough to permit illitization. The pale gray layer (EZT 54) is comparable to the yellowish layer but is less pure and contaminated by some more detrital components as shown by the higher quartz content. The blackish layer at the level of the silicified forest (EZT52) is a remarkable sediment; it has an overall mineral composition indicative of bentonite beds but the chemistry of the smectite differentiates it from the pale coloured thin layers. This smectite is most likely also of a pyroclastic origin, but was more diluted by detrital sediment. At present no explanation exists for its peculiar chemical characteristics.

The other samples examined in the section are also very rich in smectite, suggesting a volcanoclastic origin, but their obvious detrital fraction points at reworking and transport into a local lake or lagoon in a coastal plane.

In conclusion, nearly pure smectite layers of a few cm thick, reflect short intervals with intense regional volcanic activity. Between these volcanic intervals the weathered ashes become part of the normal sedimentary processes taking place in the lagoon or lake; also minor new ash falls were likely incorporated in these sedimentation processes.

The stratigraphic position of these bentonite and bentonitic clays, for the first time identified in Belgium, links this volcanic activity to the well known earliest Eocene volcanism in the North Sea Basin described in the literature.

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