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HYGRIC AND HYDRIC EXPANSION OF TUFFS EXCLUSIVELY CAUSED BY CLAY MINERALS?

Wanja Wedekind[1,] Rubén López-Doncel[2], Reiner Dohrmann[3] and Siegfried Siegesmund[1]

[1] *Geoscience Centre of the University of Göttingen, Germany*
[2] *Geological Institute, Autonomous University of San Luis Potosí, Mexico*
[3] *Federal Institute for Geosciences and Natural Resources (BGR) Hannover, Germany*

Abstract

Volcanic tuff rocks show great variety in composition, grain size, sorting and textures. However, scaling, flaking and cracking affect most of them. Various authors suggest that the swelling ability of clay minerals provokes moisture expansion, which contributes to the weathering and deterioration. Fifteen building volcanic tuff rocks from Germany, Hungary and Mexico of different ages, compositions and weathering were studied in order to determine the influence of the moisture content on their deterioration.

Hydric and hygric wetting experiments undertaken on the tuffs show that the phenomenon of the expansion certainly can be related to the content of clay minerals. However, there are tuff samples that show only moderate or no expansion, although they have a significant amount of clay minerals. This study underlines the importance of the pore space properties within the rock material for moisture expansion. The results show that the amount of micropores plays a decisive role in hydric and hygric swelling, while the amount of clay minerals seems to be less important.

On the other hand, we identified tuff types rich in clay minerals and with larger CEC values, which indicate that a stone material has a greater amount of swellable clay minerals and normally has an influence on the moisture expansion. In the presented study a correlation of the CEC-values and the moisture expansion does not appear.

Keywords: volcanic rocks, hydric and hygric swelling, clay minerals, weathering

1. Introduction

Volcanic rocks have been an important natural building stone for millennia. Their use has varied, from monumental sculptures in ancient cultures to modern buildings around the world. These rocks are valued as natural building stones mainly for three reasons. First, some of these rocks and tuffs in particular are relatively soft and easily reworked. Second, their abundance – tuff rocks are present in many places worldwide and in practically all geologic periods. Third, tuff rocks occur in a great variety of colors, composition, grain size, sorting and textures, which make these rocks very attractive for construction and artwork (Fig. 1).

Tuffs are volcanic rocks that consist of a combination of greater or lesser amount of fine silt-sized clay material, with embedded material ranging in size from sand and gravel up to block-sized fragments. The arrangement between these tuff components and the lithology itself provide for a very wide spectrum of porosities, very different textures (Fischer, 1961) and normally large amounts of clay minerals in the matrix.

Fig. 1 Volcanic rock used as building stones and for artwork: a) Rochlitz porphyric Tuff (portal at monastery Wechselburg/Germany), b) Hilbersdorf Tuff (tulip pulpit cathedral of Freiberg/Germany), c) Cantera Rosa San Miguel el Alto (cathedral Arandas/Mexico), d) Blanca de Pachuca Tuff (Palacio Postal Mexico-City), e) Tenayocátetl tuff (Aztec sculpture Tlatecuhtli, Templo Mayor, Mexico-City), f) Chiluca and Cantera gris de los Remedios (cathedral of Mexico-City), g) Cantera Amarilla Tuff (Santa Monica Church, Guadalajara/Mexico).

Yet at the same time, these characteristics make tuffs less resistant to erosion, particularly if they are exposed to the moisture and humidity (Stueck et al., 2011; Ruedrich et al., 2011; Timothy et al. 2011).

Deterioration on buildings is often concentrated in areas affected by temporary moisture infiltration and leads to different damage phenomena (Fig. 2). Moisture expansion is clearly recognized as one of the most important factors that contribute to the weathering and deterioration of building rocks (Ruedrich et al., 2011; Weiss et al. 2004), and is mostly associated with the swelling and shrinking of clay minerals (De la Calle and Suquet 1988; Jimenez Gonzalez and Scherer 2004).

In principle, there are two mechanisms that can act inside and between the clay minerals: the innercrystalline (intra-crystalline) (Madsen and Mueller-Vonmoos 1989) or the osmotic (inter-crystalline) swelling (Heim 1990).

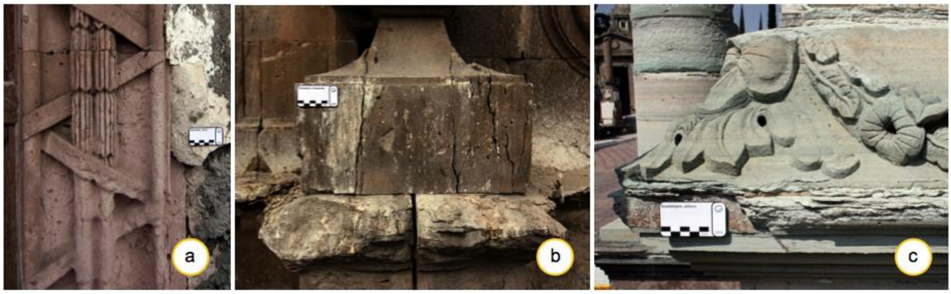

Fig. 2 Different damage phenomena observable in tuffs traced back to moister infiltration and expansion of the rock material. a) Back weathering of Cantera Rosa Tuff of San Miguel de Alto, Jalisco / Mexico. b) Extensive cracking parallel to the bedding of the Bufa Tuff in Guanajuato / Mexico. c) Scaling parallel to the bedding of Loseros Tuff found in Guadalajara / Mexico.

To understand these mechanisms, it is crucial to have control of the amount and the type of clay minerals that are present, because 1:1 and 2:1 layered clay minerals can condition this effect through their cation exchange capacity and the water uptake provoked by the electrolyte concentration in water that causes an osmotic swelling (Madsen and Müller-Vonmoos, 1989; Heim 1990). Similar to the osmotic swelling and related to the electrolyte concentration of a solution, there is also a "non-clay associated" swelling type, known as disjoining pressure (Derjaguin and Obukov 1936), which is more dependent on the size and amount of micropores in the rock (Ruedrich et al. 2011).

As seen above, microporosity plays an important role not only for the disjoining pressure, but also because with the critical pore size of 5 μm the pore water is trapped and cannot drain out of the stone (Larsen and Cady 1969). Pore water hosted in the micropores can contribute to the dissolution of certain minerals in the matrix or to the acceleration of the swelling of clay minerals. Once the rock has been affected by internal weathering, the effective porosity ratio is increased (Tugrul 2004), allowing a major water circulation in all its variants into the rock.

This phenomenon also occurs in all the other types of porosity, and their size and distribution within the rock are likewise decisive for stone durability (Benavente et al. 2004, Siegesmund and Snethlage 2011).

The main goal of this investigation is to contribute to the knowledge about moisture expansion in building stones from volcanic rocks. This study used fifteen important building volcanic tuff rocks from Germany, Hungary, and Mexico of varying ages, compositions, and weathering stages (Fig. 3).

Hydric and hygric wetting experiments carried out on the tuff rocks, as well as chemical and mineralogical analyses on the clay-fraction, including XRD of oriented slides and CEC-analysis, were performed in order to determine the total amount of swellable clay minerals and to clarify the role of the presence of 1:1 or 2:1 layered clay minerals in determining tuff and its relationship to moisture expansion.

Furthermore, the following petrophysical characteristics of the studied rocks were measured: densities (bulk and particle density), porosities (pore radii distribution, pore radii classes and average pore radius), water sorption and tensile strength, compressive strength, and E- Module determinations under dry and water-saturated sample conditions.

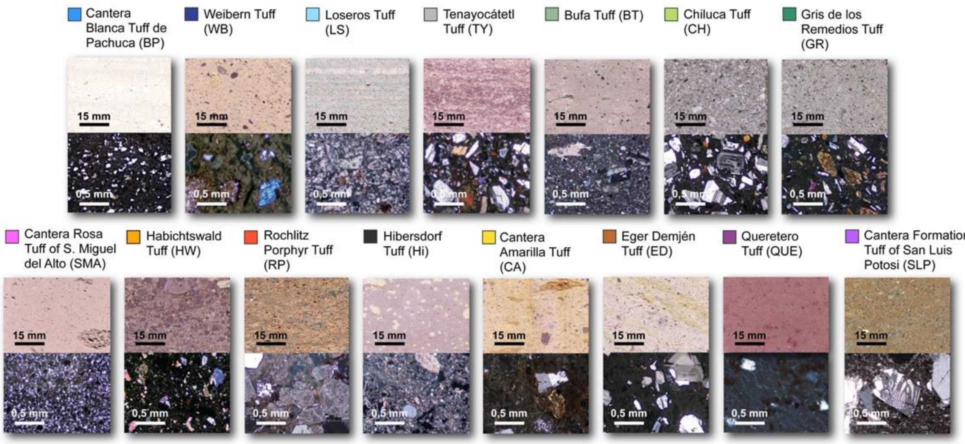

Fig. 3 The investigated volcanic rocks.

2. Petrography and mineralogy

The ages of the tuff rock studied ranged from the Permian to the Recent with chemical compositions that vary from basalt tuff to dacite tuff up to rhyolitic ignimbrite tuff. Petrographical analysis on each tuff sample was done on oriented thin sections under the polarisation microscope. In addition to optical microscopy, the mineralogical composition of bulk samples and clay fractions were analyzed using X-ray diffraction. By using the cubric-triethylentetramine method (Dohrmann and Kaufhold 2009), further information on the clay minerals was obtained. The results of X-ray diffraction and of the clay mineralogy are listed in Table 1.

3. Petrophysical Investigations - Methods

Measurement of open (effective) porosity was performed by hydrostatic weighting on stone cubes (six and a half centimeters long). Capillary water absorption was measured in three directions (X, Y, Z) on the same sample cubes. The cubes were placed into water and their weight increase measured over time.

The pore size distribution was determined by mercury porosimetry (cf. van Brakel et al. 1981). Microporosity is defined as all pores between 0.001 to 0.1 µm. The capillary active pores range between 0.1 and 10 µm.

The moisture expansion by hygric and hydric wetting of the volcanic rocks was determined on cylindrical samples (∅20 mm x 100 mm). To evaluate hydric moisture expansion, these stone cylinders were measured under water-saturated conditions. The hygric moisture expansion was carried out in a special measuring environment, connected to a climate chamber. The moisture expansion was measured stepwise, beginning with 20% relative humidity (RH) and gradually increased up to 95% with a constant temperature of 30°C. For water-saturated conditions, the samples were completely immersed in distilled water. The resolution of the displacement transducer is 0.1 µm and the accuracy is about 0.5 µm. The measurements were carried out on all samples parallel to the X-, Y-, and Z-direction (parallel to the bedding and to the foliation and perpendicular to the bedding). Further information about the clay minerals was obtained using the cubric-triethylentetramine method (Dohrmann and Kaufhold 2009).

3.1 Results

Pore size distribution can be grouped according to ideal pore size types such as equal, unequal unimodal, and unequal bimodal (Siegesmund and Dürrast 2011). Various pore types can be distinguished in the investigated volcanics.

The group of largely equal porous structure consists of the Bufa tuff and the Blanca de Pachuca tuff. Both tuffs have a high microporosity, 78% for the Bufa tuff and 90% for the Blanca de Pachuca tuff. Both volcanics also are comparable in porosity and density. The Bufa tuff has a porosity of 18% and a bulk density of 2.13 g/cm^3; the Blanca de Pachuca tuff a porosity of 15% and bulk density of 1.84 g/cm^3 (Tab. 2). Both rocks show the highest values of hydric dilatation perpendicular to their fine-layered structure in the Z direction. The maximal values of 0.8 mm/m for the Bufa tuff and 0.6 mm/m for the Blanco de Pachuca tuff could be reached (Fig. 4). In the Blanco Pachuca tuff a high amount of smectite was found, and the CEC-value with 15.6 meq/100g is very high as well. The Bufa tuff contains only a higher amount of mixed layers with the I-S (R3)-structure and a CEC-value of 3.0 meq/100g (Fig. 4, Tab. 1).

The Loseros tuff and the Rochlitz tuff belong to the group of equal pore size types with a wider spectrum of pore sizes. While the highest amount of pores of the Loseros tuff are micropores, with 73% between 0.01µm and 0.1µm, in the case of the Rochlitz tuff the greater pore sizes dominate, with 52% between 0.1µm and 1µm.

The data for porosity and density are given in table 2. The highest values of hydric dilatation of both tuffs can be measured perpendicular to their layered structure in Z-direction.

Tab. 1 XRF chemical analysis and CEC values (for abbreviations see fig. 3).

Rocks	**XRF chemical analysis (wt.%)**											**CEC**
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	MnO	TiO_2	P_2O_5	(SO_3)	meq/ 100 g
LS	73.8	15.2	0.2	3.2	0.8	3.5	1.1	0.07	0.09	0.06	0.03	7.1
BP	77.3	13.5	1.4	2.2	0.4	4.1	1.0	0.02	0.08	0.02	0.01	15.6
SMA	76.1	12.8	1.8	0.3	0.1	5.2	3.4	0.05	0.10	0.02	<0.01	2.5
CH	66.3	16.7	3.4	4.5	1.8	1.8	4.5	0.06	0.54	0.17	<0.01	0.4
TY	69.8	15.4	3.6	3.6	1.3	2.1	3.3	0.06	0.50	0.14	0.02	17.6
BT	72.8	14.1	2.6	2.6	0.6	2.5	4.2	0.06	0.23	0.12	0.01	3.0
GR	67.7	14.1	5.2	3.7	1.4	2.7	3.8	0.05	0.76	0.23	0.03	9.6
CA	71.4	14.4	3.7	0.2	0.2	4.8	4.6	0.07	0.31	0.09	0.01	4.2
SLP	74.1	13.4	2.2	0.6	0.1	7.5	1.6	0.02	0.19	0.04	<0.01	0.3
QUE	76.6	12.0	2.0	0.7	0.2	4.0	3.9	0.04	0.22	0.07	<0.02	0.3
HW	51.9	13.6	10.4	7.4	9.4	1.7	2.0	0.2	2.2	0.7	<0.03	168
WB	59.1	20.1	4.1	2.7	0.9	6.1	5.7	0.17	0.51	0.15	<0.04	4.9
Hi	74.6	14.1	2.2	0.5	0.4	7.5	0.1	0.04	0.21	0.04	<0.01	0.7
RP	72.2	23.1	3.5	0.2	0.1	0.1	0.1	0.01	0.40	0.07	<0.01	1.1
ED	71.5	14.7	3.3	2.6	0.5	4.5	2.2	0.07	0.25	0.17	<0.01	10.4

Tab. 2 Pore space properties (for abbreviations see fig. 3).

Stone type	Effective porosity [Vol%]	Particle density [g·cm^{-3}]	Bulk density [g·cm^{-3}]	Average pore radius [µm]	Micropore porosity [%]
WB	37.1	2.43	1,52	0.412	16
ED	24.56	2.29	1.48	0.495	14
HW	8.76	2.59	2.38	0.004	70
RP	27.13	2.64	1.92	0.335	21
Hi	30.24	2.63	1.83	0.145	51
BT	18.39	2.61	2.13	0.06	82
LS	7.06	2.34	2.18	0.095	95
SMA	40.8	2.56	1.53	0.988	13
CA	41.56	2.53	1.48	0.554	18
TY	4.74	2.38	2.26	0.303	47
CH	7.95	2.58	2.37	0.358	27
GR	31.03	2.55	1.82	1.285	15
BP	15.26	2.53	1.38	0.035	97
QUE	46.2	2.53	1.36	2.353	6
SLP	12.83	2.6	2.26	0.804	15

An extreme maximal value can be reached for the Losero tuff of 1.4 mm/m by hydric dilatation while the value for the hygric swelling is more than three times less. The Rochlitz tuff shows a hydric dilatation of 0,42mm/m and a much smaller hygric swelling (Fig. 4). For the Loseros tuff, a very high amount of mixed layers (I-S – R0 & R3) was found. Also the CEC-value with 7.1 meq/100g is noticeable. The Rochlitz tuff contains very high amounts of kaolinite and has a CEC value of only 1.1 meq/100g (Fig. 4, Tab. 1).

The Cantera Amarilla tuff, the Cantera tuff from San Luis de Potosi, the Cantera Rosa tuff from San-Miguel el Alto, the tuff from Querétaro, the Weibern tuff and the Eger Demjén tuff belong to the group of unequal pore size types, with an unimodal spectrum of pore sizes. The six volcanics contain mainly capillary active pores between 1µm and 10µm, 94% for the Querétaro tuff till 82% for the Cantera Amarilla. Consequently, the proportion of micropores ranges from 18% to 6% (Tab. 2). The five types of tuff show moderate hygric dilatation. The highest dilatation is noticeable for the Cantera Amarilla with 0.90m/mm in the Z-direction. In this tuff small amounts of smectite and kaolinite were found, and the CEC-value reached 4.2 meq/100g. The Weibern tuff also shows high dilatation with up to nearly 0.6 m/mm in the same direction. Its hygric dilatation shows at a relative humidity of 95% an even higher value than under water-saturated conditions (Fig. 4). In the Weibern tuff muscovite/illite were found. The CEC-value can be given with 4.9 meq/100g. The Cantera Rosa and the Querétaro tuff show a very low or no hygric or hydric dilatation. Some smectite and mixed layer K-S (R0) has been found for the Cantera Rosa tuff by reaching only a low CEC-value (Tab. 1). A minimal reduction of hydric swelling is observable in case of the Cantera tuff.

The Eger Demjén tuff shows low hygric and hydric swelling (Fig. 4). The Cantera tuff contains measurable amounts of muscovite/illite and kaolinite, reaching a CEC-value of only 0.3 meq/100 g. The Eger Demjén tuff shows moderate moisture expansion but a high CEC-value of 10.4 meq/100g.

Five of the investigated stones belong to the group of volcanics with an unequal pore size type and a bimodal spectrum of pore size.

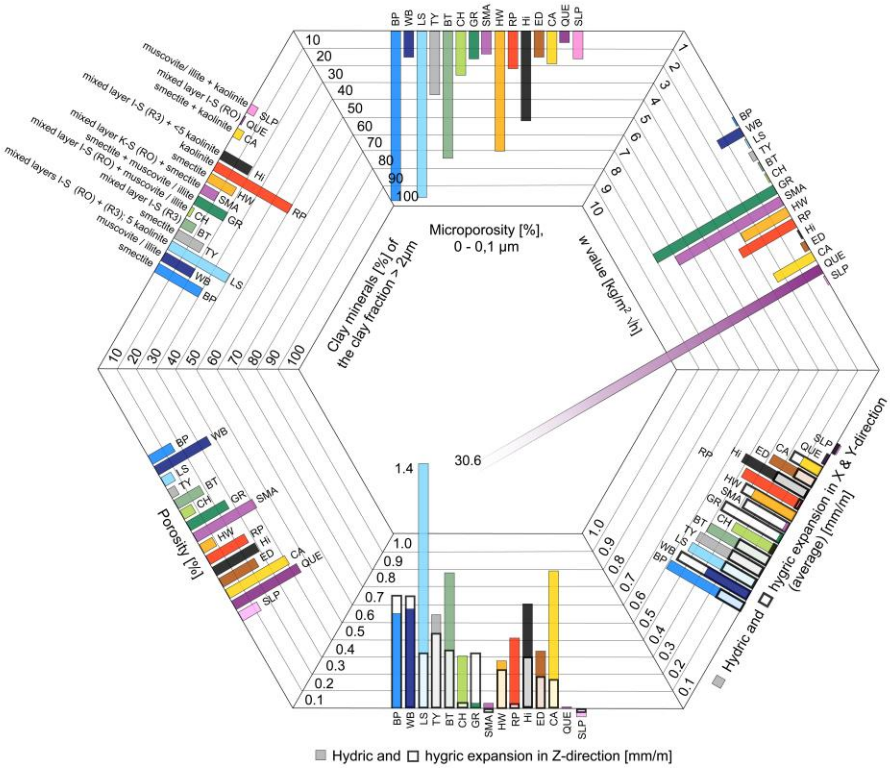

Fig. 4 Overview diagram to moisture uptake, porosity, clay minerals, CEC-values and moisture expansion of the investigated volcanics.

With the exceptions of the Habichtswald tuff and the Hilbersdorf tuff, the amount of active capillary pores is higher than the amount of micropores. The Cantera Gris de Remedios tuff has the highest amount of active capillary pores with 85%. The rest contains a weakly developed accumulation of the pore class from 0.01 to 0.1 µm. The Chiluca tuff contains 73% mostly capillary active pores, followed by the Tenayocátetl ignimbrite with a concentration of 47% of greater capillary active pores, but also a clear accumulation of micropores. For the Hilbersdorf tuff, the ratio of micropores to capillary active pores is evenly balanced. The Habichtswald tuff has a clear accumulation of micropores of 72%, but also a few greater capillary active pores.

The Habichtswald tuff shows moderate hygric and hydric dilatation and contains a high amount of smectite (Fig. 4). The CEC-value of 16.8 meq/100g is the second highest of all volcanites investigated in this study. The Hilbersdorf tuff shows higher moisture dilatation than the Habichtswald tuff and contains a high amount of mixed layer clay minerals I-S (R3). The CEC-value of 0.7 meq/100g is low (Tab.1). The Gris de Remedios tuff shows low hydric dilatation and a high hygric swelling (Fig. 4). This rock contains smectite and has a high CEC-value of 9.6 meq/100g (Tab. 1). The Chiluca tuff with around 0,3 mm/m shows moderate hydric dilatation and a very low hygric swelling. Only small traces of clay minerals of mixed layer I-S (R0) and muscovite/illite could be found.

The highest CEC-Value with 17.6 meq/100g was measured for the Tenayocátetl tuff. The tuff has only a moderate hydric and hygric dilatation but a high amount of smectite.

4 Discussion

The CEC-value describes the capacity to hold cations. Larger CEC values indicate that a stone material has a greater amount of swellable clay minerals, which normally has an influence on the moisture expansion. In the presented study a correlation of the CEC-values and the moisture expansion does not appear (Fig. 5).

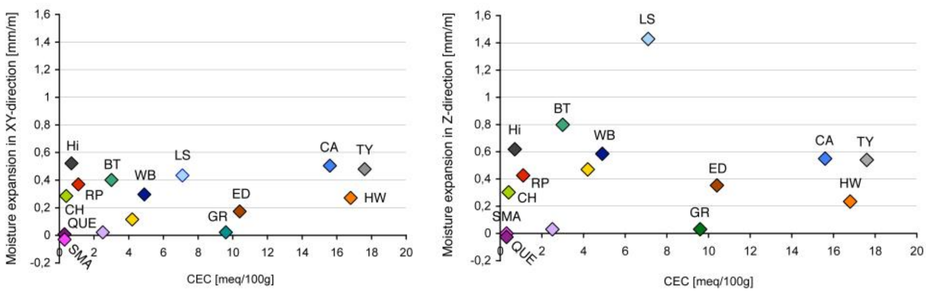

Fig. 5 Moisture expansion in all directions doesn't show a clear correlation.

Water uptake took place due to the capillary active pores. Compared with the microporosity, the averaged *w*-value should show a negative correlation. A tendency of correlation is recognizable (Fig. 6a). This correlation did not continue with reference to the relation between *w*-value and moisture dilatation (XY), which suggests that other properties also should be contemplated (Fig. 6b).

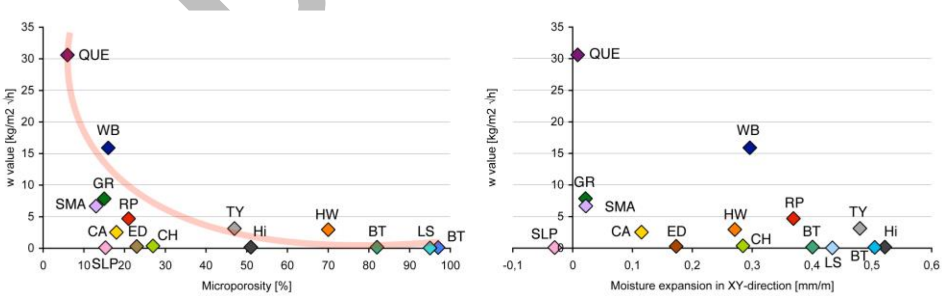

Fig. 6 a) The *w* value and the microporosity show a correlation (line). b) This tendency was not possible to carry forward in comparison of the *w* value and the moisture expansion in XY-direction.

A clear correlation is given for the microporosity in comparison to the hydric expansion (Fig. 7). In particular, a correlation can be detected for the hydric expansion in the XY-direction, probably because the swelling process is not affected due to the possible bedding structure of the stone material (Fig. 7a).

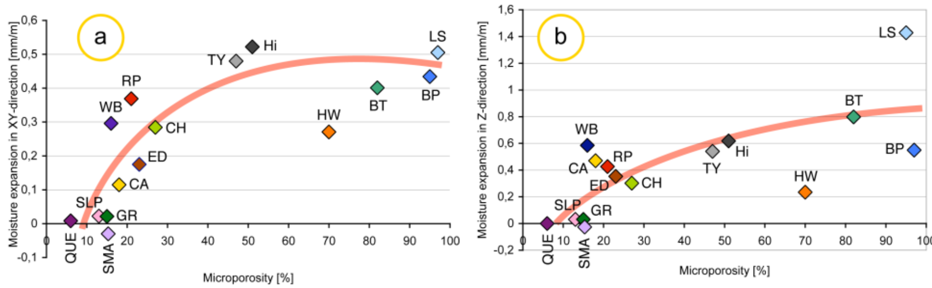

Fig. 7 a) The Moisture expansion in the XY-direction and the microporosity show a clear correlation (line). b) Only a relative weak tendency of correlation is recognizable for the Z-direction (line).

8. Conclusion

According to hydric expansion measurements, values range between 1.4 mm/m to even a reduction of length. Mostly, the expansion perpendicular to the bedding is the largest, which is due to the bedding structure.

The study underlines the importance of the pore space properties within the rock material for moisture expansion. The results show that the amount of micropores plays a decisive role in hydric and hygric swelling, while the amount of clay minerals seems to be less important.

Further studies, mainly about the clay-fraction (e.g. the total amount and type of swellable clay minerals, XRD of oriented slides and CEC-analysis), will help us to clarify the role of the presence of 1:1 or 2:1 layered clay minerals in determining tuff and its relationship to moisture expansion. Also mineralogical aspects and the cementation of the material seem to play an important role, affecting moisture expansion. Therefore further studies have to be done on the petro-physical properties of the investigated volcanic rocks.

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