

Natural analogues of bentonite reaction with alkaline groundwater: overview of the Bigbiga field area, Luzon, Philippines

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Abstract

Bentonite is one of the more safety-critical components of the engineered barrier system for the disposal concepts developed for many types of radioactive waste. However, bentonite – especially the swelling clay component that contributes to its essential barrier functions – is unstable in the high pH leachates from structural cement used in repository construction. Recently, therefore, there have been extensive efforts to better understand the interactions of alkaline fluids with bentonite, coupled with studies aimed at reducing the risk by development of low alkali cement formulations. Laboratory studies of these reactions are bedevilled by very slow reaction kinetics, so it was decided to look for natural analogues of potential alkali fluids/bentonite reaction, examples of which can be found most easily in ophiolites.

Literature surveys and preliminary field campaigns in the Philippines highlighted several sites worthy of further, more detailed study. The first priority site in the Mangatarem area of Luzon provided clear examples of fossil alkali groundwater systems reacting with bentonites in the past and, although useful indicators of the limited degree of alteration to the bentonite, they are of limited value as temporal data are missing. It was therefore decided to shift focus to a suspected site of ongoing bentonite reaction in alkali groundwaters at Bigbiga, some 35 km further south of Mangatarem.

This note provides a brief overview of:

- the overall natural analogue approach to examining potential bentonite reaction in alkali fluids
- the background information which was available on the Bigbiga area prior to detailed sampling
- the considerations behind the two drilling and trenching campaigns carried out to date

Finally, some thoughts on the likelihood of success following further work at the site are provided with a list of recommended future activities at the site.

Introduction

1.1 Background

As noted in Alexander & Milodowski (2011), bentonite is one of the more safety-critical components of the engineered barrier system for the disposal concepts developed for many types of radioactive waste. However, bentonite – especially the swelling clay component that contributes to its essential barrier functions – is unstable at high pH (cf. Savage et al., 2010). This led to some repository designs (especially for disposal of high-level vitrified waste, HLW or spent fuel, SF) that exclude use of concrete from any sensitive areas containing bentonite, due to the fact that it reacts with groundwater to produce leachates with an initial pH of up to about 13 (cf. Haworth et al., 1987).

Such an option of avoiding the problem by constraining the design was considered acceptable during early, generic studies but, as projects move closer to implementation, it is increasingly recognised that constructing extensive facilities underground without concrete – a staple of the engineering community – would be difficult, expensive and potentially dangerous for workers. This last point is especially important given the extreme sensitivity of most stakeholders towards even ‘conventional’ accidents at nuclear sites (see discussion in Alexander et al., 2007). This is especially the case in countries like Japan and the UK, where a volunteering approach to siting a repository means that repository construction could be in a technically challenging host rock.

A further area of concern involves transuranic wastes (TRU, or other high toxicity/long half-life intermediate-level waste, ILW), particularly if this is co-disposed with HLW/SF. TRU waste contains large inventories of cementitious materials and hence, in principle, could pose a risk to the engineered barrier system (EBS) of HLW/SF, even if concrete was excluded from the HLW part of the repository (e.g. scenarios discussed in Nagra’s Projekt Entsorgungsnachweis; Nagra 2002). Indeed, some designs of the EBS for various kinds of low- and intermediate level waste (L/ILW) include a bentonite layer, which is planned to act as an external barrier around concrete structures. To date, there has been no comprehensive demonstration that the performance of such a barrier can be assured for relevant periods of time.

Recently, therefore, there have been extensive efforts to better understand the interactions of alkaline fluids with bentonite, coupled with studies aimed at reducing the risk by development of low alkali cement formulations. The greatest challenge is bringing the information produced by laboratory (conventional and underground rock laboratories, URL) and modelling studies together to form a robust safety case. This is complicated by the inherently slow kinetics of such reactions and the commonly observed persistence of metastable phases for geological time periods (for a good overview of the issues involved, see Metcalfe and Walker, 2004). Clearly, this is an area where natural analogues could play a valuable role – bridging the disparity in realism and timescales between laboratory studies and the systems represented in repository performance assessment (see also discussion in Alexander et al., 1998).

1.2 Natural Analogue Concept

Although systems representative of leachates from both OPC (i.e. groundwaters with pH 12.5 and above) and low alkali cements (groundwater pH of 10 to 11) have previously been examined as natural analogues of cementitious repositories, neither produced data of relevance to the questions now being posed with regard to low alkali cements. For example, the natural cements in Jordan are closely representative of OPC-based materials (see Alexander, 1992; Linklater, 1998; Smellie, 1998; Pitty and Alexander, 2011), so the pH is too high for the current purpose. An appropriate low alkali water chemistry was studied in Oman (McKinley et al., 1988), but this project did not include any investigation of bentonite/leachate reaction as this was not of concern at the time.

There are a number of locations worldwide where such an analogue might be found, including Oman (e.g. Neal and Stanger, 1983), California (e.g. Barnes and O'Neill, 1969), Papua New Guinea (e.g. Barnes et al., 1978) and the Philippines (e.g. Alexander et al., 2008a,b). Based on a multi-attribute analysis, considering factors such as probability of finding suitable locations, relevance to Asian programmes, opportunity for training, low risk of disrupting calls for volunteer sites in Japan and cost-effectiveness, the Philippines was chosen as the focus for this new study. After preliminary field studies investigated several potential sites across the Philippines (Alexander et al., 2008a,b), the Zambales ophiolite in Luzon (Figure 1) was chosen as the focus of the new study.

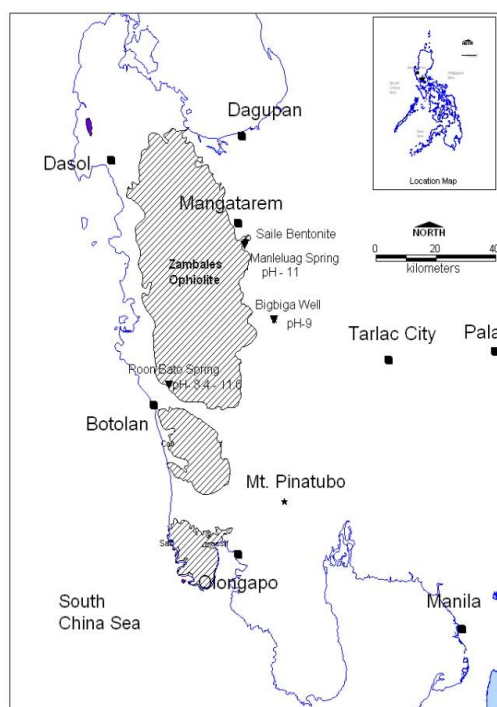


Figure 1: Regional setting of the Zambales ophiolite, showing the Acoje Block in the northwest. The Cote Block is to the south of this, separated by a major fault line. The main groundwater sampling areas are marked on the map. After Abrajano and Pasteris (1989).

Currently, the technical focus is on finding appropriate sites where the following items could be examined:

- long-term bentonite stability in contact with water analogous to low alkali cement leachates
- if possible, same system as above interacting with a range of leachate chemistries as the precise situation in a repository will depend both on the site conditions and the composition of the cementitious materials – neither of which have been fixed as yet
- evidence for gas transport through fractures in the ophiolite and potential blocking of gas transport by the bentonites

1.3 Geological setting

All alkaline groundwaters studied so far in the Philippines originate from the 20 or so known ophiolite bodies which are widely scattered throughout the archipelago (e.g. Yumul, 2007).

The definition of an ophiolite used here is as defined at the Penrose Conference of 1972 (Geotimes, 1972) and, briefly, includes:

deep (abyssal) marine sediments (including bentonites)

pillow lavas (basalt)

sheeted dyke complex

high level/isotropic gabbro

layered mafic cumulates (gabbro)

layered ultramafic cumulates

transition zone dunites and residual peridotites

If any of the above lithologies is missing, it should be called an ophiolite *complex*, but this term is frequently misused in the literature. The alkaline pH values (generally between pH 10 and 11) observed in the groundwaters are a product of the serpentinisation of the ophiolites, a reaction which is discussed in detail in Alexander and Milodowski (2011, 2013). The serpentinite mineral assemblages are very strongly reducing and the hyperalkaline waters in the Philippines are generally effervescent with H_2 and/or CH_4 gas (Figure 2), with up to 5,000 ppm CH_4 and 1,500 H_2 ppm measured during previous field work at Mangatarem. Some of the reaction pathways are also strongly exothermic, frequently producing hydrothermal groundwaters (which are often used as therapeutic springs in the Philippines).



Figure 2: Manlalueg Hot Spring, Zambales ophiolite, showing gas (H_2 and CH_4) emanating from a fracture in gabbro (from Yamakawa et al., 2009)

1.4 Why Bigbiga?

Why choose to sample at Bigbiga and not elsewhere in Luzon? To date, the Philippines Natural Analogue Project (PNAP) has been successful in identifying several sites in northern Luzon where ‘fossil’ bentonite/alkaline groundwater reaction zones can be studied. These are in the Saile quarry in the Malabobo area, Mangatarem and have been described elsewhere (e.g. Alexander et al., 2009; Arcilla et al., 2009; Honrado et al., 2009; Fujii et al., 2010; Fujita et al., 2010) and so will not be discussed further here. To obtain information on reaction rates, however, it is necessary to compare the data from the fossil systems with those from a ‘live’ system, in other words, one where bentonite interaction is currently ongoing today and which can, by a range of methods (cf. Alexander et al., 2012a), be dated.

Unfortunately, despite the clear presence of alkaline groundwaters in the vicinity of the bentonite deposits at Saile, several trenches and numerous boreholes have found no indications of an active system. This is likely due to two main reasons: first, reference to similar geological environments (e.g. Bath et al., 1988; McKinley et al., 1988; Neal and Shand, 2002; Milodowski et al., 2009; Alexander and Milodowski, 2011, 2013; Alexander et al., 2012) indicate that the alkaline groundwater flow is highly likely to be restricted to channels in a few active fractures and, second, this structural control is very difficult to identify in the heavily forested land of the Mangatarem area (it is a protected National Forest Park, so also greatly limiting possible drill sites). In addition, the seemingly most promising sites of presently-active alkaline groundwater flow, such as the Manleluag and Poon Bato Springs, have no relevant clays (smectites) present in the vicinity (Vargas et al., 2011; Alexander et al., 2012b).

In contrast to the National Park, the open scenery with few trees of the Bigbiga area have allowed the development of a better understanding of the structural controls on groundwater flow in the area (e.g. Arcilla and Pascua, 2010, to be discussed below). Finally, in a MAA (Multi-Attribute Analysis) carried out after a preliminary field campaign (see McKinley and Alexander, 2007, for details), the Bigbiga site came a close second to the Saile area. In addition to the above noted points, the logistics of sampling were good - amenable land owners meaning no excessive access restrictions, reasonable site access for drilling equipment and a welcoming local village for supplies. In conclusion, Alexander et al. (2008a) noted “The results clearly indicate the strengths of the Mangatarem site, but also indicate that further study of the Bigbiga site would be useful.” This has been followed up the last two FY with further, more detailed examination of the site and the results are presented below.

When viewed in this light, the actual site drilled was an excellent compromise: although it was not possible to find a site closer to a known active alkaline groundwater source, the area drilled is certainly within an area of active, low temperature serpentinisation as can be observed from the numerous alkali seeps and typical alkali-related tufa deposits in the Pillow Lavas (PLV) across the Aksitero River (i.e. on the western side) from the village (Figure 3).

2 Information available on the Bigbiga area prior to drilling

2.1 Regional geological setting

The Middle Eocene (44-48 Ma; Encarnación, 2004) Zambales ophiolite (i.e. the complete ophiolite suite of the 1972 Penrose classification is present; see Geotimes, 1972, for details) is classified as Tethyan and SSZ (i.e. supra-subduction zone), showing clear subduction-related geochemical signatures (e.g. presence of chromium-rich spinels, calcic plagioclase, pyroxene crystallising before plagioclase etc). It extends approximately 120km N-S and 35km E-W and the eastern part dips at $\sim 30^\circ$ to the east (Schweller et al., 1984) and has been encountered in drillholes below Mount Pinatubo (Delphin et al., 1996) to the southwest (see



Figure 3: Pillow lavas on the western side of the Aksitero River near Bigbiga village. The flat land in the foreground marks the bentonite. New alkali seeps were observed on the hillside behind the small wooden hut during the 2012 field season

Figure 4). Several Plio-Pleistocene andesitic volcanoes have punched through the southern end of the ophiolite (de Boer et al., 1980) and are present from in and around the Saile bentonite quarry in the north to the Bigbiga area in the south (see, for example, Figure 5).

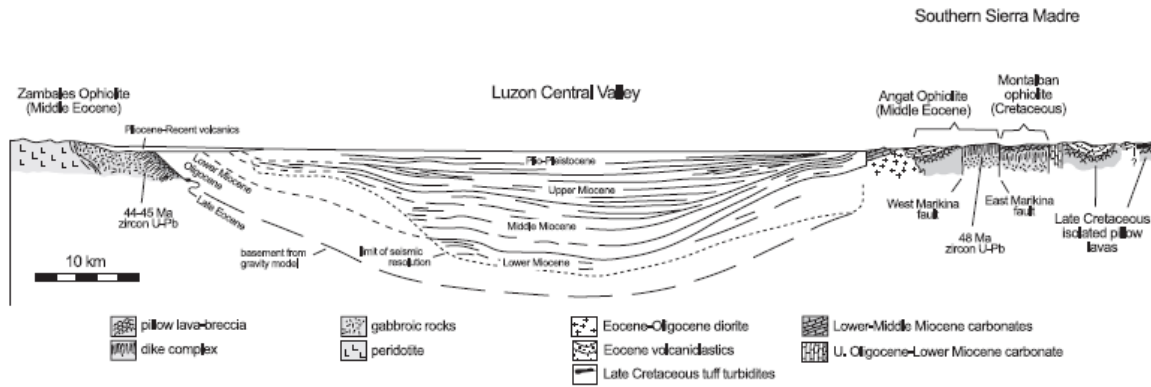


Figure 4: Cross section (from the Zambales ophiolite in the west across the Luzon Central Valley to the Southern Sierra Madre) showing how the ophiolite dips to the east under the Central Valley (from Encarnación, 2004).



Figure 5: To the south of Bigbiga, a typical Plio-Pleistocene andesitic volcanic plug which punches up through the eastwards dipping ophiolite and the alluvial sediments of the plain

The Zambales ophiolite consists of three volcanic-hypabyssal units (Yumul and Dimalanta, 1997): the Cote Block volcanic-hypabyssal rocks, the Cote dykes and the Acoje Block volcanic-hypabyssal rocks (Figure 1), with the Cote Block being of most relevance to this project. A significant body of work exists on the Zambales ophiolite (e.g. Rossman et al., 1989; Tamayo et al., 2004) and most is focussed on the northwestern Acoje Block (no doubt prompted in part by the rich mineral reserves in the area). For example, according to Sturchio et al. (1989), serpentinisation of the rocks of the Acoje Block occurred at temperatures between 30 and 350°C, but no data are available for the Cote Block for comparison. Samples have been collected from the pillow lavas, both near the Manleluag Spring and further west

and south, to assess this for the Mangatarem area and to evaluate the serpentinisation pathway (REF).

Much less work has been carried out on the sedimentary sequences in the area. In brief, it has been interpreted as part of upper tuffaceous turbidites, interbedded with basal micritic limestone of the lower clastic member of the (late Eocene) Aksitero Formation. The upper member is early to late Oligocene and consists mainly of interlayered limestone and clastics, generally indurated, highly calcareous calcirudites, calcisiltites and calcilutites.

This low-dipping, deep marine sequence of about 200m total thickness has been reported to both conformably (e.g. Schweller et al., 1984) and unconformably (e.g. Ugalde, 1999) overlies the pillow lavas and gabbros of the ophiolite and it was important to establish which view was correct. If the association is conformable, then the bentonites are almost certainly the product of marine alteration of tuffs and as old as (or just slightly younger than) the ophiolite (in particular, the pillow lavas) which would then provide a firm baseline from which to assess the bentonite reaction with alkaline leachates. If, however, the relationship was unconformable, then the bentonite history would be much less clear – they could even be related to the Plio-Pleistocene plugs, for example, and so be potentially produced by sub-aerial weathering. In this case, more effort would be required to assess their geological evolution.

Examination of the Aksitero Formation in a range of localities (in particular at the type locality at Bigbiga – see Figure 6) makes it quite clear that the relationship is conformable. For example, basal limestones were observed filling spaces in and around the pillow lavas and breccia and penetrating up to several metres into fractures and cooling joints. In places, clays lie directly on the basalts, for example at the Formation type section in the Aksitero River valley, south of Bigbiga (see Figure 2 in Schweller et al., 1984). Although exposure is generally poor, the confirmed extent of the formation is some 35km, north to south, and several hundred metres east to west. Still unconfirmed is a possible outcrop on the north side of the Cote Block (near Barlo), noted by Schweller et al. (1984).

Moving from the base of the Aksitero Formation and up into the overlying Moriones Formation, it is possible to follow the onset of deep (reportedly >1000 m, but certainly less than the calcite compensation depth), basinal sedimentation on the recently formed pillow lavas. This changes through a slow decrease in water depth (and move towards shallow water deposits including clasts of the ophiolite, wood, corals etc) as the ophiolite is uplifted and exposed (on accretion at the Manila Trench), leading to the current situation of erosion of the ophiolite contributing to infilling the Central Valley of Luzon to the east (cf. Figures 1 and 4).

It would be useful to have an estimate of the rate of uplift on the ophiolite to constrain the onset of leachate/bentonite interaction. Estimates exist in the literature (e.g. Encarnación, 2004) and these could be checked by examination of gabbro samples from the Bigbiga area. In addition, a geomorphological study of the area would provide independent information on the onset of leachate/bentonite interaction, as has been carried out for the Troodos ophiolite in Cyprus (see Alexander et al., 2012a and Alexander and Milodowski, 2013 for details).

Mineralogically, the bentonite has been reported to be montmorillonite with quartz and some glass shards (Ugalde, 1999) with CEC of 38.0 meq/100 g of Ca and 1.22 meq/100 g of Na (Fernandez, 1999) – i.e. it is a Ca-bentonite. However, a detailed mineralogical and chemical analysis is ongoing on a suite of samples which have been collected from around the Bigbiga area to provide an idea of the homogeneity of the material and to compare with the data from the Saile quarry area (cf. Alexander et al., 2009). This will also provide a baseline of the

unreacted mineralogy to compare with those samples which were collected at sites where it is suspected that interaction may have taken place (e.g. the ‘fossil’ reactions from the Saile quarry; see Fujita et al, 2010, for example). Physico-chemical characteristics should also be measured (as has been done on samples from Saile: [REF](#)) so as to allow comparison of the pristine and reacted bentonites with those planned to be used in Japanese repository projects (e.g. Figure 7).

2.2 Geology of the Bigbiga site (based on preliminary field observations)

The gabbro and diabase dyke complexes of the Zambales ophiolite were observed in the Manlelueg area (e.g. Alexander et al., 2008) and, by analogy, were presumed to lie further west than the main Bigbiga study area. This was shown to be the case in the 2012 field season (Figure 8). The general topography of the area can be seen in Figures 9 and 10 and preliminary field investigations indicated that the bentonite generally underlay the rice paddies, lying conformably on the pillow lavas which are exposed further west as the low hills which rise above the paddy fields.



Figure 6: The Aksitero Formation type locality in the Aksitero River at Bigbiga in 2008 where the Formation can clearly be seen to lie conformably on the underlying pillow lavas (from Alexander et al., 2008a: note water buffalo standing in river for scale). This section was buried by a land slip following flooding in 2009 and is currently inaccessible

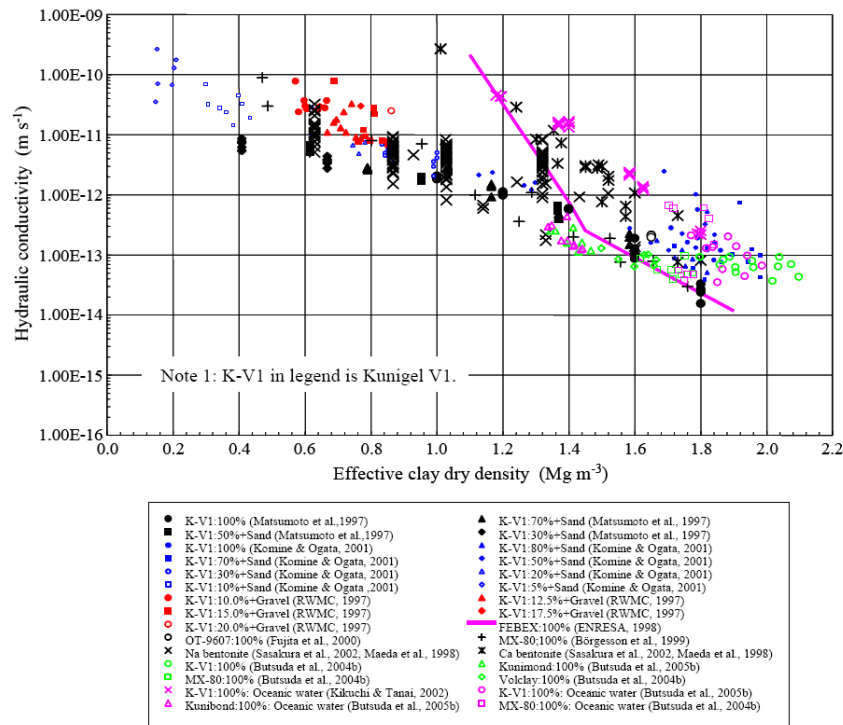


Figure 7: Relationship between effective clay dry density and hydraulic conductivity for various types of bentonite (from JAEA, 2007)

Pillow Lava Basalts and Autobreccias

The pillow lava basalts are observed along the streams, trails and excavations in the vicinity of Bigbiga village. The slightly weathered to fresh pillow lavas exhibit sub-rounded blocky to sub-rounded piles that are cemented by limestone and pink chert. Autobreccias occur at the top of the pillow lavas and the contact with the overlying pelagic sedimentary rock. Petrographic analysis of the pillow lavas in the Manlelueg area (Vargas, 2008) show that the rock is composed of 50% labradorite, 25% pyroxene, 5 % amphibole, <5% K-feldspar and quartz and ~15% opaque minerals.

The rock is aphanitic and glassy. Ophitic textures are observed in plagioclase cutting across pyroxene laths and plagioclase replacing pyroxene is also observed. At Manlelueg, gas has been observed emanating directly from fractures in the gabbro, a km or so from the bentonite (e.g. Yamakawa et al., 2008) and at Bigbiga, gas and alkali groundwater has been observed leaking from fractures in the pillow lavas (just west of borehole BBW5 at E120°22'43.4", N15°31'05.6") some 50 m from the bentonite.

Figure 8: Deeper members of the ophiolite are exposed several kms westwards from Bigbiga village

Aksitero Formation

The Aksitero formation was named after the series of rocks consisting of limestone with thin tuff layers and tuffaceous fine grained turbidite members with marly limestone interbeds found at the Aksitero river in Bigbiga (see Figures 6 and 9). The limestone member is called the Lower Bigbiga Limestone member and the upper member is the Upper Burgos member. The total thickness of the Aksitero at the type section is slightly >100 m (Schweller et al., 1984). The limestone member is ~42 m thick while the overlying turbidite member is up to 78 m thick.



Figure 9: Aerial view over the Mayantoc Field Centre, Bigbiga, of the University of the Philippines, looking west towards the pillow lavas rising above the paddy fields. Bentonite and associated sediments underlie the paddy fields (a worked out bentonite quarry can be seen on the west side of the river, immediately opposite the Field Centre)



Figure 10: Bigbigan village, looking southwest, the flat-lying paddies of the foreground conceal the majority of the bentonite. Note the low-lying pillow lava hills to the west and southwest middle ground with the higher hills in the far background marking diorite and gabbro lower in the Zambales ophiolite



Figure 11: The bentonites outcrop along both sides of the Aksitero River which lies on the west side of Bigbigan village. View approximately WSW

At Bigbiga, the Aksitero formation conformably overlies the pillow lavas and outcrops generally consist of medium to thin alternating beds of calcareous tuffaceous sandstone, siltstone and claystone. Colours range from buff to white to pale green. Some beds are compact with conchoidal fractures, while other beds are friable. The bentonites have been quarried on a small scale locally and otherwise outcrop along the river (Figure 11). Based on nannofossil studies (details in Aurelio and Pena, 2004), the age to the Aksitero Formation is between, Late Eocene to Lower Oligocene. The sample at ‘Suaco’ examined by Schweller et al. (1984) is dated Lower Oligocene and is still part of the Bigbiga Limestone member of the Aksitero Formation.

In the Saile quarry area, the bentonite is known to be fractured in places and similar fracturing has been identified at Bigbiga (e.g. Figure 12). However, unlike at Saile, the fractures are clearly water-conducting at present. Although buffering reactions would appear to have already reduced the pH to near-neutral, the groundwater temperature (30°C) is more representative of the deeper groundwaters from boreholes BBW1-4 (29-32°C) than the river water (26°C in mid-October 2011 during the sampling pictured here), probably indicating a deeper source (this could be checked with stable isotopes and groundwater dating).



Figure 12: Water-conducting fractures in bentonite at the west side of the river, Bigbiga

Moriones Formation

The Moriones formation is named after a sequence of sedimentary rock consisting of interbedded sandstone, shale, conglomerate and minor limestone found along the Aksitero river, unconformably overlying the Aksitero Formation (see Figure 13). This formation has an estimated thickness between 1,000 to 1,500 m (Aurelio and Pena, 2004) and is Early to Late Miocene in age based on nannofossils. Although known to be present from previous studies of the area, the Formation has not yet been examined in detail by the field team at Bigbiga due to the limited field time available so far.

2.3 Structural controls

The Bigbiga area has been the scene of a recent aerial structural survey (Figures 14 and 15) and this has identified several likely dominant structural controls on the deep groundwater flow in the area (Arcilla and Pascua, 2010; cf. Vargas, 2008, for the Manleluag area).



Figure 13: Contact of the Aksitero (bottom half of view) and Moriones (top half) Formations in the Saile quarry. Note the cobbles and coarse sandy nature of the matrix at the base of the Moriones Formation (from Vargas, 2008)



Figure 14: An UP crew conducting an aerial photo survey of the Bigbiga area, summer, 2009

Of direct relevance here are the roughly N-S trending structures which can be seen to control much of the surface flow patterns. This structure is exemplified on the land of the Field

Centre where one seep (BBW1) and three producing boreholes lie on one such lineation. A further borehole (BBW5) lies on the same lineation, just a few 100 m to the north of the Field Centre. **I can't say much more on this, never seen the report....**

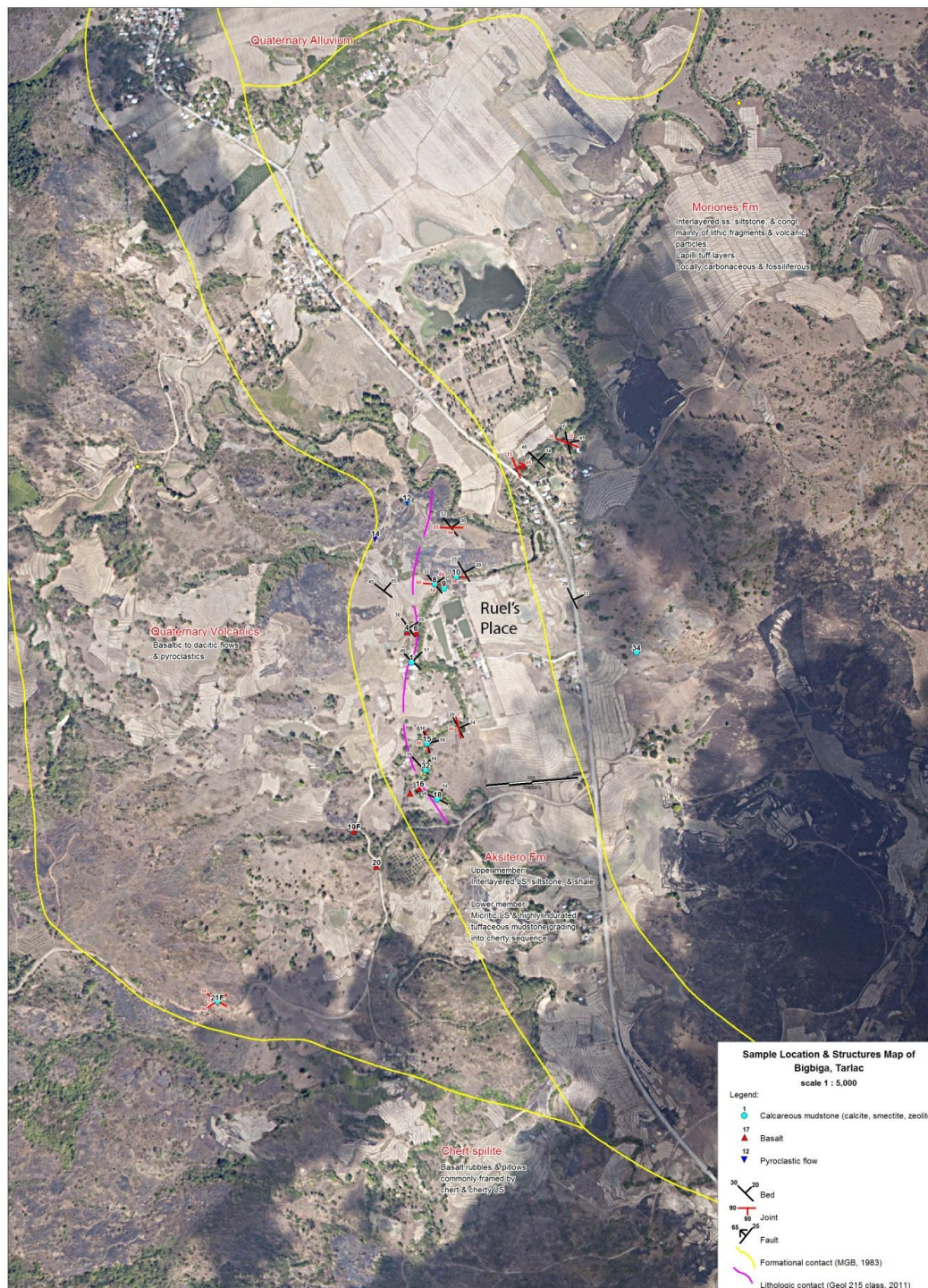


Figure 15: Lithology of the Bigbiga area based on UP field work and the structural analysis of Arcilla and Pascua (2010)

2.4 Hydrochemistry

In addition to samples taken at the surface of the bentonite (e.g. Figure 12), deeper groundwaters have also been sampled from the suite of boreholes (BBW1 to 4, moving from the southwest to the northeast) at the Mayantoc Field Centre and the analytical data from these are presented in Table 1.

Detailed modelling of the two datasets presented in Table 1 has not been carried out here due to the significant differences in the concentrations measured in the two laboratories (see also the data for boreholes BB-09 and BB-10 in appendix 1). However, in general, when compared with the higher pH systems at Manleluag (e.g. Vargas et al., 2009), the data would tend to suggest there has already been some reaction between the ‘original’ ophiolite-derived alkaline groundwater and the bentonite.

Table 1: selected data from analyses by the Universities of Hokkaido and Keio (all ppm)

Sample name	pH	Ca ²⁺	Na ⁺	K ⁺	Mg ²⁺	F ⁻	Cl ⁻	Al	As	SO ₄ ²⁻
BBW3	9.2/9.6	nd	15.80	0.51	nd	0.57	4.28	nd	nd	51.21
BBW1	8.4	1.21	14.56	0.64	nd	0.39	3.98	nd	nd	41.32
BBW2	9.3	nd	10.92	0.88	nd	2.97	8.22	nd	nd	0
BBW4	6.7/6.9	12.72	28.00	0.86	1.33	nd	72.60	nd	nd	138.09
BBW3	9.1/9.4	0.05	91.20	2.14	1.24	nd	4.04	0.01	0.14	50.22
BBW1	8.4	5.82	83.60	4.32	0.09	nd	4.08	0.15	0.13	44.49
BBW2	9.6	0.80	65.60	3.54	5.29	nd	7.75	0.14	0.15	0.21
BBW4	7.6	87.40	143.00	2.54	5.17	nd	94.91	0.01	0.12	257.51
M1-09-HWP-01	10.89	22.30	22.20	1.72	0.14	nd	18.82	nd	nd	1.08

BBW1 – Keio samples BBW1 – Hokkaido samples M1-09-HWP-01 Manleluag sample for comparison nd – no data

This is implied by the generally lower pH, Ca, Na and K and the higher Mg concentrations than seen in the Manleluag groundwaters. The Al content is slightly higher too, as would be expected, but little weight should be put on these data due to the high probability that Al colloids are present (cf. Alexander et al., 2011). Likewise the SO₄ concentrations as this may simply reflect oxidation effects on the borehole during open-hole sampling.

As a general recommendation, QMS (quality management system) guidelines for groundwater sampling and laboratory analyses should be followed rigorously (cf. Kunimaru et al., 2009, 2010, 2013; Ota et al., 2009) to allow meaningful inter-comparison of data from different operators.

3 The three drilling and trenching campaigns carried out to date

3.1 The considerations behind the sampling campaigns

In addition to the above presented information which was collected during the preliminary site assessment which showed bentonite conformably overlying pillow lavas from which alkali seeps could be observed emerging in close proximity to the bentonite, other aspects were behind the choice of the Bigbiga area. These included:

- a friendly and open relationship with the person who identified himself as the local landowner on the favoured sampling site on the east side of the Aksitero River
- the presence of alkali groundwaters in boreholes on the above mentioned landowners property. These boreholes appeared to penetrate through the bentonite to the pillow lavas below (although note no corelogs were available)
- the site structure was such that any pillow lava/bentonite contact (and therefore potential zone of reaction) was known to be near-surface and so accessible by shallow boreholes and/or trenches
- identification of several lineations which were thought to control groundwater movement at depth: the clearest being the N-S lineation penetrated by boreholes BBW1 to 4

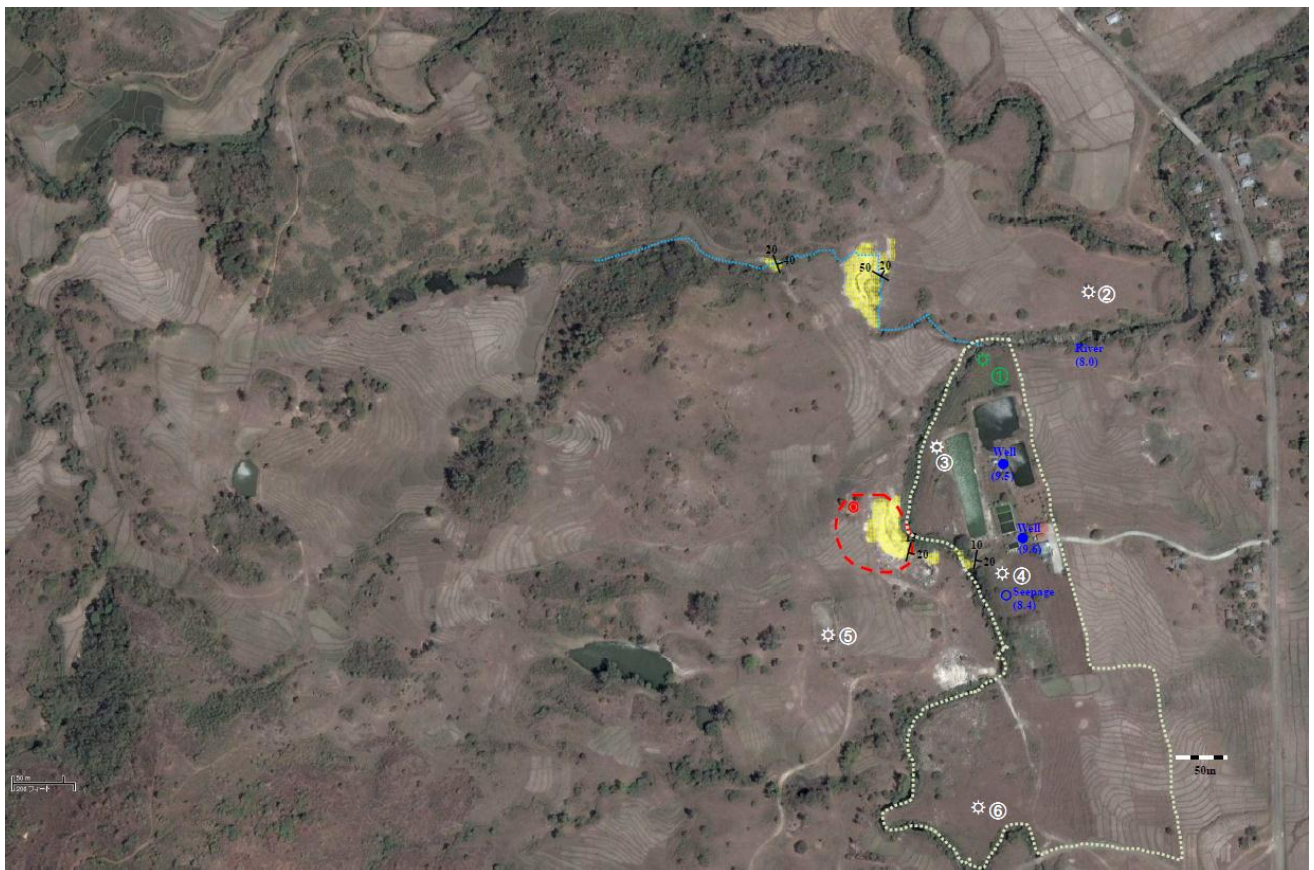


Figure 16: Proposed drill sites for the 2010 field campaign. Yellow dotted line is the farm boundary, red dotted line highlights a worked out bentonite quarry, ● existing borehole or sampling point, ☼ proposed borehole site, ☼ first priority borehole

Consequently, in the autumn of 2010 it was planned to drill research boreholes on the farm at Bigbiga and arrangements were made with the supposed land owner to establish access etc. Unfortunately, the sampling vanguard were effectively thrown off the land when it turned out that the ‘landowner’ was only the true landowner’s brother and consequently had no right to offer the sampling team access to the land. Thus the planned sampling campaign (cf. Figure 16) had to be abandoned and, following frantic negotiations by UP with the landowner of the area to the west of the river, it was decided to drill several boreholes in this area to attempt to access the base of the bentonite. Unfortunately, the structural relationships in this area were less clear than to the east of the river so it was decided to attempt to access the base of the bentonite at reasonably shallow depth based solely on the observed outcrop patterns in the sampling area.

This focussed the drillholes (and trenches) in two main areas: first near the river where outcrops indicated likely areas to successfully access the base of the bentonite and, second, further west where the bentonite presumably thinned out against the outcropping pillow lavas. The decision making was supported by a gas survey which attempted to determine methane and hydrogen leaking from the ophiolite into the soil.

3.2 The gas survey in 2010

The gas survey was carried out across the west side of the river (see Figure 17) in areas of interest identified in the 2010 field programme and highlighted in the structural survey of Arcilla and Pascua (2010). The full details of the methodology is provided in Obayashi (2011), but, briefly, a steel tube is inserted some 30 - 50 cm into the soil and a sample is drawn through the methane and hydrogen meters by small, integral pumps (Figures 18 and 19). In total, some 11 sites were sampled (see Table 2) as part of the site reconnaissance and an additional 5 were sampled post-sampling (including 4 boreholes and 1 trench).

As the results in Table 2 show, gas was only noted at four sites, two of them being in boreholes, one in bentonite, the other in pillow lavas. It also has to be remembered that the methane signatures possibly also include input from the paddy fields, especially at sites BBG08, 13 and 15. This could be checked by re-sampling these sites and analysing the methane isotopically although, even here, the essentially inorganic methane from the ophiolite (cf. Neubeck et al., 2011) can be ‘contaminated’ by additional biological processes in the ophiolite groundwaters (e.g. McCollom and Seewald, 2007). This is less the case for hydrogen gas, but even here, it cannot be simply assumed that it is an indication of abiogenic processes (cf. Sherwood-Lollar et al., 2007). Nevertheless, the fact that sample BBG15 is on a clear fault line which can be traced in the river bank some 5 m away, suggests that the hydrogen measured in this case is ophiolite derived. This also suggests that alkali groundwaters may be present in borehole DH02A, either at greater depth in the pillow lavas or somewhere nearby in the fractured pillow lavas at the top of the pillow lava pile (see below). As such, this site may be worth further, more detailed, examination at a later date.

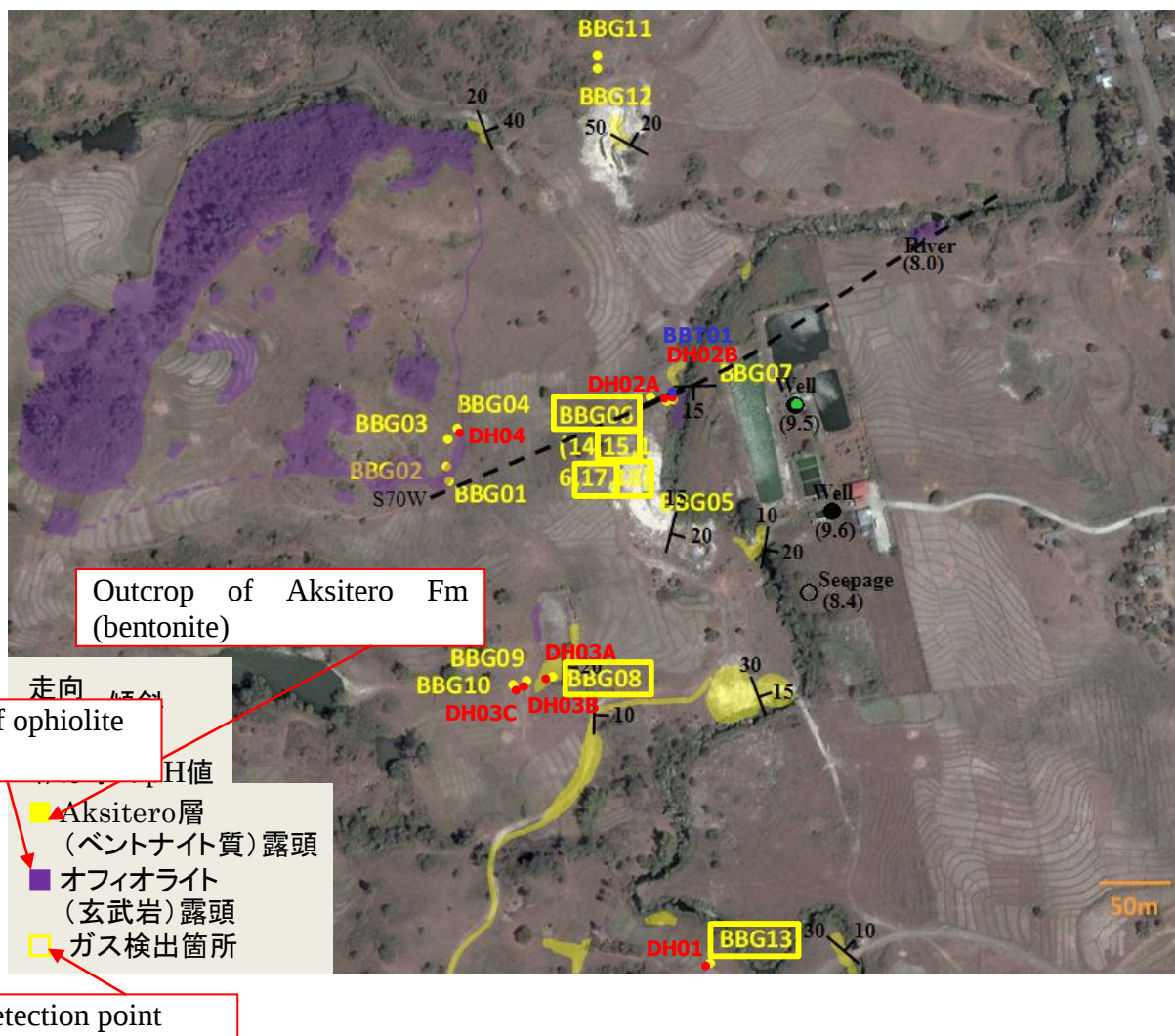


Figure 17: Gas survey sampling positions during the 2010 field campaign (from Obayashi, 2011)



Figure 18: Gas sampling at Bigbiga: soil gas analysis via steel probe rammed into the ground, left, sampling in thin soil over pillow lavas, right



Figure 19: gas meters deployed to sample gas with steel probe into the soil, left, or in a borehole, right

Table 2: Results of gas survey at Bigbiga area (Obayashi, 2011)

	Concentration (ppm)		date	Classification※	geological condition	Remarks
	CH ₄	H ₂				
BBG01	0	0	11/8	①	Surface soil (on basalt)	On assumed fault system
BBG02	0	0	11/8	①	Surface soil (on basalt)	On assumed fault system
BBG03	0	0	11/8	①	Surface soil (on basalt)	
BBG04	0	0	11/8	①	Surface soil (on basalt)	
BBG05	0	0	11/8	③	Aksitero	BBT01 (Trench 1)
BBG06	140	0	11/8	①	Surface soil (on Aksitero)	On assumed fault system
BBG07	0	0	11/8	①	Surface soil (on Aksitero)	On assumed fault system
BBG08	530	130	11/8	②	basalt	DH03A
BBG09	0	0	11/8	②	basalt	DH03B
BBG10	0	0	11/8	②	basalt	DH03C
BBG11	0	0	11/8	③	Aksitero	On the contact (dry)
BBG12	0	0	11/8	③	Aksitero	On the contact (wet)
BBG13	560	6	11/8	②	Aksitero	DH01(in the heavy rain)
BBG14	0	0	11/10	①	Surface soil (on Aksitero Fm)	On assumed fault system
BBG15	103	60	11/10	①	Surface soil (on Aksitero Fm)	On assumed fault system
BBG16	0	0	11/10	①	Surface soil (on Aksitero Fm)	On assumed fault system

DH = drillhole, ① = measurement by following photographs ② = measurement at borehole mouth ③ = measurement at outcrop

3.3 The drilling and trenching campaign 2010

3.3.1 Introduction

As noted above, the 2010 drilling campaign had to be changed with little warning and so borehole positioning was done on the basis of field observation and the Arcillo and Pascua (2010) structural analysis. In the event, a total of 7 boreholes (Figure 20) were drilled (Figure 21), reaching the bentonite/pillow lava interface at between 1.5 and 5 m depth (Figure 22).

3.3.2 Drillcore description

Below a generally thin soil layer, the Aksitiro Formation on the west side of the Aksitiro River are characterised by massive limestones (some clay enriched - see Figure 23) with thin, interbedded tuffs and tuffaceous sandstones. In all these cores, the base of the Aksitiro lies conformably on the pillow lavas and the base displays some form of bleaching.

As has been observed in Cyprus, the top of the pillow lavas is often highly fractured and appears to be the likely conduit of groundwater flow. This is certainly the case in Cyprus, where this zone is a significant aquifer across much of the island (Alexander et al., 2012a). Interestingly, this zone at Bigbiga also includes frequent zones and blebs of serpentinised pillow lava, suggesting recent/ongoing production of alkaline groundwaters at shallow depth. This is consistent with field observations in the 2012 programme where tufa deposits were noted at the top of the pillow lava hill immediately west of borehole DH04.



Figure 20: Position of the 7 boreholes (DH) drilled during the 2010 field campaign (Obayashi, 2011)



Figure 21: small diameter drilling machine on-site at Bigbiga. Borehole DH01, left and borehole DH04, right

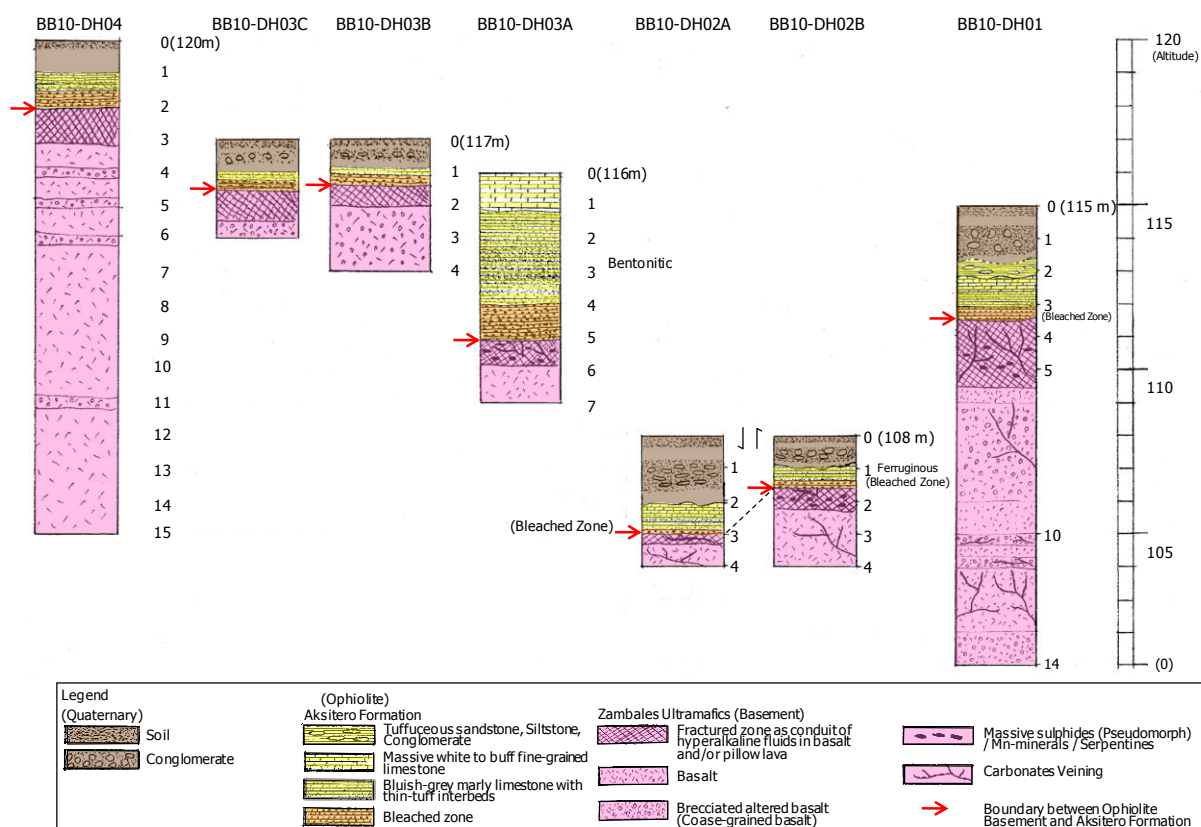


Figure 22: Borehole logs from the 2010 field sampling campaign (Obayashi, 2011). Note the significant variation in the depth of the top of the pillow lava and in the thickness of the Aksitero Formation

It is not impossible that these fluids represent deep groundwaters, but the erosion surface suggests isolated, localised, shallow production triggered by rainwater infiltration in the lavas. This could be checked by short boreholes instrumented with peizometers at the hilltop to establish if there are any indications of deep flow emerging at the hilltop via fractures. The head for such flow is almost certainly present due to the Zambales peaks to the west, but localised flow seems more likely at present.



Figure 23: 0.04 m diameter drillcore of blue-grey marly limestone at the base of the Aksitiro Formation, borehole DH01 (Obayashi, 2011)

Of the 7 boreholes drilled, only DH04 produced groundwater in any quantities. This artesian flow had a pH of 6.88 when sampled immediately after completion. More recently, the pH has been **XXXXX**



Figure 24: Trench BBT01 on west side of the Akrotiro River (see Figure 17 for the position) being excavated by hand

What is clear from Figure 22 is that there is a significant topography across the top of the pillow lavas, with about 17 m vertical difference across the 200 m distance between boreholes DH04 and DH02A/B. This is similar to the situation in the much better exposed pillow lavas of the Troodos ophiolite in Cyprus and will be discussed further below.

3.3.3 Trench descriptions

One trench, BBT01, was dug at Bigbiga during the 2010 field campaign (Figure 24), at the site of borehole DH02B (see Figure 17). The borehole appeared to have reached the pillow lavas at approximately 1 m deep. Based on the presence of a normal fault several metres away in the river which has lifted the pillow lavas to the surface, this depth seemed reasonable but, on excavating the trench, it transcribed that the ‘basement’ was in fact a very large basalt boulder. The trench was excavated to 2 m deep before being abandoned with no sign of the pillow lavas.

3.4 The gas survey in 2011

Permission to enter the farm on the east side of the Aksotiro River (see Figure 16) was obtained for field season 2011 and additional groundwater samples were collected from the existing farm boreholes (do we have the results?). In addition, a gas survey was carried out, covering a regular grid from the most northerly borehole on the farm (marked as ‘well’ on Figure 17), running due south to the small spring on the east river bank (incorrectly positioned and marked as ‘seepage’ on Figure 17). The spring is actually just outside the property boundary at the southwest limit of the boundary (see Figure 16).

Very little gas was observed (table of data?) other than at the mouth of the existing farm boreholes and at the seepage (where gas bubbles to the surface), suggesting rather good barrier performance from the bentonite underlying the farm. Sampling was carried out in the rainy season and it could also be the case that the saturated soil smothered any faint signals emanating from other fracture systems in the pillow lavas. As such, it may be worth re-sampling under drier conditions in the future.

3.5 The gas survey in 2012

By the 2012 field season, the farm had been purchased by UP and is now known as the UP Mayentoc Field Centre which is run and maintained by NIGS. This certainly makes property access effortless and provides an excellent logistics centre for any field work in the wider area. The most recent gas survey took place during the 2012 field season as an aid to deciding the positions of the drillholes. The area to the north and west of the farm fishponds (around planned borehole 1 and 3 on Figure 16) was surveyed, with the grid recorded by GPS measurements (Figure 25).



Figure 25: GPS positioning of the gas survey grid at the UP Field Centre

Once again, few gas anomalies were encountered (Figure 26 Table of results available?), but those which were noted were compared with the results of the shallow resistivity survey of

the area (Medina and Arcilla, 2011 - don't have a copy) which was carried out in 2011 and were used to guide the positioning of the drilling rig.



Figure 26: gas sampling at the Field Centre during the 2012 field campaign

3.6 The drilling campaign 2012

3.6.1 Introduction

Three new boreholes were drilled in the 2012 field campaign, all within the boundary of the Field Centre (Figure 27). DH05 and DH07 were positioned on the low ground just above the east bank of the Aksitero River whereas DH06 was effectively on the lineation on which the farm boreholes lie.



Figure 27: Position of boreholes DH05, -06 and -07 at the Mayentoc Field Centre

DH05 and -07 were positioned so as to minimise the depth to the bentonite/pillow lava interface and DH06 was positioned to intercept the lineation. It was decided not to drill DH06 too close to the northern farm borehole in case it damaged, or in some way short-circuited, flow to the borehole and this politically motivated decision obviously compromised borehole positioning.

3.6.2 drilling activity

The boreholes were produced by the same UP drilling team and rig as previously employed (Figure 28 and Table 3). As before, the target was the base of the bentonite, so drilling was stopped shortly after encountering the pillow lavas. Following completion of each hole, they were cased very simply with 58 mm O.D. plastic piping to prevent borehole wall collapse.



Figure 28: UP drilling rig and team drilling borehole DH05

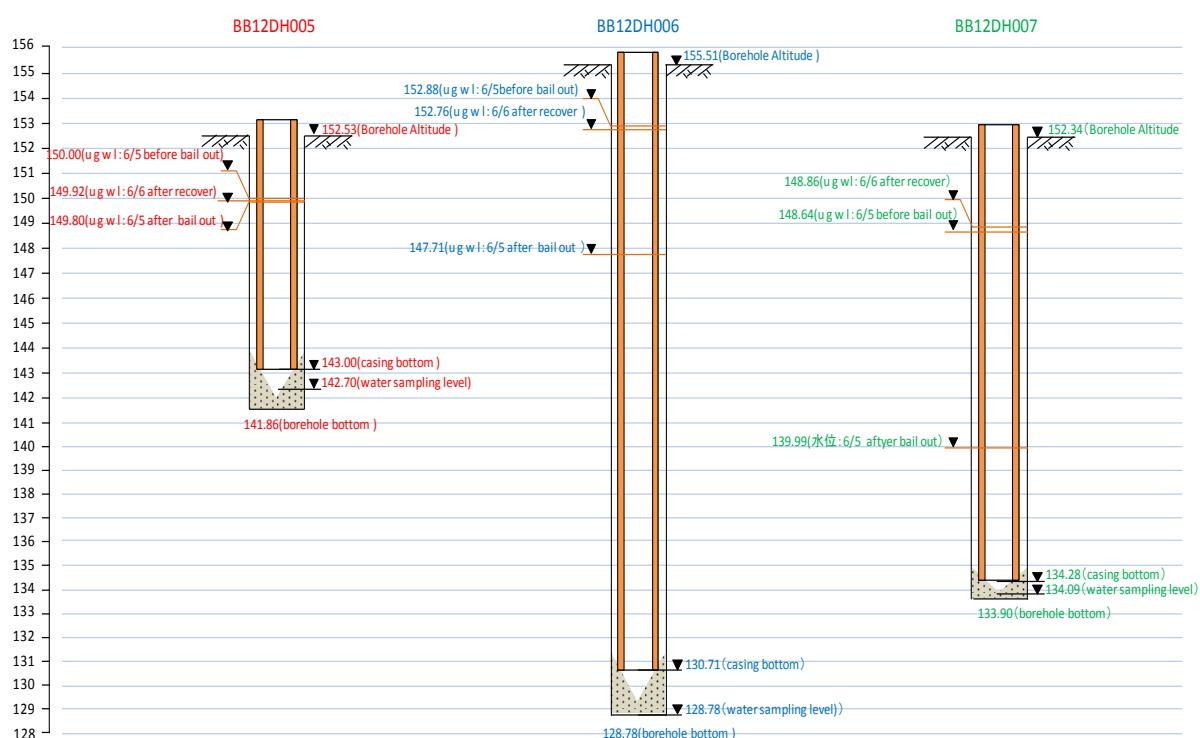


Figure 29: borehole completion details

The space between the liner and the borehole was filled with gravel to stabilise the borehole (Figure 29), but this, of course, does not prevent groundwaters from different levels mixing, so any samples have to be considered as mixed source. Currently, the only analyses available (Table 4) were collected 2 days after completion of the last well and so will also reflect

contamination from the drilling fluid (water from the nearby fishponds) and rock flour (from the drilling).

3.6.3 drillcore descriptions

The drillcores were examined on-site (Figure 30) and the preliminary core description drawn up. Comparison of Figures 27 and 29 also indicates significant topography on the pillow lava surface (cf. section 3.3.2).



Figure 30: Preliminary drillcore description was produced on-site

The detailed core descriptions are shown in appendix 2 and several points are worth noting:

- the bentonite, when present, is generally in thin horizons, but more massive, clay-rich limestones (of the Lower Bigbica unit) are more widespread
- clays are also common in the fault gouge in the fault zone described in borehole DH06 and -07
- the bleached zone present just above the pillow lavas in boreholes DH01 to -04 is also present in DH05, but not in -06 or -07
- the lateral continuity of the sediment beds appears to be highly limited between these three, relatively close boreholes
- there is no obvious alteration of the clays near the base of the sediment column, but this was also the case at Saile (and in Cyprus; see Alexander and Milodowski, 2013)
- in some cases, the pillow lavas are highly altered and calcite veins are common throughout the pillows and brecciated basalt

The bentonite in all 10 boreholes drilled so far at Bigbica is distributed throughout the sediment column and so is unlike the massive bentonites further north at Saile nor at the Parsata site in Cyprus.

Table 3: Drilling information

name		BB12-DH005	BB12-DH006	BB12-DH007
Coordinate (location)		N15°30'52.8",E120°22'51.2"	N15°30'51.3",E120°22'53.5"	N15°30'53.4",E120°22'51.6"
Borehole altitude (m)		152.53	155.51	152.34
Drilling length(GL-mabh)		10.67	26.73	18.44
Bottom altitude (m)		141.86	128.78	133.90
Casing length (m)		9.95	25.15	18.41
length from GL to top of casing (m)		0.42	0.35	0.35
Casing bottom altitude (m)		143.00	130.71	134.28
Watersampling level (m)		142.70	128.78	134.09
Borehole size		NQ(Φ75.3mm)		
Casing OD and ID		OD:φ58mm,ID:φ54mm		
Operating Schedule				
5/29	10:00	Drilling Start	*	*
	18:30	Drilling Completion	*	*
5/30	10:00	*	Drilling Start	*
6/1	17:20	*	Drilling Completion	*
6/2		*	*	Drilling Start
6/3		*	*	Drilling Completion
6/4	10:00	borehole casing insertion		
	11:30	Washing inside of Borehole		
6/5	13:10	*	152.88(before bail)→bail out 55L→147.71(after)	*
	14:10	*	*	148.64(before bail)→bail out 34L→139.99(after)
	14:30	150.00(before bail)→bail out 19L→149.80(after)	*	*
	15:00	*	water level : 152.73,water sampling	*

			NO.W009～W011	
		*	*	water level : 147.64, sampling NO.W012～W014
		water level : 149.99、 water sampling NO.W015～W017	*	*
6/6	12:50	*	water level : 152.76,water sampling NO.W018～W020	*
		*	*	water level : 148.86,water sampling NO.W021～W023
		water level : 149.92,water sampling NO.W024～W026	*	*

Table 4: Mixed groundwater (plus drilling fluid) from boreholes DH05 - DH07

name	DH005				DH006				DH007			
	N15°30'52.8",E120°22'51.2"				N15°30'51.3",E120°22'53.5"				N15°30'53.4",E120°22'51.6"			
	GK- W15	H- W16	GK- W24	H- W25	GK- W09	H- W10	GK- W18	H- W19	GK- W12	H- W13	GK- W21	H- W22
Sampling by	G Univ.	H Univ.	G Univ.	H Univ.	G Univ.	H Univ.	G Univ.	H Univ.	G Univ.	H Univ.	G Univ.	H Univ.
date	6/5	6/5	6/6	6/6	6/5	6/5	6/6	6/6	6/5	6/5	6/6	6/6
pH	6.92	6.74	7.05	6.96	7.17	6.95	7.36	7.31	7.19		7.36	7.29
Temp. (°C)	28.8	28.7	28.9	29.8	29.5	29.7	29.4	29.4	28.7	28.8	28.8	29.6
ORP(mV)	191	15	77	—	60	78	20	—	168	23	161	—
Na		1.37		7.83		3.10		2.98		1.82		2.11
K		0.09		0.39		0.10		0.10		0.08		0.11
Ca		7.62		0.64		3.18		3.65		3.03		3.38
Si		23.93		20.37		9.60		20.54		16.37		21.20
Al		ND		ND		ND		ND		ND		ND
Mn		ND		ND		ND		ND		ND		ND
Mg		0.36		0.09		0.31		0.30		0.13		0.15
Fe		ND		ND		ND		ND		ND		ND
HCO3	—	—	—	—	—	—	—	—	—	—	—	—
NO2		ND		ND		ND		ND		ND		ND
NO3		ND		ND		ND		ND		ND		ND
PO4		ND		ND		ND		ND		ND		ND
SO4		8.99		14.76		0.98		0.86		13.47		11.28
Cl		3.56		6.16		0.26		0.23		3.28		2.51
Br		ND		ND		ND		ND		ND		ND
F		0.54		0.60		0.05		0.06		0.79		0.71

4 Discussion and thoughts on the likelihood of success following further work at the site

The Bigbiga site was chosen to investigate further for numerous pragmatic logistical reasons, but also because of the suspected presence of alkaline groundwaters in the pillow lavas in the area combined with previously reported bentonite quarries and the presence of the Aksitero Formation type locality. Looking at these features again:

Logistics

This remains a strong point for any further work in the area. The development of the Mayentoc Field Centre by UP has improved this significantly. The working relationship with the local community also remains strong, although this should be nurtured further.

Alkali groundwaters

The chemistry of the existing Field Centre boreholes (see appendix 1) is strongly suggestive of alkali groundwaters. However, it looks likely that the boreholes also intercept shallow flow paths (NB presence of cattle faecal material in samples collected by UP microbiologists) and it is likely that dilution with freshwater (probably from the river) is ongoing. The new boreholes in 2012 (DH-05 to -07) appear to have groundwater inflow, but the only samples obtained so far are heavily contaminated with drilling fluid (i.e. water from the fishponds on site – see Table 4). Further sampling could be carried out here, but, once again, the simple nature of the borehole liners (Figure 29) does not allow unambiguous sampling of deep groundwaters, so it is debatable whether this is worth the effort. *Apart from pumping, cleaning and sampling boreholes DH-05 to -07 once, there seems little point in further borehole sampling unless proper sealed liners can be fitted, so bypassing any shallow flowpaths. Alternatively, install a double-packer at depth in the pillow lavas and install another double packer at the bentonite/pillow lavas interface. This will allow sampling of the 'original' alkaline groundwater in the pillow lavas and the probably 'reacted' groundwater at the base of the bentonite. Repeated groundwater sampling should be carried out over a year. This should be considered for boreholes DH-05 to -07, but only after analysis of the marly claystone on top of the pillow lavas (see comments below)*

The boreholes drilled in 2010 and 2011 were all dry, apart from DH04 which has been used to supply the local farmer. Unfortunately, this has not been sampled on a regular basis to check the pH evolution. This would be of great use in developing the site conceptual model. *This could easily be done using pH papers and the farmer could be trained in their use very rapidly.*

There are clear, small scale seeps in the pillow lava exposures in the hills to the west of the river: indeed, new examples were found in the 2012 field season (Figure 31). These may have been visible because of the dry weather at the time accelerating tufa production by evaporation of the alkali waters. It is unclear if these simply represent local production of alkali groundwaters via ongoing low temperature serpentinisation or if the source is at depth. *This could be checked with short boreholes instrumented with peizometers at the hilltop to establish if there are any indications of deep flow emerging at the hilltop via fractures. This would be of great use in developing the site conceptual model.*

Groundwater sampling and analytical standards leave much to be desired with the differences in the produced data (e.g. Table 1 and appendix 1) between the two main sampling groups being astonishingly large. Any project which plans to feed information into a repository safety

case has to produce data which can be trusted and this is certainly not the case at the moment. A full cradle-to-grave QMS for groundwater sampling, sample transport, laboratory analyses and data handling (e.g. Kunimaru et al., 2009, 2010, 2011, 2013) should be initiated to ensure that all steps of the project are of high enough quality that the data can be used to develop repository safety case by the Japanese implementers and regulators. Otherwise, there seems little point in further groundwater sampling.

Figure 31: New tufa deposits from weak alkali groundwater seeps in the pillow lavas west of the river. The hillside above (and due west of) borehole DH04 were covered in new tufa in May 2012. Note rucksack on left of middle ground for scale



Bentonite

While the data on the fossil bentonite/groundwater systems from Saile are very useful, the first priority has to be sampling a 'live' system. The chances that such a system can be captured at Bigbiga remain high, but is the bentonite observed in the area appropriate? There is certainly no evidence of the massive bentonite beds of the Saile area with any material observed to date restricted to thin beds within the limestones and sandstones of the Aksitero Formation (see appendix 2). However, the marly claystone which is observed immediately

above the pillow lavas in boreholes DH-01 to -04 and DH-06 to -07 (and is probably present in -05 too, but much of this horizon has been lost in this borehole) may well contain swelling clays and may be a perfectly viable alternative to ‘true’ bentonite. **Appropriate analysis of the marly claystone needs to be carried out urgently to assess the content of swelling clays.**

If the marly claystone turns out to be an inappropriate alternative, then serious thought must be given to finding an alternative site

Flow through bentonite (see Figure 12): in the groundwaters collected from fractured bentonite in the Aksitero River, although buffering reactions would appear to have already reduced the pH to near-neutral, the groundwater is still warmer (30°C) than the river water (26°C in mid-October during sampling). **This probably indicates a deeper source (cf. temperatures from boreholes BBW1 to 4) and this could be checked with stable isotopes and groundwater dating.**

To allow better comparison with the industrial bentonites planned to be used in Japanese repository projects., it is important that the physic-chemical characteristics of the Bigbiga (and Saile) bentonite should also be measured. **It is suggested that parameters such as swelling pressure, unconfined uniaxial compressive strength, plasticity index etc should be defined.**

Gas

To date, the results of the gas surveys have been patchy. **It is suggested that deeper probes are assessed as the clay-rich soils may be blocking gas reaching the near surface. Further sampling at sites identified by geophysical surveys (see below) is also recommended.**

Others

Geophysics

The preliminary shallow resistivity survey was not carried out in enough detail to be of any great help. A more detailed resistivity survey was carried out in CNAP (details in Alexander and Milodowski, 2013) and this proved very useful for identifying likely zones of shallow groundwater. In CNAP, this was allied to a Ground Penetrating Radar (GPR) survey which indicated the highly fractured nature of the site, allowing appropriate fracture zones (and so potential conduits of rising groundwater) to be identified for sampling. **It is proposed that a detailed resistivity survey be carried out at the site, but that the priority target area is the west side of the river (including around borehole DH02A – see comments in section 3.2). If resources allow, a more detailed survey of the Field Centre could be included. A GRP survey of both areas would also be beneficial.**

Dating the system

It is not enough to show the absence or presence of bentonite reaction with alkaline groundwaters, some idea of the likely fluxes of reactants is crucial to estimating reaction rates etc. In CNAP, an attempt was made to date the initiation of the alkali groundwater flow by two means: a broad based geomorphological study and natural decay series analysis of both groundwaters and bentonites. For the geomorphological survey, both a regional scale (i.e. Zambales wide) and a local scale (Aksitero River catchment area) survey would be necessary. For the natural decay series, appropriate samples of bentonite and groundwater must be analysed. **To provide quantitative data on bentonite reaction, it is proposed that an analysis of**

the reactant fluxes be attempted at Bigbica. It is recommended that a two-pronged examination of the likely initiation date of the alkali groundwater system be carried out as this proved to be very useful in CNAP.

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6 Appendices

Appendix 1: Repeat groundwater analyses for boreholes BB-09 and BB-10

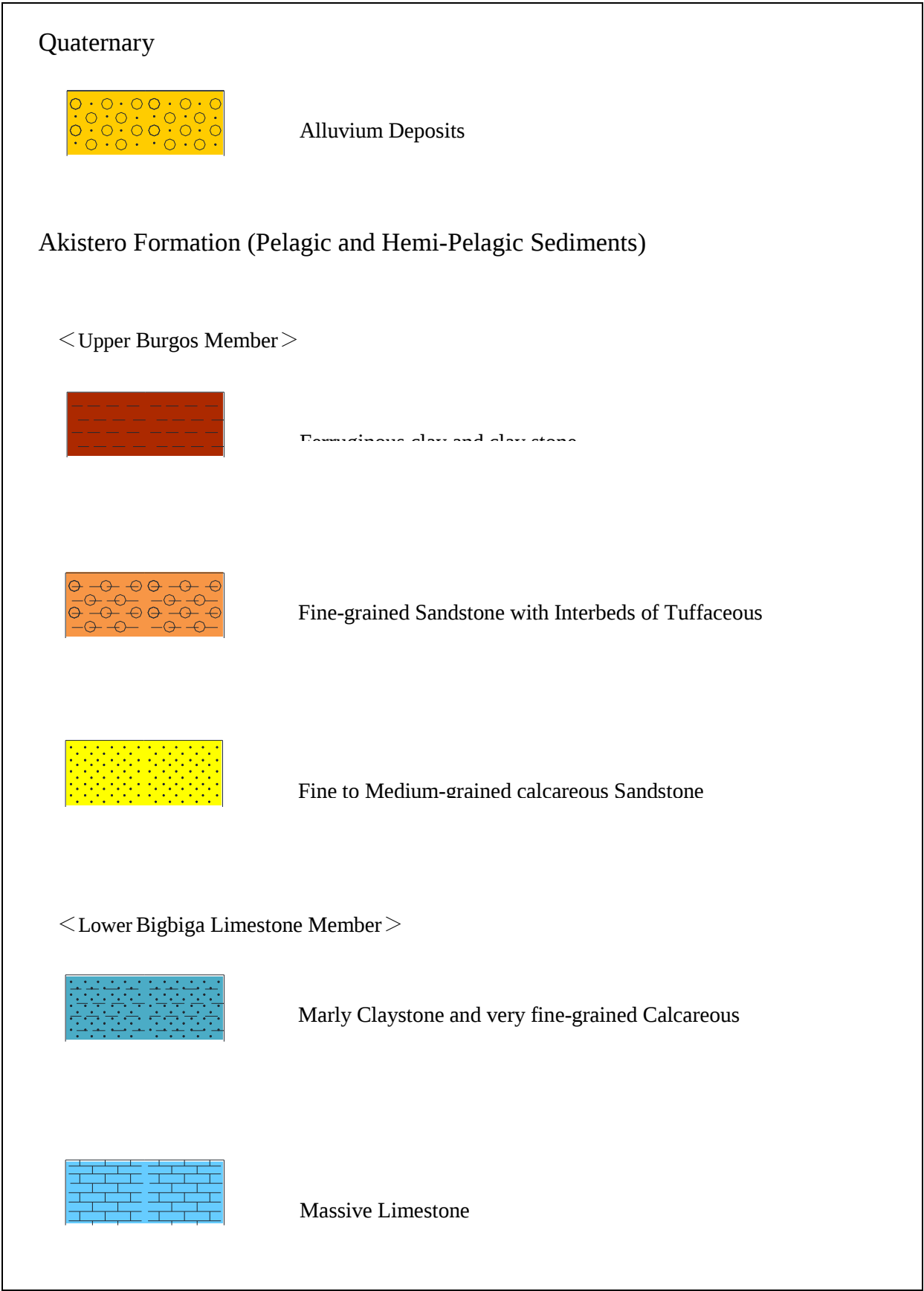
Appendix 2: Borehole descriptions for DH-04, -05 and -06

Appendix 1: Repeat groundwater analyses for boreholes BB-09 and BB-10

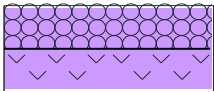
Sample No.	pH	Temp. (℃)	ORP (mv)	EC (mS /m)	DO (mg /l)	CO ₂ (mg/l)	カチオン濃度 (ppm)								アニオン濃度 (ppm)								備考	
							Na ⁺	K ⁺	Ca ²⁺	Si ⁺	Al ³⁺	Mn ²⁺	Mg ²⁺	Fe (2 ⁺ +3 ⁺)	HCO ₃ ⁻	NO ₂ ⁻	NO ₃ ⁻	PO ₄ ³⁻	SO ₄ ²⁻	Cl ⁻	Br ⁻	F ⁻		OH ⁻
BB-10-KWP-01	7.68	28.6	63	48.2	8.48	15.4	61.60	2.82	37.88	31.63	0.00	0.15	6.22	0.54	—	—	—	—	—	—	—	—	—	井戸水(地下4m)
BB-10-KWP-02	8.44	32.2	175	14.8	21.18	40.6	19.28	8.28	11.40	12.91	0.00	0.28	5.09	0.33	—	—	—	—	—	—	—	—	—	溜池(ボーリング(DDH-3)の背後)
BB-10-KWP-03	8.76	33.4	170	20.2	25.67	53.1	12.58	5.30	15.61	13.18	0.00	0.00	4.56	0.26	—	—	—	—	—	—	—	—	—	溜池
BB-10-KWP-04	7.03	35	170	9.9	11.86	23.5	6.27	5.13	5.03	12.01	2.98	0.01	2.63	2.31	—	—	—	—	—	—	—	—	—	溜池
BB-10-KWP-05	7.49	31.5	216	24.4	8.52	115.8	16.72	3.99	19.53	9.02	0.00	0.00	7.75	0.41	—	—	—	—	—	—	—	—	—	溜池(DDH-2近)
BB-10-KWP-06	9.37	29.5	22	45.1	2.3	168.4	117.20	1.97	1.93	29.75	0.00	0.00	0.01	0.27	—	—	—	—	—	—	—	—	—	井戸水(Fishpondのアルカリ地下水)
BB-10-HWP-01	6.88	30.64	224	21.40	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	4.73E-06	溜池(DH04近傍)
BB-10-HWP-02A	8.44	32.40	265	12.48	16.10	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	—	4.34E-06	溜池(DH03近傍、Aはbasalt, Bはbentonite)
BB-10-HWP-02B	8.40	32.50	268	12.50	14.18	—	10.2	16.6	50.7	13.90	0.045	0.323	9.95	0.042	68.59	0.635	1.129	0	39.788	1.791	0	0.014	1.06E-05	
BB-10-HWP-03	8.76	33.40	225	15.69	18.62	—	4.21	8.12	15	7.35	0.039	0.568	5.52	0.041	—	0.407	0.646	0	1.189	1.163	0	0.006	2.20E-07	溜池(DH03Cの背後)
BB-10-HWP-04	7.03	35.00	74-61	7.55	4.92	—	13.8	6.13	75.5	7.87	0.006	0.065	19.1	0.048	—	0.481	0.956	0	1.323	1.725	0	0.008	4.68E-07	溜池(DDH-2近)
BB-10-HWP-05	7.49	30.60	338	23.00	5.57	—	60.2	7.2	78.3	34.73	0.013	0.113	12.2	0.211	—	0.491	1.203	0	2.846	0.772	0	0.009	4.79E-08	
BB-10-HWP-06	9.37	30.10	252	42.60	1.06	—	107	0.215	0.599	34.66	0.012	0.061	0.092	0.035	153.89	0.979	1.199	0	1.332	1.543	0	0.106	3.42E-05	井戸水(Fishpondのアルカリ地下水)
BB-09-KWP-01	9.24	29	28	44	5.76	—	15.797	0.512	0	—	—	0	0	0	—	0	0	0	51.210	4.283	0	0.572	—	井戸水(Fishpondのアルカリ地下水)(09年)
BB-09-KWP-02	8.38	28.8	10	42.6	5.02	—	14.555	0.644	1.212	—	—	0	0	0	—	0	0	0	41.322	3.980	0	0.394	—	河道の湧水(09年)
BB-09-KWP-03	9.3	31	30	34.7	5.02	—	10.922	0.878	0	—	—	0	0	0	—	0	0	0	0	8.223	0	2.968	—	井戸水(Fishpondの旧揚水井)(09年)
BB-09-KWP-04	6.67	29.4	-76	113	3.75	—	28.003	0.855	12.717	—	—	0	1.331	0	—	4.944	0.045	0	138.088	72.600	0	0	—	河道の割れ目湧水(09年)
BB-09-HWP-01	9.09	29.40	-242.0	42.80	—	—	91.20	2.140	0.052	36.10	—	—	1.240	—	—	—	—	—	50.221	4.041	—	—	—	井戸水(Fishpondのアルカリ地下水)(09年)
BB-09-HWP-02	8.45	30.90	-202.0	41.10	—	—	83.60	4.320	5.820	33.60	—	—	0.093	—	—	—	—	—	44.486	4.079	—	—	—	河道の湧水(09年)
BB-09-HWP-03	9.60	32.50	-9.0	35.00	—	—	65.60	3.540	0.797	1.240	—	—	5.290	—	—	—	—	—	0.211	7.750	—	—	—	井戸水(Fishpondの旧揚水井)(09年)
BB-09-HWP-04	7.63	30.80	-41.0	106.6	—	—	143.0	2.540	87.40	23.20	—	—	5.170	—	—	—	—	—	257.512	94.910	—	—	—	河道の割れ目湧水(09年)

Appendix 2: Borehole descriptions for DH-04, -05 and -06

Legend



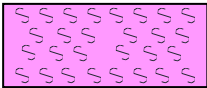
Zambales Ultramafics (Ophiolite Basement)



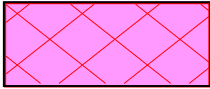
Massive and Auto brecciated Spilitic Basalts with



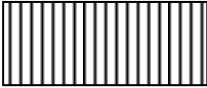
Brecciation (Crush Breccia /Fault Breccial



SPILITIC



SPILITIC BRECCIA



SPILITIC BRECCIA

Drill Hole Number : BB12-DH05
 Location : N15°30'52.8" E120°22'51.2"
 Elevation : 152.53m
 Core Size : NQ
 Total Footage : 10.69m
 Drilling Operation : 29 May,2012 (9:30 a.m.)~30 May,2012 (18:30 p.m.)

Depth(m)	Columnar Section	CR*	RB**	Sample Number*** (Sample Location)	Lithology	Remarks
0.79				(HU001) BB12-DH05-RW001	Quaternary Alluvium Deposits (Unconsolidated Sediments : cobble sized conglomerates)	
1.08					Aksitero Formation (Pelagic and Hemi-Pelagic Sediments)	
1.50				BB12-DH05-RW002 (HU002)	▷ Crushed Ferruginous coarse grained sandstone due to Paleo weathering	Bentonic Layer < Bitumen Inclusion >
1.63				BB12-DH05-RW003 (HU003)	▷ Light grey, coarse grained sandstone with Interbeds of Tuffaceous Layer including elongated Pumice Fragments	
2.10				BB12-DH05-RW004 (HU004)	▷ Light yellow to grey Calcareous Tuffaceous sandstone	< Bitumen Inclusion >
2.37					▷ Light blue to gray, very fine grained Tuffaceous Calcareous sandstone with elongated Pumice Fragments	
2.93				(HU005) BB12-DH05-RW005	▷ Light grey, fine grained Calcareous Tuffaceous sandstone with elongated Pumice Fragments	Bentonic Sediments < Bitumen Inclusion >
3.31				BB12-DH05-RW007 (HU007) BB12-DH05-RW006 (HU006)	▷ Light grey to Light brown, fine grained, Calcareous Tuffaceous sandstone with elongated Pumice Fragments	(Mn – staining)
3.80						Bentonic Layer
3.93				BB12-DH05-RW008 (HU008)	▷ Fine to medium grained Calcareous Sandstone with Interbed of Light buff Tuffaceous layer including elongated Pumice Fragments	
4.20				BB12-DH05-RW009 (HU009)	▷ Light grey, fine grained, Calcareous Tuffaceous sandstone with elongated Pumice Fragments	
4.30						Bentonic Sediments < Bitumen Inclusion >
4.51						
4.90				(HU010) BB12-DH05-RW010	▷ Fine grained, Calcareous Sandstone with elongated Pumice Fragments	
5.52				BB12-DH05-RW011 (HU011)	▷ Light grey to White grey, fine grained, Calcareous Tuffaceous sandstone with elongated Pumice Fragments	Bentonic Sediments < Bitumen Inclusion >
5.62				BB12-DH05-RW012 (HU012)		
6.00						
6.48				BB12-DH05-RW013 (HU013)	▷ Light Grey to White grey, very fine grained, Tuffaceous Sandstone with elongated Pumice Fragments	Bentonic Sediments < Bitumen Inclusion >
6.61						
7.29				BB12-DH05-RW014 (HU014)	▷ Light grey, fine to medium grained Tuffaceous sandstone with elongated Pumice Fragments	Bentonic Sediments
7.61						
8.06				(HU015) BB12-DH05-RW015	▷ Light grey to White grey, fine to medium grained Tuffaceous Sandstone with elongated Pumice Fragments	Bentonic Sediments
8.51				BB12-DH05-RW016 (HU016)		Bentonic Sediments
9.00						
9.16				(HU017) BB12-DH05-RW017	▷ Light grey, fine grained, Calcareous Tuffaceous sandstone with elongated Pumice Fragments	Bentonic Sediments [Bleached zone]
9.68				BB12-DH05-RW018 (HU018)	▷ Light Grey to Light brown, fine grained Calcareous sandstone with elongated Pumice Fragments	
9.97				BB12-DH05-RW019 (HU019)	▷ Light green, medium grained, Tuffaceous sandstone with elongated Pumice Fragments	
10.69				BB12-DH05-RW020 (HU020)	Zambales Ultramatics (Basement) Massive and Auto brecciated Spilitic Basalts with scattered zones of Pillow Lava ▷ Brecciated Spilitic Basalt with carbonates veinings	

CR* : Core Recovery (Core-lacked Zone)

RB** : Range of Brecciation (Crush-Breccia, Fault-Breccia / Fragmentation, Fracturing, cracking)

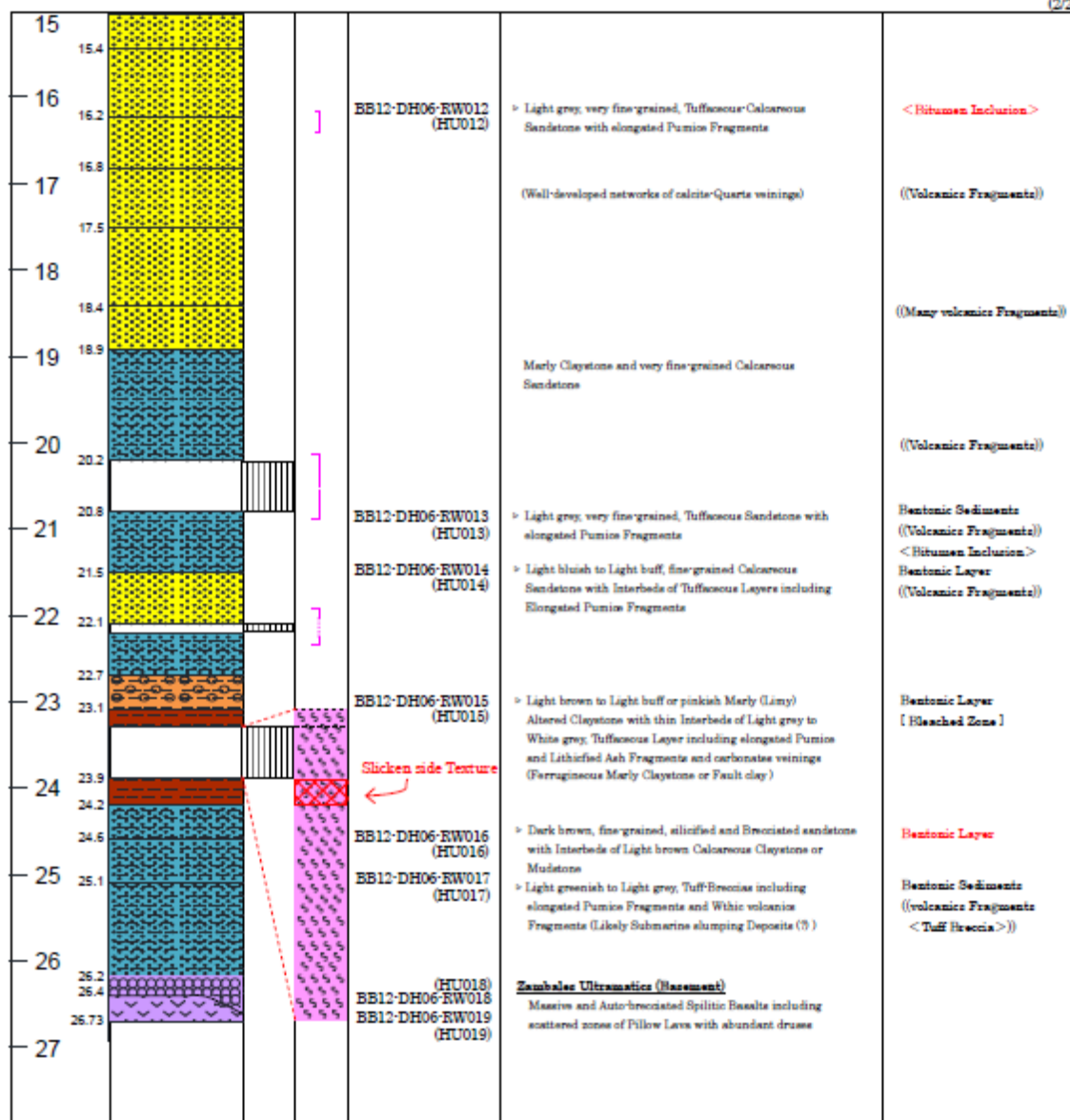
Sample Number*** : RW (RWMC), HU (University of Hokkaido)

Drill Hole Number : BB12-DH06
 Location : N16°30'51.3" / E120°22'53.5"
 Elevation : 155.50m
 Core Size : NQ
 Total Footage : 26.73m
 Drilling Operation : 30 May,2012 (10:10 a.m.)~1 June,2012 (15:30 p.m.)

(1/2)

Depth(m)	Columnar Section	CR*	RB**	Sample Number*** (Sample Location)	Lithology	Remarks
0.2					<u>Quaternary</u> (Alluvium Deposits : Unconsolidated Sediments)	(cobble sized Conglomerates)
0.4					<u>Altiplano Formation</u> (Pelagic and Hemi-Pelagic Sediment)	(silt)
0.7					Ferruginous clay and claystone due to Paleo weathering	Bentonitic Sediments
0.8				BB12-DH06-RW001 (HU001)	White to Light grey, very fine-grained Calcareous Tuffaceous Sandstone	
1.1						
1.4						
1.6						
1.7						
1.8				BB12-DH06-RW002 (HU002)	Light blue to grey fine-grained Sandstone with Interbeds of Tuffaceous Layer including elongated Pumice Fragments	Bentonitic Layer < Bitumen Inclusion >
2.0						
				BB12-DH06-RW003 (HU003)	Light yellowish to grey, fine to medium grained Tuffaceous Calcareous Sandstone with elongated Pumice Fragments	Bentonitic Sediments < Bitumen Inclusion >
2.9						
3.0						
3.3						
3.4						
3.6				BB12-DH06-RW004 (HU004)	(Fine to Medium-grained Tuffaceous and Calcareous Sandstone)	Bentonitic Sediments
3.7					(Interbeds of Tuffaceous Layer including elongated Pumice and Ash Fragments)	Bentonitic Layer
4.0						
4.1						
4.6						
5.1						
5.2				(HU005)	Grey to dark grey, medium-grained Tuffaceous Sandstone with Elongated Pumice Fragments and Carbonate veinings + (Zeolite?)	Bentonitic Sediments (Bleached Zone)
5.7				BB12-DH06-RW005 (HU005)	Light Grey, fine-grained Tuffaceous-Calcareous sandstone with elongated Pumice Fragments	< Bitumen Inclusion >
5.8				BB12-DH06-RW006 (HU006)		
6.6					Light greenish to Light grey, very fine-grained Tuffaceous Calcareous sandstone with elongated Pumice Fragments	Bentonitic Sediments
6.7				BB12-DH06-RW007 (HU007)		
7.6					Light yellowish to grey, fine to medium grained Calcareous Sandstone with Interbeds of Tuffaceous Layers including elongated Pumice Fragments	Bentonitic Layer (Bleached Zone)
7.8				BB12-DH06-RW008 (HU008)		< Bitumen Inclusion > (volcanic Fragments)
8.6						
8.8				BB12-DH06-RW009 (HU009)	Grey to Light bluish, very fine-grained Calcareous Sandstone With Interbeds of Tuffaceous Layers including elongated Pumice Fragments	Bentonitic Layer (Bleached Zone)
9.6						< Bitumen Inclusion > (volcanic Fragments)
9.7					Massive grey claystone	(volcanic Fragments)
10.6						
10.8					Very fine-grained Tuffaceous and Calcareous Sandstone With elongated Pumice Fragments and networks of Calcite-Quartz veinings	Bentonitic Sediments
11.6				BB12-DH06-RW010 (HU010)	Light grey, fine to medium grained, Calcareous-Tuffaceous Sandstone with elongated Pumice Fragments	Bentonitic Sediments (Abundant volcanic Fragments)
12.8						
13.6				BB12-DH06-RW011 (HU011)	Light grey, very fine-grained Calcareous sandstone and Marly claystone	< Bitumen Inclusion >
14.6					White to Light buff fine-grained Massive Marly Claystone (Limy Claystone)	

(2/2)



CR* : Core Recovery (Core-lacked Zone)

RB** : Range of Brecciation (Crush-Breccia, Fault-Breccia / Fragmentation, Fracturing, cracking)

Sample Number*** : RW (RWMC), HU (University of Hokkaido)

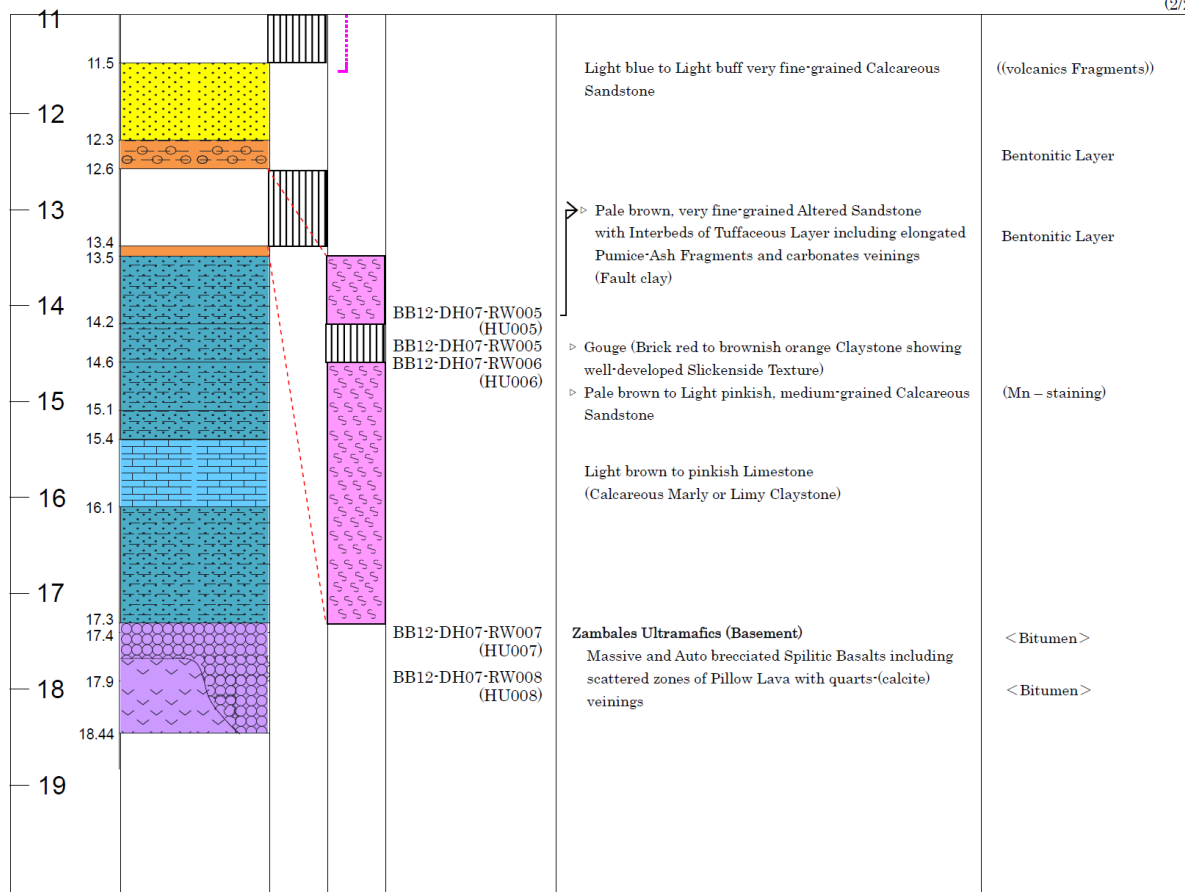
Drill Hole Number : BB12-DH07
 Location : N15°30' 51.3" / E120°22' 53.5"
 Elevation : 152.34m
 Core Size : NQ
 Total Footage : 18.44m
 Drilling Operation : 1 June,2012 (16:00 p.m.)~3 June,2012 (17:00 p.m.)

(1/2)

Depth(m)	Columnar Section	CR*	RB**	Sample Number*** (Sample Location)	Lithology	Remarks
0.7					<u>Quaternary</u> Alluvium Deposits (Unconsolidated Sediments)	Cobble sized Conglomerates and soils
1.7					<u>Aksitero Formation (Pelagic and Hemi-Pelagic Sediments)</u>	
1.9					Ferruginous clay and Claystone due to Paleo weathering	
2.3				BB12-DH07-RW001 (HU001)	Light buff to yellowish, very fine-grained calcareous Sandstone and Siltstone with Interbeds of Light buff Tuffaceous Layer including elongated Pumice Fragments	Bentonic Layer [Bleached Zone]
2.6				BB12-DH07-RW002 (HU002)	White grey to Light grey, fine-grained Calcareous Tuffaceous Sandstone with Interbeds of Tuffaceous Layer including elongated Pumice Fragments	Bentonic Layer < Bitumen inclusion >
2.9						
4.2						
4.3					Bluish to grey fine-grained Calcareous Sandstone	Bentonic Sediments (Mn – staining)
4.5						Bentonic Layer ((Volcanics Fragments))
4.7						
5.2						
5.6				BB12-DH07-RW003 (HU003)	White to Light grey, fine-grained Calcareous Tuffaceous Sandstone with Interbeds of Tuffaceous Layer including elongated Pumice Fragments and quartz-calcite veinings	Bentonic Layer [Bleached Zone] ((Volcanics Fragments))
6.8				BB12-DH07-RW004 (HU004)	Dark grey to Yellowish white, graded fine to medium-grained Calcareous Tuffaceous Sandstone	Bentonic Layer (Mn – staining) ((Volcanics Fragments))
7.7						
8.2						Bentonic Layer
8.5						Bentonic Layer ((Volcanics Fragments))
8.7					Very Fine-grained Sandstone with Interbeds of Tuffaceous Layer including elongated Pumice Fragment and associated with networks of calcite-quartz veinings	
9.5						Bentonic Layer [Bleached Zone] ((Volcanics Fragments))
9.7						
10.3						Bentonic Layer [Bleached Zone] ((Volcanics Fragments))
10.6						Bentonic Layer [Bleached Zone] ((Volcanics Fragments))
11.5						

BB12-DH07

(2/2)



CR* : Core Recovery (Core-lacked Zone)

RB** : Range of Brecciation (Crush-Breccia, Fault-Breccia / Fragmentation, Fracturing, cracking)

Sample Number*** : RW (RWMC), HU (University of Hokkaido)