THEMATIC ISSUE



A multilevel U-tube sampler for subsurface environmental monitoring

Xuehao Liu^{1,2} · Qi Li¹^(D) · Ranran Song¹ · Zhiming Fang¹ · Xiaochun Li¹

Received: 23 March 2016/Accepted: 9 August 2016 © Springer-Verlag Berlin Heidelberg 2016

Abstract Sampling fluid in boreholes is always challenging from the standpoints of maintaining integrity and minimizing contamination. This paper focused on the U-tube sampling technology. Firstly, according to initial development motivation, briefly comparison was conducted with available fluid sampling technologies based on driving forces and sample quality. Then, the development history of the U-tube sampling technology was introduced, as well as a comprehensive investigation of its site applications around the world. In particular, a novel multilevel U-tube sampler was designed in China specifically for shallow subsurface, and it had been proven both technically reliable and economically feasible in several carbon capture, utilization, and storage (CCS/CCUS) projects, including the Shengli CO2-enhanced oil recovery (CO2-EOR) project, the Shenhua CCS demonstration project, and the Jilin CO₂-EOR project. Finally, the benefits, drawbacks, and future developments of U-tube sampling technology were pointed out as a conclusion. The U-tube sampler used in shallow subsurface is quite suitable in dedicated monitoring projects which specifically require

This article is part of a Topical Collection in Environmental Earth Sciences on "Subsurface Energy Storage", guest edited by Sebastian Bauer, Andreas Dahmke, and Olaf Kolditz.

☑ Qi Li qli@whrsm.ac.cn

¹ State Key Laboratory of Geomechanics and Geotechnical Engineering, Institute of Rock and Soil Mechanics, Chinese Academy of Sciences, Xiaohongshan 2, Wuchang, Wuhan 430071, China

² Wuhan Center of Geological Survey, 69 Guanggu Road, Wuhan 430502, China three-dimensional tracing and accurate fluid analysis in a long term.

Keywords U-tube · Multilevel fluid sampling · Subsurface storage · Environmental monitoring · CCUS

Introduction

Fluid sampling is common and important in many geological activities. In order to examine the physical, chemical, and biological conditions of the subsurface geological environment, it is crucial to get underground fluid samples of maintaining integrity, minimizing contamination, and continual steady recovery amount for the coming hydrobiogeochemical analysis. The results of the analyses are used to predict possible changes in those conditions with time and to determine the subsurface conditions. Some examples are predicting changes in the chemical quality of water with time and distance in the event of environmental contamination and remediation, investigating the origin of pollution solutes in waste disposal areas or other types of contaminated sites, monitoring the exact arrival of a CO₂ $(H_2S \text{ or } CH_4)$ plume, or groundwater tracers. The analyses can also be used to determine the exact shape of a breakthrough curve of a CO₂ (H₂S or CH₄) plume in a deep reservoir, its possible leakage into the shallow subsurface environment in the vicinity of the underground storage of waste or CO₂, and the recovery of underground resources such as CO₂-enhanced oil recovery (CO₂-EOR) (Hovorka et al. 2013; Jenkins et al. 2015; Yang et al. 2013).

However, collecting unhindered samples of the target formation in time does pose challenges for many reasons, from formation contamination by drilling and backfill to maintaining accuracy and integrity during sampling from several targeted layers. There are also many other substantial engineering activities associated with the retrieval of fluid sampling in a practical borehole. In one review (Parker 1994), it was stated that there can be significant problems with degassing and loss of oxidizable and volatile inorganics under certain circumstances with almost all samplers. In addition, it has been shown that the effect of sampling devices on the quality of groundwater was greater than any material effects by polyvinyl chloride (PVC) or steel (Houghton and Berger 1984). According to Barcelona et al. (1984, 1985), the sampling mechanisms for collecting groundwater samples are among the most error-prone elements of monitoring programs.

Existing groundwater sampling equipment could not always suit to the increased engineering demands, although sampling fluid from boreholes is a generally accepted practice for hundreds of years, including wireline samplers and pump samplers. These unsuitable situations include negligible changes in temperatures (T) and pressures (P) during a sampling process from a downhole, and the chemical instability of multi-phase and multi-component fluids. In addition, three-dimensional tracing or monitoring demands that high accuracy samples be obtained from multilayers through one borehole. These situations create the need for major improvement of traditional downhole sampling technologies as well as the development of new methods.

U-tube sampling technology was selected and its research and development (R&D) is the focus of this paper, after comparing several fluid sampling technologies for use in boreholes above. First, a brief history is presented of the related technology developments and engineering applications around the world. Next, we present an in-depth assessment of the development of U-tube technology in China. A novel U-tube sampler is highlighted. The sampler is specially designed with all plastic materials and has multiple layers for shallow subsurface conditions.

Overall, the paper summarizes the rapid development of U-tube sampling technology, in order to identify the required data to make informed decisions concerning applications, which is suitable for permanent subsurface sites that require accuracy for both deep and shallow wells.

The U-tube technology and its application around the world

Our primary motivation of R&D the U-tube sampler in shallow subsurface is to verify CO_2 leakage from deep reservoir into shallow aquifers, and how it could affect the subsurface environment around human beings once leakage really happen. Thereafter, the monitoring results of U-tube from CO_2 geological utilization and storage areas might at least have three main merits to different relevant interested

parties. For project builders, it could be used to provide early warnings of whether CO_2 leakage and possible locations it happens, thus effective engineering solutions could be made at the first time, like reducing the CO_2 injected pressure, plugging the leaking faults or boreholes. For public, especially residents near the injection areas, it could provide continual and solid evidence to enforce the public confidence and improve the public acceptance (Chen et al. 2015; Lei et al. 2014). For governments and regulators, it should be trying to illustrate what the environmental risks might be at long term and whether these risks are in control.

Undoubtedly, to achieve the goals above, several monitoring techniques should be combined together, which at least include 3D seismic, soil gas, atmospheric, and shallow groundwater (Boreham et al. 2011). As for shallow groundwater monitoring, several borehole fluid sampling technologies could be selected, such as wireline fluid sampling, pump sampling, and gas-operated sampling.

As far as we concerned above, it was divided into three sections in this chapter. Firstly, why U-tube sampling technology is unique in some occasions and thus could not be replaced by other existing sampling technologies. Thereafter, an overall description of U-tube sampling technology is presented, including the origin and vital technological developments. Finally, a comprehensive list of its site applications around the world is addressed in detail.

Comparison of different borehole fluid sampling technologies

Borehole fluid sampling technology typically can be divided into three basic categories based on their different driving forces, i.e., wireline tools, pumps, and gas-operated samplers, which are shown in Fig. 1.

Wireline fluid sampling devices, which also are called grab samplers (Parker 1994), are characterized by wireline tools from the labor force to a tractor as driving force, by valve-tripping mechanisms, and by a volume-limited sampler which is connected to wireline tools down the wellbore to the target depth, including positive displacement, vacuum, flow-through samplers, and bailers. This technique is cost-effective, and packers could recycle during sampling. However, it is not suitable for use in long term and frequently monitoring projects due to cross-contamination and time-consuming in preparation, retrieving, and flushing of the equipment when it used to sampling from multilayers through one borehole immediately.

Unlike wireline fluid sampling that use wireline tools, there are a group of fluid sampling methods that depend on pumps to transfer the borehole fluid to the wellhead. These include the line shaft turbine pump, electric submersible pump, peristaltic pump, inertial pump, gas-driven bladder

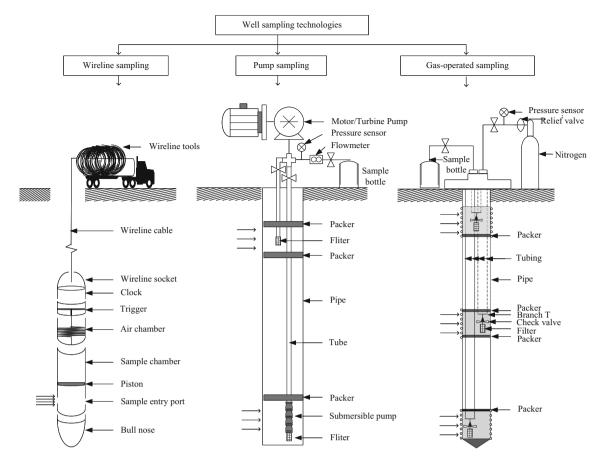


Fig. 1 Schematic diagrams of three typical kinds of well sampling technologies

pump, and vacuum pump. Research studies have identified several problems associated with pump sampling including (1) higher pumping rates result in samplers with higher turbidities and larger particles (Puls and Powell 1992); (2) there is a potential for contamination of the sample from direct contact with the body of the pump its impeller, or gasoline; and (3) they might cause volatilization, oxidation, precipitation, adsorption, and ion-exchange reactions during pumping (Parker 1994). In other words, the precision of this method probably cannot satisfy demands of monitoring subtle changes due to potential CO_2 leakage.

Gas-operated sampling method blows out reservoir fluids from a tube using an inert carrier gas, such as compressed nitrogen. As a new choice, the U-tube sampling technology is quite suitable in dedicated monitoring projects which specifically require three-dimensional tracing and accurate fluid analysis. It can obtain fluid samples from multiple underground layers smooth and steady, which reduce the impact on the flow field because the fluid flow is minimal, slow, and under constant pressure. It could conduct sampling quite frequent and coexist with other downhole measurement equipment and operations.

It seems that the U-tube possibly satisfies our high technical requirements. However, the reality is that there is none trial or application around the world using the U-tube in shallow subsurface, and no potential company or research group could provide. As far as we known, it is partly because this sampling technology is quite new, which would be shown clearly from its history below. More importantly, the U-tube samplers were thought very fragile for breakdown and easy to block. That means several technical difficulties still need to overcome.

The history of the U-tube sampling technology

It would be beneficial to understand the U-tube which specifications from its technology improvement history. Unlike line tools lifting or pumping, the driven force of the U-tube is nitrogen displacement. This means that its tube lines should be cycled from wellhead to each sampling sites underground. Therefore, a check valve could be crucial in this case. However, it is also the check valve and cycled tube lines that result in a fragile U-tube system.

The porous cup sampler was thought to be one of the origins of the U-tube sampler, which was described by Briggs and McCall (1904) as "artificial roots," and at that time, it was the only practicable means of obtaining an in situ sample of water from the soil. Thereafter, an

improved type of soil–water sampler was introduced by Wagner (1962), and it was called a "suction lysimeter" by Parizek and Lane (1970). It consisted mainly of a porous ceramic cup, a connecting lip around the top of the cup to provide vacuum conditions with airtight and watertight seals. When ceramic cup contacts with moist soil, the pores in the wall of this cup would be filled with fluids because of capillary suction. The sizes of the pores are so tiny that the fluid sample is held inside them with a force that is sufficient to seal the cup against air pressure (Wagner 1962).

During sampling, air is evacuated from the air chamber by a vacuum on the surface, and the suction that is applied to the water in the porous ceramic cup is overcome by applying a counter force to the air–water interface. Water will move into the sample bottle through the air chamber if the applied vacuum is stronger than the suction in the soil around the ceramic walls.

Unfortunately, the porous ceramic samplers cannot sample fluids held in a formation that is more than 10 m deep, because the driving force is provided by the suction from an air vacuum, which is limited to 1 atm or about 100 kPa. The tubes or connections without cement may result in an inadequate airtight seal, resulting in either a rapid loss of vacuum or the inability to create a vacuum initially. Incomplete backfilling and tamping also could result in voids around the porous cup (Freifeld et al. 2005).

These obstacles were not resolved until (1973) when (Wood) illustrated an advanced design by placing a new element, i.e., a poppet check valve, into the sample collection assembly, without using vacuum and pressure application to pressure the cup (England 1974). This is shown in Fig. 2 as stage 1.5 of the U-tube technology. In addition to the introduction of the check valve, another vital improvement was the U-shaped tubes, which replaced the traditional two separated tubes. Technically speaking, based on a check valve, formation fluid could seep into the buried porous cup and storage rather than being drawn in by the suction of a vacuum pump. The U-shaped tubes prevented pressurization of the porous cup much better than had been the case before. These two novel designs make the driving force alternate between the suction of the air vacuum and the displacement of the compressed nitrogen; alternating sequence with the check valve allows for sequential differential pressure, inflow from the formation, then pushing up of the fluid out of the extraction line. This great and vital improvement made this sampler possible by allowing fluid sampling to occur at pressures greater than the suction lift off (~ 10 m) and theoretically at any depth.

Nevertheless, it still was called a porous cup sampler at that stage, partly because of the vacuum that was involved on the surface collection equipment for removing the sample and partly due to the porous ceramic cup that still was used for the downhole collection of fluid from the formation. In addition, at least two doubts remained about the representativeness of the fluid samples obtained using this technology. One was that the concentrations of the solutes collected from the macro-pores probably were different from those collected from the micropores, and the other doubt concerned the problem of adsorption of ions by the ceramic cell of the porous cup (England 1974).

Thirty years later, Freifeld et al. (2005) developed a U-tube sampler for acquiring borehole fluid and conducted several projects that successfully demonstrated the efficacy of this technology at depths greater than 2000 m (Freifeld 2010). These field applications proved Wood's claim that the technology theoretically would allow sampling fluids at any depth. The field applications also made substantial contributions to validating the theoretical and practical capabilities of U-tube sampling technology, which meant that the technology was ready for commercialization.

Applications of the U-tube sampler around the world

Figure 2 shows that U-tube sampling technology first appeared on the scene in 2004. As a fairly new addition to the market, it has already become a mainstay among the well sampling tools used to acquire fluid samples related to carbon capture and storage (CCS) sites all over the globe (Wolff-Boenisch and Evans 2014). The technology is used to monitor "the exact arrival of a CO_2 plume and to determine the exact shape of a break-through curve of groundwater tracers" (Wolff-Boenisch and Evans 2014).

To date, according to the published data that are available, U-tube sampling technology is being used in 13 typical field applications around the world. From the first site in 2006—the Frio Brine Pilot Project (Freifeld 2010; Freifeld et al. 2005)-U-tube sampling technology has undergone rapid and comprehensive development in engineering design and commercialization over the recent 10 years. Compared to the old versions of samplers that were limited to a depth of 10 m in the 1970s, today, U-tube samplers can be deployed at depth exceeding 3000 m, such as the Cranfield CCS project at a depth of 3200 m (Parizek and Lane 1970). Meanwhile, the design improvements and increasing applications of this technology that are occurring in China are quite impressive. To date, nearly 50 % of the field applications of this technology all around the world are in China, even though none of these sites had been deployed the samplers at depths exceeding 500 m yet. However, there have been indications that more ambitious plans will be undertaken in the near future.

Figure 3 shows the distributions as well as selected information, and more details about these applications are given below. Based on the technical provider and the scale

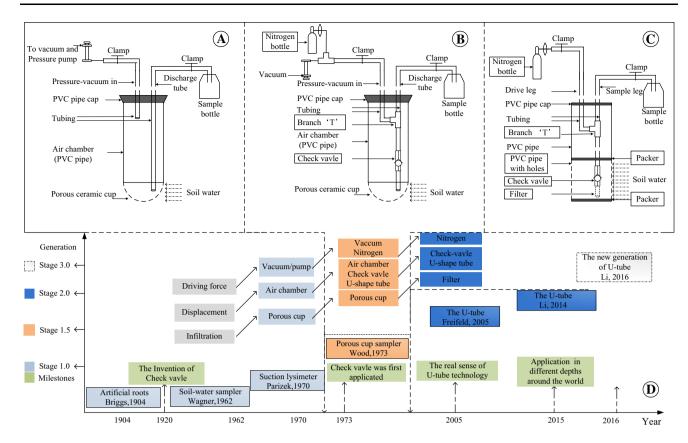


Fig. 2 History of the development of U-tube sampling technology

of projects, these sites are divided into three groups, i.e., cases 1–7, cases 8–9, and cases 10–13. The technologies in cases 1–7 were provided either directly or indirectly by the Lawrence Berkeley National Laboratory (LBNL) in the USA. Cases 8–9 were pilot tests that were conducted in China. Regarding cases 10–13, these U-tubes were designed especially for use with shallow subsurface and they were developed by the Institute of Rock and Soil Mechanics, Chinese Academy of Sciences (Li et al. 2014).

It is worth to note that the seven cases 1–7 were introduced in detail by Freifeld (2010). In order to display a comprehensive view of the field applications around the world, only vital information and useful technical problems encountered in engineering were selected here.

The first deep borehole application of U-tube samplers was conducted in the Frio Brine Pilot Project (Dayton, Texas) from 2004 (Xu et al. 2010). The U-tube located at a depth of 1.5 km with a typically volume of 118 L. Problems aroused in this site, including hydrate formation and freezing, unexpected large volumes of gases and high moisture and salt content (Freifeld 2010).

The second application was the CO_2CRC Otway Project (Melbourne, Australia) (Paterson et al. 2013; Stalker et al. 2015). A bottom hole assembly containing three U-tubes was lowered 2 km. One significant hurdle was the

unexpected presence of natural waxy alkanes. All three U-tubes have been functioning since their initial deployment in October 2007 (Freifeld 2010).

The third located "at the High Lake massive sulfide deposit (Nunuvut Territory, Canada), with a depth of 350 m by hand. The sampling tubes were eventually blocked due to the presence of heavily-mineralized zones and temperatures of about -6 °C. Seven samples were collected prior to the blockage of the tubes" (Freifeld 2010).

The fourth application located in Amargosa Valley (Yucca Mountain, Nevada) (Tsang et al. 1999), and four U-tubes were fitted at two separate locations lower than a depth of 400 m (Freifeld 2010). However, the fluid that was produced initially was significantly contaminated with what was suspected to be bentonite, which might have come from the backfill materials. However, after repeated operation, the turbidity of the fluid finally decreased somewhat (Rutqvist et al. 2008).

In the Greenland Analogue Project near Kangerlussuaq (Oy 2011; Stackhouse et al. 2010), U-tube samplers were deployed below the permafrost, which was used jacket and heater lines to prevent freezing successfully while sampling (Freifeld 2010).

U-tube samplers were deployed at a depth of 3.2 km (subsea) in the Cranfield field (Li and Li 2015), and CO_2

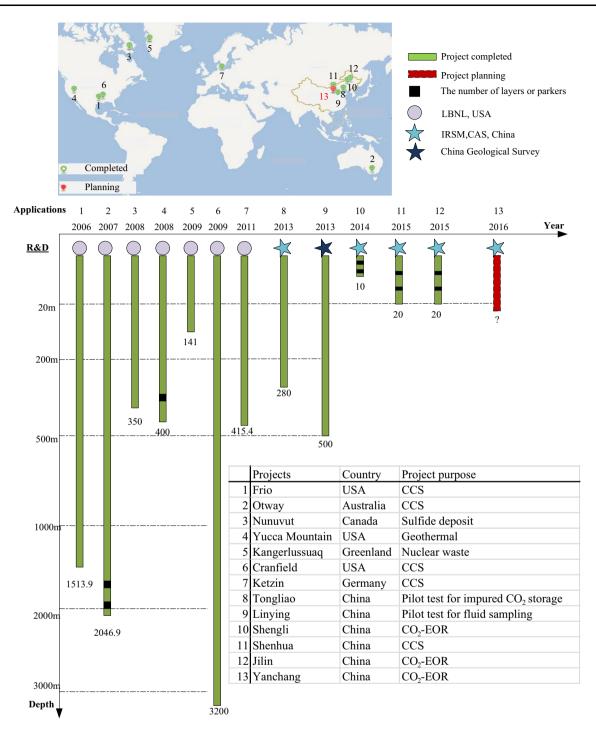


Fig. 3 Projects or pilot tests of the U-tube sampling technology around the world

storage demonstration sites were located 20 km east of Natchez in the southwest of Mississippi, USA (Hosseini et al. 2013; Lu et al. 2012). By the end of August 2011, 3 billion kilograms of gas (~95 % CO₂ mixed with CH₄) had been injected and stored (Choi et al. 2013). Unfortunately, the U-tube samplers temporarily are blocked by solids from December 5 to December 12, 2009, so a downhole Kuster sampler was deployed urgently (Lu et al. 2012).

At the Ketzin pilot site to detect hydraulic and geochemical impact on groundwater, the U-tube sampling system was located at 415.4 m in 2011, which requires less equipment and reduced effort but initially higher installation costs (Martens et al. 2014). Regarding the precision of sampling, the mean of 0.6 and 1.5 % residual drilling mud is represented in proportion of the U-tube samples until November 2012, in comparison with 7.2 and 8.8 % from the pump test (Barth et al. 2015).

The cases 8–9 about pilot tests of the U-tube in China were shown below. And the cases 10–13 using the novel U-tube in shallow subsurface will be illustrated in next chapter.

The Tongliao pilot-scale experimental site was located northeast of Tongliao City, Inner Mongolia, China (Wei et al. 2015; Zhu et al. 2015). The field site for the experimental injection of CO₂ into a shallow aquifer was chosen in the Qianjiadian depression in the southwestern Songliao Basin; 200,000 kilograms of CO₂ was injected into the aquifer at depths of 180-250 m, and a monitoring scheme was used to assess the migration behavior and geochemical impact of the co-injection of CO_2 and air (Zhu et al. 2015). The project was continued for one year with a CO₂ storage tank, liquid CO₂ pump, air pump, heating apparatus, gas mixing apparatus, well fields, and monitoring apparatus. The U-tube samplers used a tube with a diameter of 6.25 mm and a 20-m stinger with a check valve through a pneumatic packer. The pulse delay method was used during the sampling operation in order to simultaneously obtain a thimble-sized amount of gas from the geological formation and measure the downhole pressure by self-referral. Approximately 3.2 L of gas at the conditions of the reservoir condition was sampled each time. However, this U-tube was used in a borehole without a multilevel sampling system and proper zonal isolation due to the limitations imposed by the engineering complexity and lack of experience, which led to relative uncertainty because the fluids in various geological layers were sampled together, and they were diluted by the large volume of water in the casing.

The Linying field test, located in Linying, Henan Province, China, was conducted by China Geological Survey in 2014 to demonstrate its U-tube sampling technology (Li et al. 2015). An existing geothermal well was selected that had a depth of 1206 m and a diameter of 159 mm at depths below 168 m. Sampling operations were conducted three times during one week at depths of 270, 370, and 500 m, respectively. The sampling volume was 23.3 L, and the maximum flow rates were 55.9 L/h, with a driving pressure of 4.9 MPa for 12 min. It is worth noting that the deepest location of the check valve was 400 m, and there was only a one layer U-tube in the borehole without a packer. It has been reported that this U-tube currently is blocked and cannot be operated at this time.

R&D of multilevel U-tube sampler for shallow subsurface in China

Novelty and specialization

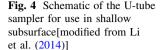
In consideration of the disadvantages of traditional borehole fluid sampling technologies and the challenges or limited behaviors mentioned above about the U-tube technology, a novel U-tube sampler was designed and developed specifically for shallow subsurface, i.e., less than 200 m. The design was based on Freifeld's work (Freifeld et al. 2005) and the experience that was gained from the Tongliao pilot-scale experiment (Wei et al. 2015) and the Shengli CO₂-EOR project (Li et al. 2016), and this novel U-tube sampler has proven to be technologically and economically feasible so far in the Shenhua CCS project and the Jilin CO₂-EOR project.

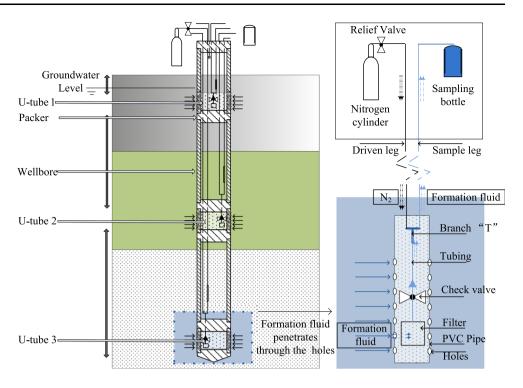
After getting a quick overview of the U-tube above, the composition and operation procedure of the novel U-tube sampler are revealed in detail here. Figure 2c shows a schematic of the modified U-tube sampler, which mainly consist of filter, check valve, U-shaped tube (including tubing, branch "T," driven leg, and sample leg), fluid chamber (including perforated pipe and PVC pipe cap), and packer. Firstly, residual fluid in fluid chamber should be vented through the U-shaped tube before sampling, which the tube using the polyurethane with an outside diameter of 8 mm and inside diameter of 5 mm. Therefore, original formation fluid would permeate into the fluid chamber due to the formation's hydrostatic pressure (Freifeld 2010) after venting. Then, to collect a sample, force the drive leg through a nitrogen cylinder to recover original formation fluid using compressed gas, and thus, the sample bottle located at wellhead could obtain the fresh fluid sample at the sample leg of the U-shaped tube. Finally, repeat these collection cycles to get samples from different layers underground (Fig. 4).

There are at least two typical improvements in this novel U-tube with shallow subsurface. Firstly, the quality of sample is improved heavily through finely design; besides, the connection of original formation fluid with atmosphere was cut off by a check valve. Inspired by the framework of the porous cup sampler, the fluid chamber was designed specially. Its capacity was three times of the rated sampling volume at wellhead, to alleviate the impact of residues. Moreover, the flow rate of formation fluid seepage into this fluid chamber was controlled by the diameter and total area of the tiny holes, in order to balance a slow flow field as possible underground and an acceptable sampling interval at wellhead.

For another, blockage and fragility are overcome to a certain degree, which were thought the fatal flaw of a U-tube sampler and had caused several failures during site applications before. It could be partially due to check valve cannot work correctly or enduringly in condition of a high amount of sediment or turbidity (Parker 1994), which is the core component in a U-tube sampler and cannot be repaired or replaced once deployed on site. There are three effective measures we can think of. In the first place, it can never too careful to choose a filter. On

Environ Earth Sci (2016)75:1194





account of the numerous tiny particles and severe contamination from the drilling fluid to damage check valve, a porous filter combined with a filter screen of 500 meshes was used to tackle this challenge. Filters from markets at this stage are still not good enough to prevent microparticles destroying check valve, but a porous filter with a specific permeability we selected here could stop or slow down the flow rate of these dangerous particles until sedimentation of drilling fluid. Secondly, trying to slow down the flow rate of original formation fluid through the fluid chamber before filtering is demonstrated an effective procedure to alleviate blockage and improve the durability of the U-tube sampler. Lastly, it is worth implementing a mature testing procedure on the whole system before deploying into borehole, in order to eliminate potential technical problems. Other improvements or differences used in this version of the U-tube samplers include all materials are plastic, including tubes, check valves, and filter mesh, all of which are low cost and quite easy to deploy onsite.

While it is worth noting that this U-tube (for shallow subsurface less than 200 m) is not sophisticated as the original U-tube developed by Freifeld (2005). Due to the tiny change of P/T condition during sampling process, problems like phase separation of sample, hydrate formation, solids precipitation, or large amount of unexpected gases once appeared in deep would not occur for shallow subsurface. However, the novel U-tube for shallow subsurface has been demonstrated technical and economic feasibility in China first. So far, more than thirty sets of the U-tube deployed have functioned well without any technical problem over 2 years, at the depth of 10–30 m, after overcoming several obstacles encountered at laboratory and failures happened during demonstrated testing of the Shengli CO₂-EOR project.

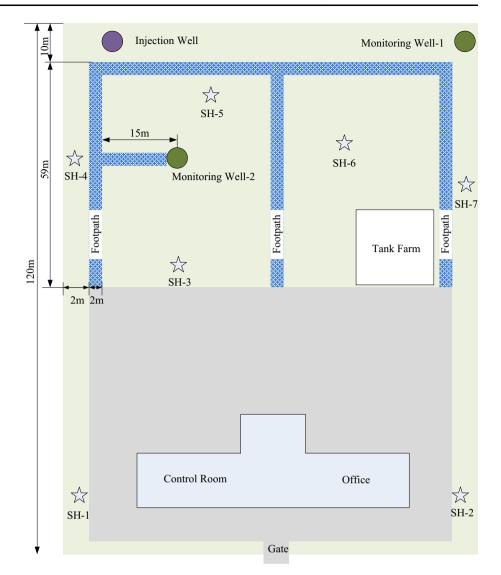
Three field applications

The cases 10–13 in Fig. 3 using the novel U-tube samplers were deployed at the Shengli CO₂-enhanced oil recovery (CO₂-EOR) project (Li et al. 2014; Lv et al. 2015), the Shenhua CCS project, the Jilin CO₂-EOR project, and plan to be deployed at the Yanchang CO₂-EOR project.

The Shengli CO₂-EOR project (Dongying, Shandong Province, China) began in 2012. In this project, 1.5 million kilograms of CO₂ with a purity of 99.5 % was injected into the (CO₂-EOR) oilfield to recovery. The U-tube sampling technology was used in this demonstration project to monitor and identify potential CO₂ leakage from the subsurface (Li et al. 2014; Liu et al. 2015) and also to assess induced health, safety, and environmental (HSE) risks (Li et al. 2013).

Three sets of U-tubes were deployed in the first stage at a depth of 10 m to obtain samples of groundwater and soil gas with a maximum three-block separation, e.g., -2, -6, and -10 m (Table 1). The preliminary analyses of the underground fluids were obtained both on-site and at a laboratory, and the results were significantly different from the analysis of nearby ditch water. It is worth noting that only soil gas above the water table was acquired, and the

Fig. 5 Distribution of the U-tube samplers at the Shenhua CCS demonstration site. *Symbol stars* indicate the location of the U-tube sampler



residual gas of unsaturated aquifers was excluded in this version of the U-tube sampler.

However, the U-tube samplers were broken after first sampling operation, which was due in part to severe blockages in tubes and/or damage to the check valves because of clay and sand particles. There was evidence of contaminated fluid from the sample-leg tube and of a much higher driving pressure during sampling operation, even no sample could be from certain broken layers at last. To look into these matters, a typical broken U-tube sampler was dug out and checked out carefully and thoroughly. We found that some of the check valves could be no longer closed in reverse, and several blockages due to clay and sands had occurred in tubes, especially which look crooked or full of twists and turns. Some other technical problems were also identified and overcome by design alteration, such as underground water cannot seepage continually into the fluid chamber due to internal air accumulation, and tubes were easy to connect in false position which makes the U-tube not work properly.

The Shenhua CCS demonstration project (located in Ordos City, Inner Mongolia, China) was the first pilot project for deep saline aquifer storage of captured CO_2 in China, with a capacity of 0.1 million tonnes of CO_2 per year from an existing coal-to-liquids facility (Liu et al. 2014; Wu 2014). With its pioneering multilayered injection and layer-by-layer monitoring, this project used one injection well and two monitoring wells, with the latter being used to monitor the temperature/pressure changes and obtain fluid samples in the storage layers and above the cap rock.

There are seven sets of U-tube samplers deployed at the field site (Figs. 5, 6), and each U-tube sampler contains three layers to obtain samples of groundwater at depths of 12, 16, and 20 m, and it contains two layers to collect soil gas at depths of 2 and 8 m. To date, after overcome



Fig. 6 Sampling tests at the Shenhua CCS project

Table 1Monitoringframework of the U-tubesamplers at the Shengli CO2-EOR project

Main type	Objectives	Frequency
Soil gas	1. CO ₂ concentrations in soil gas	Monthly
	2. Soil gas components	
Fluid samples at -2 m	1. Obtain continuous geochemical water	Monthly
Fluid samples at -6 m	2. CO_2 leakage or not	
Fluid samples at -10 m	3. Fluids subsurface transport	

blockage, fragile, and other technical obstacles, all seven of the U-tube samplers have been running successfully nearly two years since their deployment in 2014. There are several sampling data revealed in Table 2 at this stage, which samples from two multilevel U-tube samplers at different seasons. The pH varies from 6.5 to 7.4 and decreases by the depth of sampling underground. Also, the original data sampling from -16 to -20 m shows obviously differentiation. Furthermore, there are certain correlations between the concentration of Cl⁻, Mg²⁺, Ca²⁺ changed by depth and sampling time, as well as correlations and the conductivity and pH value. These sampling data are evident that no CO₂ leakage happened yet in the Shenhua CCS demonstration project, which are consistent with other monitoring results including atmospheric CO₂ concentration and soil CO₂ flux monitoring in this stage (Guo et al. 2015).

In addition, eight sets of U-tube samplers deployed in the Jilin CO_2 -EOR project (located in Songyuan City, Jilin, China) in November 2015 are all functioning properly. Moreover, as a part of monitoring program, preliminary results in this site were also revealed (Zhang et al. 2015).

Drawbacks and future plans of the U-tube sampling technology

Wireline fluid sampling is suitable for use in the temporary and short-term periodic monitoring projects, due to the sampling device should be retrieved from the well. Pump sampling and U-tube sampling rely on the dedicated installation of components in the well. Another crucial difference between U-tube sampling and other types of samplers is to produce single-phase samples without degasification or precipitation, due to its potential excellent behavior in pressure and temperature control during sampling deeper than 1000 m. The third difference between them is the sampling volume and whether steady or not, since wireline sampling methods collect a grab sample the other a steady stream of sample. Additionally, both pump samplers and U-tube samplers can collect multiple-layer samples that depend on packers to isolate the different horizons.

An ideal case is to have several samplers available to substitute for each other in case of failure/blockage. The Frio site in Texas and the Cranfield site in Mississippi are examples of sites

		TT		5		Co E				A. 2+ / F /	+
INUMDER	Sampling time	ЬН	TDS (mg/L)	Sal	EC (µs/cm)	I (C)	CI (mg/L)	SO4 ⁻ (mg/L)	Na' (mg/L)	Mg ⁻¹ (mg/L)	Ca ⁻ (mg/L)
SH-2-16-1	May. 30, 2015	7.192	I	I	1755	25.8	494.576	61.916	27.846	37.604	330.776
SH-2-16-2	Aug. 31, 2015	6.541	432	0.1	405	18.4	462.08	40.826	16.688	29.986	347.786
SH-2-16-3	Nov. 11, 2015	7.017	463	0.1	457	17.6	I	44.368	13.732	13.172	97.652
SH-2-20-1	May. 30, 2015	7.338	I	I	917	25.6	196.51	53.47	23.264	22.64	165.592
SH-2-20-2	Aug. 31, 2015	6.612	1683	0.8	1698	19	59.642	13.584	12.074	10.682	101.57
SH-2-20-3	Nov. 11, 2015	7.135	416	0.1	415	18.7	11.882	27.366	13.37	11.522	88.41
SH-3-16-1	May. 30, 2015	7.402	I	I	1301	25.3	315.64	65.986	28.524	468.3	235.888
SH-3-16-2	Aug. 31, 2015	6.705	1078	0.5	1076	15.7	286.792	74.728	19.456	36.788	251.806
SH-3-16-3	Nov. 11, 2015	6.982	2410	1.2	2410	17.4	768.356	109.114	25.18	79.322	388.874
SH-3-20-1	May. 30, 2015	7.596	I	I	471	25.5	72.296	45.054	20.946	18.054	96.47
SH-3-20-2	Aug. 31, 2015	6.811	2230	1.1	2240	16.6	69.992	20.536	14.176	13.918	106.858
SH-3-20-3	Nov. 11, 2015	6.82	1081	0.5	1081	19.1	238.31	44.332	16.722	31.362	168.894

(We also syste litech whie accu engilots E date mat bloc Num and A tech fore resp alon U-tu such pacl sele bent cher com and

means no data in this column. Partly because equipment cannot work and not measured at May 30, 2015. Partly due to under detection limit, like CI⁻ from SH-2-16-3

Page 11 of 13 1194

at which pump samplers, U-tube samplers, and Kuster I samplers were used for the different injection and monitoring wells (Wolff-Boenisch and Evans 2014). Bailers and pump samplers also worked together in a multi-level groundwater monitoring system introduced by Einarson and Cherry (2002).

In conclusion, as a new choice, the U-tube sampling technology is quite suitable in dedicated monitoring projects which specifically require three-dimensional tracing and accurate fluid analysis in a long term. Nevertheless, several engineering failures had happened during application and lots of technical breakthroughs still remain to be overcome.

Different kinds of technical problems have occurred to date in the field applications. For example, hydrate formation and freezing unexpectedly occurred at the Frio and natural waxy alkanes at the Otway (Freifeld 2010), blockage at the Cranfield (Hosseini et al. 2013), at the Nunuvut (Freifeld 2010), as well as the Shengli CO_2 -EOR and the Linying field test in China.

Also, several concerns remain about the U-tube sampling technology in addition to those mentioned above. First and foremost, the U-tube sampler seems quite fragile with respect to blockage of the check valve and failure or leakage along the length of the tubing. Secondly, the number of U-tube assemblies installed in a limited hole is restricted, such as 5 layers in a diameter of 100 mm. Thirdly, the packer used to seal off the overlying formation should be selected cost and technical feasibility, since cement or bentonite clay added to the drilling hole might impact the chemical quality of the downward percolating water.

Over years, manufactures have improved the design, construction, and materials used in many sampling devices, and in some cases, this has improved their performance. However, new fluid sampling technologies should be developed and added to the market to satisfy the increasing engineering demands, such as accuracy and cost-effective overall. Like flow rates for pumps sampling, an important consideration in future for the U-tube using in deep is temperature control, which can affect the solubility and volatility of constituents in the groundwater (Parker 1994). These and other important concerns should be addressed with respect to U-tube sampling technology to improve the accuracy of the samplers even further.

Design and material issues for sampling devices will still need to be considered in future studies. Specifically, new generation of the U-tube or modified related multilevel sampling technology would hopefully appear in near future.

Summary

Ĵ.

Borehole sampling technologies are typically divided into three categories based on driving forces, i.e., wireline sampling, pump sampling, and gas-operating sampling. A brief comparison of different sampling methods is presented, especially on the effects of water quality. Then, we focused on improvements in the U-tube sampling technology, which are shown below:

- The history of development. It was in (1973) that Wood used a check valve in an early version of suction-lift sampling technology. Thereafter, Freifeld developed the U-tube sampling technology in 2004, and this is the time to make the U-tube real sense.
- 2. Applications around the world. Beginning from 2005, 7 of the 13 sites have been mostly deployed in North America, within it the deepest application beyond 3200 m. Another 5 applied in China after 2009.
- 3. Although the current applications in China are not as deep as the ones in North America, it is worth noting that a novel U-tube sampler was designed specifically for shallow subsurface. So far, three field applications demonstrate the sampler feasible both technologically and economically.

Finally, the benefits, drawbacks and future developments of U-tube sampling technology were concluded.

Acknowledgments The authors thank the members of the large interdisciplinary team working on several CCUS projects in China for their significant contributions to this work. In addition, Zhang Yuanyuan from the Shengli oilfield, Wang Yongshen from the Shenghua CCS demonstration project, Zhang Deping and Li Qing from the Jilin oilfield, and Wei Ning, Wang Haibin and Xiao Wei from the Institute of Rock and Soil Mechanics are acknowledged with great appreciation for their coordination of the pilot tests and field sampling activities. Li Qi is grateful for the financial support provided by the Natural Science Foundation of China (Grant No. 41274111).

References

- Barcelona MJ, Helfrich JA, Garske EE (1985) Sampling tubing effects on groundwater samples. Anal Chem 57:460–464
- Barcelona MJ, Helfrich JA, Garske EE, Gibb JP (1984) A laboratory evaluation of ground water sampling mechanisms. Ground Water Monit Rev 4:32–41
- Barth JAC, Nowak ME, Zimmer M, Norden B, van Geldern R (2015) Monitoring of cap-rock integrity during CCS from field data at the Ketzin pilot site (Germany): evidence from gas composition and stable carbon isotopes. Int J Greenh Gas Control 43:133–140. doi:10.1016/j.ijggc.2015.10.017
- Boreham C, Underschultz J, Stalker L, Kirste D, Freifeld B, Jenkins C, Ennis-King J (2011) Monitoring of CO₂ storage in a depleted natural gas reservoir: gas geochemistry from the CO2CRC Otway Project, Australia. Int J Greenh Gas Control 5:1039–1054. doi:10.1016/j.ijggc.2011.03.011
- Briggs LJ, McCall AG (1904) An artificial root for inducing capillary movement of soil moisture. Science 20:566–569. doi:10.1126/ science.20.513.566
- Chen Z-A, Li Q, Liu L-C, Zhang X, Kuang L, Jia L, Liu G (2015) A large national survey of public perceptions of CCS technology in China. Appl Energy 158:366–377. doi:10.1016/j.apenergy.2015. 08.046

- Choi J-W, Nicot J-P, Hosseini SA, Clift SJ, Hovorka SD (2013) CO₂ recycling accounting and EOR operation scheduling to assist in storage capacity assessment at a U.S. Gulf Coast depleted reservoir. Int J Greenh Gas Control 18:474–484. doi:10.1016/j. ijggc.2013.01.033
- Einarson MD, Cherry JA (2002) A new multilevel ground water monitoring system using multichannel tubing. Ground Water Monit Rem 22:52–65. doi:10.1111/j.1745-6592.2002.tb00771.x
- England CB (1974) Comments on 'a technique using porous cups for water sampling at any depth in the unsaturated zone' by Warren W. Wood. Water Resour Res 10:1049
- Freifeld BM (2010) The U-tube: a new paradigm in borehole fluid sampling. Sci Drill. doi:10.2204/iodp.sd.8.07.2009
- Freifeld BM, Trautz RC, Kharaka YK, Phelps TJ, Myer LR, Hovorka SD, Collins DJ (2005) The U-tube: a novel system for acquiring borehole fluid samples from a deep geologic CO₂ sequestration experiment (1978–2012). J Geophys Res Solid Earth 110:B10203. doi:10.1029/2005JB003735
- Guo J, Wen D, Zhang S, Xu T, Li X, Diao Y, Jia X (2015) Potential and suitability evaluation of CO₂ geological storage in major sedimentary basins of China, and the demonstration project in Ordos Basin. Acta Geol Sin Engl Ed 89:1319–1332
- Hosseini SA, Lashgari H, Choi JW, Nicot J-P, Lu J, Hovorka SD (2013) Static and dynamic reservoir modeling for geological CO₂ sequestration at Cranfield, Mississippi, USA. Int J Greenh Gas Control 18:449–462
- Houghton RL, Berger ME (1984) Effects of well-casing composition and sampling methods on apparent quality of ground water. Paper presented at the proceedings of the fourth national symposium on aquifer restoration and ground water monitoring, The Fawcett Center, Columbus, Ohio, USA, pp 203–213
- Hovorka SD, Meckel TA, Trevino RH (2013) Monitoring a largevolume injection at Cranfield, Mississippi—project design and recommendations. Int J Greenh Gas Control 18:345–360. doi:10. 1016/j.ijggc.2013.03.021
- Jenkins C, Chadwick A, Hovorka SD (2015) The state of the art in monitoring and verification—ten years on. Int J Greenh Gas Control 40:312–349. doi:10.1016/j.ijggc.2015.05.009
- Lei X-L, Li X-Y, Li Q, Ma S-L, Fu BH, Cui YX (2014) Role of immature faults in injection-induced seismicity in oil/gas reservoirs: a case study of the Sichuan Basin, China. Seismol Geol 36:625–643. doi:10.3969/j.issn.0253-4967.2014.03
- Li Q et al (2014) A novel shallow well monitoring system for CCUS: with application to Shengli oilfield CO₂-EOR project. Energy Proced 63:3956–3962. doi:10.1016/j.egypro.2014.11.425
- Li J, Li X (2015) Analysis of U-tube sampling data based on modeling of CO_2 injection into CH_4 saturated aquifers. Greenh Gases Sci Technol 5:152–168
- Li Q, Liu G, Liu X, Li X (2013) Application of a health, safety, and environmental screening and ranking framework to the Shenhua CCS project. Int J Greenh Gas Control 17:504–514. doi:10.1016/ j.ijggc.2013.06.005
- Li Q, Song R, Liu X, Liu G, Sun Y (2016) Monitoring of carbon dioxide geological utilization and storage in China: a review. In: Wu Y, Carroll J, Zhu W (eds) Acid gas extraction for disposal and related topics. Wiley-Scrivener, New York, pp 331–358. doi:10.1002/9781118938652.ch22
- Li X, Pan D, Ye C, Zheng J (2015) Experiment and research of air exchange underground water sampler. Yangtze River 46:43–45+64
- Liu L-C, Leamon G, Li Q, Cai B (2014) Developments towards environmental regulation of CCUS projects in China. Energy Proced 63:6903–6911. doi:10.1016/j.egypro.2014.11.724
- Liu X, Li Q, Fang Z, Liu G, Song R, Wang H, Li X (2015) A novel CO₂ monitoring system in shallow well. Rock Soil Mech 36:898–904. doi:10.16285/j.rsm.2015.03.001

- Lu J et al (2012) CO_2 -rock-brine interactions in lower Tuscaloosa formation at Cranfield CO_2 sequestration site, Mississippi, USA. Chem Geol 291:269–277
- Lv GZ, Li Q, Wang S, Li X (2015) Key techniques of reservoir engineering and injection-production process for CO₂ flooding in China's SINOPEC Shengli Oilfield. J CO2 Util. doi:10.1016/j. jcou.2014.12.007
- Martens S, Möller F, Streibel M, Liebscher A (2014) Completion of five years of safe CO₂ injection and transition to the post-closure phase at the Ketzin pilot site. Energy Proced 59:190–197. doi:10. 1016/j.egypro.2014.10.366
- Oy P (2011) The greenland analogue project: yearly report 2009. Olkiluoto, Eurajoki, Finland
- Parizek RR, Lane BE (1970) Soil–water sampling using pan and deep pressure-vacuum lysimeters. J Hydrol 11:1–21
- Parker LV (1994) The effects of ground water sampling devices on water quality: a literature review. Ground Water Monit Rem 14:275
- Paterson L et al (2013) Overview of the CO2CRC Otway residual saturation and dissolution test. Energy Proced 37:6140–6148. doi:10.1016/j.egypro.2013.06.543
- Puls RW, Powell RM (1992) Acquisition of representative ground water quality samples for metals. Ground Water Monit Rem 12:167–176. doi:10.1111/j.1745-6592.1992.tb00057.x
- Rutqvist J, Freifeld B, Min K-B, Elsworth D, Tsang Y (2008) Analysis of thermally induced changes in fractured rock permeability during 8 years of heating and cooling at the Yucca mountain drift scale test. Int J Rock Mech Min Sci 45:1373–1389
- Stackhouse BT et al (2010) Chemical and microbial analysis of a talik in western Greenland. Paper presented at the American Geophysical Union (AGU), Fall Meeting San Francisco, 13–17 December, 2010, CA
- Stalker L, Boreham C, Underschultz J, Freifeld B, Perkins E, Schacht U, Sharma S (2015) Application of tracers to measure, monitor and verify breakthrough of sequestered CO₂ at the CO2CRC Otway Project, Victoria, Australia. Chem Geol 399:2–19. doi:10. 1016/j.chemgeo.2014.12.006

- Tsang YW et al (1999) Yucca mountain single heater test final report: Yucca mountain site characterization project. Lawrence Berkeley National Laboratory, Berkeley, CA. doi:10.2172/6495
- Wagner GH (1962) Use of porous ceramic cups to sample soil water within the profile. Soil Sci 94:379–386
- Wei N, Li X, Wang Y, Zhu Q, Liu S, Liu N, Su X (2015) Geochemical impact of aquifer storage for impure CO₂ containing O₂ and N₂: Tongliao field experiment. Appl Energy 145:198–210. doi:10.1016/j.apenergy.2015.01.017
- Wolff-Boenisch D, Evans K (2014) Review of available fluid sampling tools and sample recovery techniques for groundwater and unconventional geothermal research as well as carbon storage in deep sedimentary aquifers. J Hydrol 513:68–80. doi:10.1016/j.jhydrol.2014.03.032
- Wood WW (1973) A technique using porous cups for water sampling at any depth in the unsaturated zone. Water Resour Res 9:486–488
- Wu X-Z (2014) Shenhua group's carbon capture and storage (CCS) demonstration. Min Rep 150:81–84. doi:10.1002/mire. 201400006
- Xu T, Kharaka YK, Doughty C, Freifeld BM, Daley TM (2010) Reactive transport modeling to study changes in water chemistry induced by CO₂ injection at the Frio-i brine pilot. Chem Geol 271:153–164. doi:10.1016/j.chemgeo.2010.01.006
- Yang CB, Romanak K, Hovorka S, Holt RM, Lindner J, Trevino R (2013) Near-surface monitoring of large-volume CO₂ injection at Cranfield: early field test of SECARB Phase III. SPE J 18:486–494
- Zhang L, Ren B, Huang H, Li Y, Ren S, Chen G, Zhang H (2015) CO₂ EOR and storage in Jilin oilfield China: monitoring program and preliminary results. J Petrol Sci Eng 125:1–12. doi:10.1016/ j.petrol.2014.11.005
- Zhu Q, Li X, Jiang Z, Wei N (2015) Impacts of CO₂ leakage into shallow formations on groundwater chemistry. Fuel Process Technol 135:162–167. doi:10.1016/j.fuproc.2014.11.042