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Contamination Control for Scientific Drilling Operations

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Abstract

Drilling is an integral part of subsurface exploration. Because almost all drilling operations require the use of a drill fluid, contamination by infiltration of drill fluid into the recovered core material cannot be avoided. Because it is impossible to maintain sterile conditions during drilling, the drill fluid will contain surface microbes and other contaminants. As contamination cannot be avoided it has to be tracked to identify those parts of the drill core that were not infiltrated by the drill fluid. This is done by the addition of tracer compounds. A great variety of tracers is available and the choice depends on many factors. This review will first explain the basic principles of drilling before presenting the most common tracers and discussing their strengths and weaknesses. The final part of this review presents a number of key questions that have to be addressed in order to find the right tracer for a particular drilling operation.

Introduction

Exploration of deep subsurface environments relies on drilling. Many different drilling techniques are being used, the choice is mainly based on the type of sediment or rock to be penetrated and the maximum depth that has to be reached. Almost all drilling operations require the use of a drill fluid for cooling the bit, transporting cuttings out of the borehole and stabilizing the well (Kallmeyer et al., 2006). The most simple drill fluid is water, but in many cases this is not sufficient and additional compounds have to be added, e.g. clay minerals to increase specific gravity of the fluid and/or thickeners of variable composition (natural compounds like cellulose or guar gum or synthetic polymers).

Sterile drilling - an impossibility

As the drilling fluid travels from the surface down to the drill bit, it comes in contact with many surfaces (holding tank, pump, pipes, etc.) and will inevitably transport some surface material down into the borehole. From a geomicrobiological or biogeochemical perspective drilling is a very dirty business, even in relatively small drilling operations it is impossible to maintain sterile conditions or avoid contamination of the drill core with foreign compounds like hydrocarbons. A drill core is never completely pristine but will always have at least some contamination on the outside. A drilling rig and the associated drill rods very quickly add up to a total weight of many tons. The weight of the drill string varies significantly between different diameters and wall thicknesses, as a rule of thumb a 6 m piece of drill string for a deep drilling operation weighs between 100 and 150 kg, so a kilometre of string weighs 18 to 25 tons. Such huge amounts of equipment are much too large and heavy to be sterilized. Even if it were possible to sterilize an entire drill string of up to several kilometres length, it would be a futile exercise because as soon as the drill string enters the drill hole it will be immediately contaminated with microbes from the surrounding sediment or rock. Due to the fact that the drilling fluid is usually an opaque mixture of water and suspended particles that are larger than microbial cells, the fluid can neither be filter-sterilized nor UV treated. In cases where only small volumes of water without any additives can be used as a drill fluid, then pre-sterilization is indeed an option. However, this option is limited to rather small operations. In normal sized drilling operations, the massive volumes of drill fluid of up to hundreds of cubic meters and flow rates of hundreds of litres per minute preclude any sterilization. At best, the drilling equipment is thoroughly cleaned before use to avoid contamination with foreign hydrocarbons from the pipe grease or other chemicals and the drill mud is prepared with clean tap water instead of well or river water. There are several ways to keep the drilling operation as clean as possible, for example employing very strict cleaning protocols and carefully designing the operations around the drill rig with contamination avoidance in mind (Russell et al., 1992). So even under the best possible conditions drilling inevitably causes infiltration of non-sterile drilling fluid into the core, not just along cracks and fissures but also into the pore space of even undisturbed fine-grained sediments (McKinley and Colwell, 1996b; Smith et al., 2000a; Smith et al., 2000b). While drill fluid contamination is problematic for many analyses, it poses particular challenges for geomicrobiological studies.

Compared to the surface, microbial cell abundances in the subsurface are several orders of magnitude lower. As an example for typical cell abundances, a coastal shallow marine or lacustrine sediment contains between 10^6 to 10^8 cells cm^{-3}. In very organic-rich sediments from upwelling areas or other eutrophic systems, cell abundances are in the 10^9 cells cm^{-3} range but can reach, or in rare cases even exceed, 10^{10} cells cm^{-3} (Andrén et al., 2015). At the other end of the spectrum are deep subsurface sediments, which normally only have cell densities around 10^3 to 10^4 cells cm^{-3} (e.g. D'Hondt et al., 2015; Kallmeyer et al., 2012) or even less (Inagaki et al., 2015). So there are several orders of magnitude difference in cell abundance between the shallow and the deep subsurface. Thus, even the slightest infiltration of drilling fluid into a deep subsurface sample (in the order of nanoliters per cm^3 sediment) renders the sample unsuitable for microbiological and also certain geochemical investigations (Yanagawa et al., 2013). One could argue that it should be possible to avoid any contact of the drill fluid with surface sediments, use clean but not necessarily sterile equipment and to employ strict contamination control. All this is possible and has been done in in the past with various degrees of success. Still, preparation of the drill fluid is a key issue for minimizing contamination. Due to the large volumes of water that are required and the often remote location of the drill site it is often impossible to use relatively clean tap water; instead water has to be sourced locally from wells, springs, rivers or lakes instead. In ocean drilling the drilling liquid of choice is normally surface ocean water. However, even in the most extreme oligotrophic parts of the world's ocean cell abundance at the surface is still around 10^5 cells mL^{-1} (D'Hondt et al., 2011). In coastal waters or lakes cell abundances are in the 10^6 cells mL^{-1} range or higher (e.g. Daley and Hobbie, 1975; Noble and Fuhrman, 1998). So even under the best possible conditions, the drill fluid will have a cell concentration that is orders of magnitude higher than a deep subsurface sample and it will inevitably infiltrate at least into the outer layers of the drill core.

Drilling techniques

Drilling is an integral part of deep earth exploration. The review of (Wilkins et al., 2014) provides an excellent overview of terrestrial scientific drilling operations. A recent book (Stein et al., 2014) describes the state of the art and future challenges of deep life exploration in the marine realm.

Most scientists that had to deal with a large-scale drilling operation will agree that a very close collaboration between the science and drilling team a project from the earliest possible date a project is vital for its success. While no scientist should try to tell the drillers how to drill, one should be able to formulate the specific needs of a scientific drilling campaign. In my experience even the most unusual requirements can be accommodated with relative ease if they are communicated early enough. It also helps tremendously when scientists and drillers both speak the same language in terms of technical terms and definitions. Therefore before I start with a description of the different drilling techniques I would like to introduce the most common terms and definitions as they are often mixed up or used incorrectly, leading to confusion and misunderstandings.

Glossary of common terms

Drilling is a technique by which a hole is created, normally by a rotating drill bit, sometimes also by hammering. Drilling almost always includes the use of a drilling fluid. For scientific purposes drilling almost always includes the recovery of a core, but there are cases where no core is recovered and only the hole itself is of interest. Also, there are cases where only specific intervals are cored, the rest is just drilled, not cored. In industry coring is rarely done as it is very time consuming and therefore expensive. A key feature of drilling is the use of a so-called drill string, which is a set of steel pipes that connect the drill bit at the bottom of the hole with the drill rig at the surface. The drill string is extended by addition of more pieces of drill pipe. The drill fluid is pumped through the drill string, exits through the drill bit and travels upward through the drill hole, carrying the drill cuttings to the surface.

Coring is a term that is used to describe the retrieval of a core, irrespective whether drilling was used or not. Mainly in limnology and oceanography, cores of unconsolidated sediment are taken without drilling but with a gravity or piston corer (see below). Even in operations with continuous coring over hundreds of meters a core is not taken continuously but in sections of variable lengths, usually between 3 and 10 meters. Between the individual cores there is usually a small gap caused by mechanical disturbances from the drill bit or the core catcher. In cases where an absolute continuous record without gaps is required, e.g. for stratigraphy or paleoclimate research, multiple holes are being drilled. Coring at different holes will be arranged with an overlap in order to cover the gap in one hole with a continuous core in the other. So if a single core run will be 10 m long, then the first core of the second hole will be drilled only to 5 m depth to have sufficient overlap.

Core tube or core barrel: The central piece of any corer is the core tube or barrel, as it will retrieve and hold the core. The core barrel is usually made from metal, as it has to withstand the mechanical forces during coring and protect the inner plastic liner (see below) from breaking. Depending on the type of coring operation the core barrels have to be very sturdy and thick-walled to withstand the forces that occur when pushing into the sediment, especially in piston coring. These core barrels can weigh up to hundreds of kilograms, depending on the total length of the corer. For example the long piston corer from Woods Hole Oceanographic Institution has steel core barrels with wall thicknesses of 1.25”; each barrel is 20’ long and weighs 1500 lbs (fig 1).

Liner is a tube (normally plastic) that sits inside the core barrel and collects the core. After bringing the core barrel to the surface the liner with the core inside is pulled out of the barrel (fig 2a) and laid down horizontally and usually cut into sections of 1 or 1.5 m length to allow for easier handling (fig 2b). To prevent the core from falling apart it is left in the liner. Depending on the type of analysis, the horizontally placed core, including the liner, is either split open horizontally to see the internal structure of the cored material or cut vertically into small intact core pieces, the so-called whole round cores (WRC). The latter is commonly used geomicrobiological or biogeochemical research because the exposure to oxygen is reduced.

For taking short cores (usually <1 m) from very soft sediment small gravity corers are being used that do not have a separate core barrel and liner but only a single tube of either steel or plastic in which the core is collected. The recovered core is then usually pushed out of the tube, immediately sampled and the tube reused.

Core catcher: Core loss normally occurs once the corer is lifted up and the core slides down. To help prevent this loss of core, there is usually a check valve at the top of the core that lets the water out that is displaced by the drill core but closes and creates suction to keep the core inside the tube as soon as the corer is pulled upwards This way a vacuum is created that keeps the sediment inside the tube. However the longer the core the more likely suction alone will not hold the core in place and some loss will occur. To improve core recovery a core catcher is installed at the bottom of the core tube (fig 3).

Different mechanisms are being used but the most common one is a circle of plastic or metal lamellae, forming a circular flexible barrier. The core pushes the lamellae to the side and glides past them when entering but when the core tries to fall back out, the lamellae close and keep the core in place. However, the lamellae can disturb softer sediments as they enter. This is more the case with metal core catchers than with softer plastic ones. By choosing a core catcher with the right stiffness a compromise between optimal sample integrity and minimal core loss can be found.

Drilling/Coring techniques

Industry has developed many different drilling and coring techniques for almost any rock type or environment imaginable. However, only a few play a role in scientific drilling but even those few offer many options and might confuse a scientist at first.

In rare cases airlift drilling has been used for scientific purposes (Colwell, 1989; Colwell et al., 1992; McKinley and Colwell, 1996b), where air or an inert gas like Argon was used to lift the cuttings out of the hole. Given the rarity of airlift operations in scientific drilling it will not be further discussed in this paper. The study of (Colwell et al., 1992) is recommended for a good overview of the air-drill technique.

The next few paragraphs will describe those techniques that are most often used in scientific drilling.

Piston and gravity coring: Both techniques do not require the use of any drilling liquids and thereby reduce the chance of contamination with foreign material. However, they only work in soft, unconsolidated sediments, not in hard rocks. Quite often gravity or piston coring operations are called drilling, which is not correct; the correct term is gravity or piston coring. There is however an exception to this rule and that is hydraulic piston coring (see below), which is in fact a drilling technique. While the difference between coring and drilling seems rather semantic, it does make a major difference in terms of contamination control. While for piston or gravity coring operations contamination control is of minor concern and usually not employed due to lack of drilling fluid, it is absolutely crucial for drilling operations. So having a core sample from a drilling or coring operation does make a difference for many analyses.

In its simplest form a gravity corer consists of a core barrel that is closed by a flap valve at the top and a set of weights that allows it to penetrate the soft sediment. It hangs vertically on a rope or cable on which it is lowered towards the sea or lake floor. While a gravity core is lowered rather slowly from the vessel into the lake or seafloor, a piston corer is only lowered to a few meters above the bottom and then dropped in free fall by releasing a loop of additional cable. In a gravity corer the cable is connected to the head of the corer, whereas in a piston corer it is connected to a piston that sits at the bottom of the barrel. The length of the free fall is calculated so that when the corer touches the seafloor, the cable stops the piston right above the sediment surface while the corer continues its movement and penetrates the sediment. The vacuum created by the immobile piston on the sediment surface prevents the sediment from compaction caused by the downward moving tube. This way the sediment enters the tube comparatively undisturbed and allows for longer and heavier coring systems compared to gravity cores. Both gravity and piston corers have proven to provide largely undisturbed cores, but the short free fall of the piston corer allows for deeper penetration at the cost of operating a more complicated and failure-prone system. The maximum depth limit for gravity coring is usually in the 10m range. Piston corers allow for deeper penetration. The largest systems (Calypso corer of French research vessel Marion Dufresne and Woods Hole Long Core system, fig 1) reach maximum depths around 50 m.

Hydraulic piston coring (HPC): This is the most common technique for recovering non- to semi-consolidated sediment from greater depths beyond the limits of gravity or piston coring. In ocean drilling operations this technique is also called APC (Advanced Piston Coring). Depending on the sediment properties cores in excess of 400 m length can be recovered by this technique (Pälike et al., 2010). For HPC operations a core tube with a liner inside, a flap valve on top and a core shoe (fig 3&4) with a core catcher at its bottom forms a close seal at the bottom end of the drill string, so the drill fluid inside the string can be pressurized. Eventually the pressure exceeds the breaking strength of the shear pins that hold the core tube in place. By selecting the right number and type of shear pins, the drillers can adjust to varying lithologies. After breaking of the shear pins the core tube shoots forward and pushes into the sediment. The core shoe has to cut through the sediment and might get damaged when hitting harder layers or pieces of gravel (fig 4). When the core tube has come to a stop, a circular drill bit will drill around the core tube and extend the drill hole. During this operation the drill string will move downward and push the core tube back inside the string. After extending the hole to the maximum depth that was reached by the core tube, the core tube will be pulled upwards for retrieval of the drill core. HPC is usually used in combination with wireline coring (see below).

Extended Core Barrel (XCB): In case the sediment becomes too stiff for HPC but is still too soft for rotary drilling other tools are being used, the most common one is XCB, which features a short (<50cm) core barrel that extends forward from the actual drill bit, hence the name extended core barrel (fig 3). As the drill bit moves downward, the core barrel pushes into the sediment ahead of the bit. There are different versions of this tool, some have a non-rotating barrel, others have a rotating one. In some cases the barrel is spring loaded, others are fixed. There are different names for these tools: extended nose, extended shoe, extended core bit, etc. As different as these systems are, they all have in common that the quality of the recovered cores is usually not as good as the HPC cores. Very often they are mechanically disturbed and contaminated by drilling fluid and therefore unsuitable for geomicrobiological analyses. Despite many years of development, there is still no suitable technique available that delivers high quality cores from sediments that can neither be cored by HPC or RCB.

Rotary drilling (RCB): For real hard lithologies rotary drilling is being used. If the sediment is still too soft, then it will be fractured into small pieces and more or less destroyed. In RCB operations a core bit equipped with either three or four rotating cones (fig 3) or a ring-shaped crown cuts the rock.

Wireline Coring: Wireline coring is used in almost every deep drilling operation where coring is an important component. It can be used in both rotary coring and hydraulic piston coring operations. Its main advantage is that it is not necessary to pull the entire drill string out of the hole in order to retrieve the core and replace the liner but to retrieve many consecutive cores through the drill string. In shallower terrestrial drilling operations (up to a few tens to hundred meters) with stable boreholes it is not much of a problem to pull out the entire drill string for every few meters of core. In cases where stability of the borehole is an issue, e.g. in sandy aquifers, pulling the string out of the hole should be avoided as this will cause additional disturbance that might lead to a caving or even collapse of the hole.

Pulling the drill string out of the hole becomes a huge problem for deeper drilling operations or for operations from drill ships or swimming platforms. It takes several hours to move one or more kilometres of string in and out of the hole. Although a re-entry into a drill hole on the sea or lake floor is possible it is technically challenging and requires specially equipped ships or drilling platforms with highly accurate dynamic positioning systems. Also, a large re-entry cone has to be installed on the sea floor to help re-entering the drill string into the hole. Given the technical difficulties and required additional equipment and time for a re-entry it might be cheaper to drill a new hole instead of re-entering an existing one.

For wireline coring a unit consisting of a core tube with a liner inside, a flap vale and a core-retrieval mechanism (the so-called spearhead or fishneck, fig 5) at the top and a core catcher at the bottom is sent through the drill string to the bottom of the hole, where it attaches itself to the bottom segment of the string, the so-called bottom hole assembly (BHA). After coring has commenced, a cable with a catching mechanism, the so called overshot assembly is lowered through the string, connects itself to the core tube and releases it from the BHA. Then the core tube is pulled up. Upon retrieval the core tube is laid horizontally, the core catcher removed and the liner with hopefully a core inside pulled out of the core tube. Then a new liner is loaded, the core catcher fixed and the unit is ready for another trip down the drill string.

Contamination tracers

Because contamination cannot be avoided, at least not completely, it is essential to trace contamination of the drill core to identify uncontaminated samples. In order to assess the degree of infiltration a tracer is added to the fluid. In order to attribute the detected tracer to the infiltration of drilling fluid into the sample it is necessary that tracers (1) have no natural source, (2) are easy to detect even at extremely low concentrations, (3) are chemically inert.

Several techniques have been used in past drilling operations to assess microbial contamination, including fluorescent dyes (Pellizzari et al., 2013; Phelps et al., 1989; Russell et al., 1992), Perfluorocarbon tracers (PFT) (Colwell et al., 1992; House et al., 2003; Lever et al., 2006; Russell et al., 1992; Smith et al., 2000a; Smith et al., 2000b) microsphere tracers (Colwell et al., 1992; Kallmeyer et al., 2006; Smith et al., 2000b; Yanagawa et al., 2013), dissolved salts like lithium bromide (Haldeman et al., 1995), potassium bromide (Phelps et al., 1989) and barium (Chapelle and Lovley, 1990), sulfonic acids (Hirtz et al., 2001), foreign microbes like cyanobacteria (Colwell et al., 1994) or fluorescent proteins (Juck et al., 2005). Other studies used molecular biological techniques to differentiate between the microbial community of the drill fluid and the sample (Chandler et al., 1997; Gronstal et al., 2009).

(Gronstal et al., 2009) give a good overview of the different scientific drilling operations and their respective methods for contamination control.

Fluorescent dyes

Fluorescent dyes like fluorescein or Rhodamine are inexpensive, easy to handle and allow sensitive detection of contamination (Russell et al., 1992). Another major advantage is the fact that fluorescein is non-toxic and has been used as a groundwater tracer for a long time. In areas with strong legal constraints on drilling, e.g. close to drinking water wells, fluorescein might be the only tracer for which a permit can be obtained as the authorities already have experiences with it. The detection limit for fluorescein in an aqueous solution without any interfering compounds is in the order of 0.05 ppb (Gunderson et al., 2002), however in actual geologic samples the detection limit is more in the range of 1 ppb (Pellizzari et al., 2013), other dyes are in a similar range. Detection and quantification is easy, the only required equipment is a fluorometer. Pellizzari et al. (2013) provides a good description of the protocol to extract the dye, in this case fluorescein, from the drill core sample and to measure a large number of samples at once with a plate reader.

In most cases the dye concentration in drill mud is in the ppm range (mg L^{-1}), resulting in a detection range of ca. three orders of magnitude. Although a detection limit in the ppb range sounds impressive, it also means that a drill fluid contamination in a concentration around 0.5 µl cm^{-3} would not be detected, assuming a dye concentration of 1 ppm. Using normal lake or seawater with a microbial cell concentration of ca. 10^6 cells cm^{-3} as a drill fluid, then the sample contains up to 500 foreign cells per cm^3. For a drill core from shallow depths or from an organic-rich deposit with concomitantly high cell counts (i.e. above 10^5 cells cm^{-3}) these 500 cells cm^{-3} only represent less than 0.5 % of the community. Depending on the planned analysis this might not be much of an issue. However, even in shallow subsurface samples with a large indigenous microbial population, introduction of 500 foreign cells from the surface renders the sample unusable for cultivation approaches because subsurface microbes normally have much lower metabolic activity and growth rates than surface microbes (Hoehler and Jorgensen, 2013; Jørgensen, 2011) and can therefore be outcompeted by the introduced microbes.

For deep subsurface environments with only a few hundred or thousand cells per cm^3 the situation is much worse as even such a small (and in case of a dye tracer undetectable) contamination would massively change the microbial community composition. Considering the fact that a contamination of 0.5 μl drill fluid per cm^3 of sample would fall below the minimum detection limit, a more sensitive contamination control method would be required.

Also, fluorescent dyes have other potentially problematic features. Diehl and Horchak-Morris, 1987) showed that fluorescein is sensitive to light degradation. Normally the drill fluid or drill mud is stored in large holding tanks that are open at the top. So while it is easy to mix the tracer with the drill fluid in the holding tanks, it might decay to a certain degree and thereby change its initial concentration and lower the minimum detection limit.

Another aspect that has to be taken into account when using fluorescent dyes is their sensitivity to low pH values. In a detailed study about the fluorescence intensity of fluorescein and several other compounds that are being used as tracers (Zhu et al., 2005) showed that intensity remains largely stable in the alkaline range up to pH 10.5, but decreases in the acidic range, this trend seems to be more pronounced at higher concentrations.

Sorption of fluorescent dyes onto clays is another important aspect that has to be taken into account (Magal et al., 2008). Clays are a common drill mud additive, they are used to increase density in order to improve the capability of the mud to carry cuttings out of the hole and to stabilize the walls of the drill hole. The sorption characteristics depend on the type of mineral and dye, so no general recommendations can be given, only the strong advise to carefully test the tracer before deciding on its use in a drilling operation. Also, in a detailed study about factors that influence fluorescence intensity of fluorescein (Weidner et al., 2011) showed that dissolved Fe^{2+} and Mn^{2+} can significantly decrease the signal. While this might not be of general concern it should be taken into account when drilling through metal-rich formations or through aquifers with iron-rich waters.

When drilling through organic-rich deposits like peat or coal the drill fluid can extract substantial amounts of humic substances, which react with the fluorescent dyes and cause quenching, thereby decreasing the fluorescence signal (Hafuka et al., 2015). Moreover, fluorescent dyes will stain the entire drilling fluid in a bright colour (fig 6), which might cause problems for disposal of the mud after drilling. Although fluorescent dyes are the tracer that is most easy to obtain and use, there are several limitations that have to be taken into account as they limit the applicability of fluorescent dyes in deep drilling campaigns

Perflourocarbon Tracers (PFT)

Perfluorocarbon tracers (PFT) are fluorinated hydrocarbons that have no known natural source. These tracers have been used extensively in drilling operations on land and at sea (Lever et al., 2006; McKinley and Colwell, 1996a; Russell et al., 1992; Smith et al., 2000a). A common PFT tracer is Perfluoromethylcyclohexane, which has boiling point of 76°C, its solubility is ~1 mg/L in water and 10 g/L in methanol (Colwell et al., 1992). Its low solubility in water combined with a low boiling point facilitates gas phase partitioning through heating of the sample followed by quantitative headspace analysis via electron capture gas chromatography (GC-ECD). This is by far the most sensitive detection method for any tracer, reaching down to levels of ca. $2*10^{-12}$ g PFT, which translates to a minimum detection limit for drill mud infiltration in the range of 4 to 5 nl cm^{-3} (Lever et al., 2006).

Because PFT has such a low solubility in water and a low boiling point it cannot be pre-mixed into the drill mud but has to be fed constantly into the mud immediately before being pumped down the drill hole. The normal rate at which the PFT is introduced into the mud stream is 1 mg/L. For feeding the PFT into the mud stream HPLC pumps have proven to be a good choice. Another PFT that has been used in scientific drilling is gaseous Halon 1211 (Gronstal et al., 2009). During drilling operations at the Cheaspeake Bay impact structure a gas mixture of 1% Halon in N_2 was added into the mud stream to reach a final concentration of 1 ppm Halon. Due to the pressure in the borehole the gas completely dissolved into the drill mud.

Core samples for contamination control have to be taken as quickly as possible after retrieval of the core to avoid any losses due to evaporation (Gronstal et al., 2009). Samples have to be placed immediately in gas-tight (usually glass) vials and sealed with a septum. After heating the vial to facilitate partitioning of PFT into the headspace a small (0.5 to 5 ml) gas sample is taken from the headspace and analysed via GC-ECD. Smith et al. (2000a,b) note that the glass syringe used for transferring the sample from the vial to the GC should also be heated to 70° C to avoid adsorption of PFT onto the glass walls. Due to its high volatility and extremely low detection limit, they recommend that all handling of PFT should be carried out in a well-ventilated area and away from the drilling operations or sample handling areas to avoid false positives.

Given the high sensitivity of detection, PFT should be the method of choice for any biogeochemical or geomicrobiological coring operation. However, there are good reasons for not using it. The first one is the required technology for delivering the tracer into the drill mud. Because it cannot be premixed, a small pump has to be installed and the tracer fed into the mud stream at a rate that is proportional to the flow rate of the mud pump. Depending on the type of drill rig and the willingness of the drilling team to cooperate, it might be technically and strategically challenging or impossible to install a delivery system for PFT. The second reason for not using it under all circumstances is its high volatility. If a contamination control sample is not taken shortly after retrieval of the core, an unknown fraction of PFT will have evaporated from the sample. So under conditions where the cores cannot be subsampled immediately after retrieval, e.g. in lake drilling operations, where only a very small drilling barge is being used that does not provide any space for detailed subsampling, the PFT will be gone by the time the cores reach the laboratory. And even if an initial subsampling immediately after retrieval is possible, the high volatility of PFT still precludes detailed contamination control at a later date. In many cases highly specialized and laborious, time-consuming analyses are not carried out at the drill site, but later in the home lab. Usually each group that plans detailed analyses in the home lab receives a single larger WRC from a sampling interval instead of subsampling immediately for each measurement. This way the processing time at the drill site can be minimized, which is often necessary to keep up with the core flow. All further subsampling is then carried out right before the analyses. In order to make sure that every individual sample is in fact not contaminated it would be highly desirable to measure the level of contamination on every individual sample prior to analysis. When using PFT this would not be possible because the PFT has long since evaporated.

Another issue is the detection of PFT, which requires a GC with an electron capture detector (ECD). An ECD is mainly used for pesticide detection and in pharmaceutical research, it is not a common detector in biogeochemistry and therefore not readily available in most geomicrobiology and biogeochemistry laboratories. So unless a suitable system can be borrowed, it has to be included in the budget. For a one-off operation it might be too expensive. In cases where largely unknown lithologies are drilled it is extremely helpful to have at least a few contamination control results right at the drill site in order to adjust the drilling strategy. This however requires a GC-ECD on site, including a supply of a suitable high-purity carrier gas. Depending on the location of the drilling operation, this might be a very challenging task. PFT is the standard contamination control tracer on both drill ships of the Integrated Ocean Discovery Program (IODP), but they have all required equipment permanently installed and all necessary protocols in place, optimized over decades. In other operations, e.g. smaller drilling operations at remote locations like on a tropical lake in the rainforest of Sulawesi (Russell et al., 2016), in the desert (Cohen et al., 2016) or in the high arctic (Dallimore et al., 2005) it might be a considerable challenge to organize the logistics to have such a system up and running at the drill site.

Microspheres and other particulate tracers

Another very popular tracer are microspheres, sometimes also called microbeads. These are microbe-sized fluorescent plastic particles. Microspheres are available from several suppliers in many different colours and size ranges. The rationale for using microspheres is that particles that have the same size as microbes will, when mixed into the drill mud, penetrate the sample in a similar fashion as microbes and therefore mimic their distribution in a sample. Detection and quantification of microspheres by fluorescence microscopy is relatively easy and fast (fig 7). For contamination control often green fluorescent microspheres with 0.5 µm diameter are used, as this is the average size of a subsurface microbe (Kallmeyer et al., 2012) and detection is possible with the same optical filter set as for the common stains Acridine Orange or SYBR Green I. The minimum detection limit is between 10^5 and 10^6 particles cm^{-3} (Kallmeyer et al., 2006), similar to subsurface cell counting without cell separation (Fry, 1988; Kallmeyer, 2011; Morono et al., 2009). However, separating the microspheres from the sample by density separation can lower the detection limit by at least one order of magnitude (Kallmeyer et al., 2006). Using the equation of (Kallmeyer et al., 2008) and assuming that the drill fluid has a microbial cell concentration of 10^6 cells ml^{-1} and a microsphere concentration in the range of 10^9 particles ml^{-1} (Friese et al., submitted) calculated the minimum detectable concentration of drill mud infiltration to be 117 nl cm^{-3}, or 117 foreign cells cm^{-3}. This is better than fluorescent dyes but not as sensitive as PFT.

Microspheres have many advantages over fluorescent dyes and PFT. Except for temperatures >100° C that can be encountered in drilling operations in geothermal systems (Yanagawa et al., 2013), they are inert under most physical and chemical conditions and they do not evaporate. However they do have one major drawback, and that is their price. Microspheres are sold as aqueous suspensions with a concentration around 10^{12} particles ml^{-1}. When aiming for a concentration of 10^9 particles ml^{-1}, then one ml of microsphere suspension will be sufficient for one litre of drill mud. Considering the price of tens of dollars per millilitre of microsphere solution and the usual volumes of drill mud in the range of thousands of litres the use of microspheres very quickly reaches financial limits. Only in very rare instances (Kallmeyer et al., 2006) microspheres were directly mixed into the drilling mud. In most operations the tracer is packed into small plastic bags and taped to the core catcher at the bottom of the core where it bursts open once the sediment enters the liner (fig 8). This way the core is bathed in a high concentration of microspheres without the need to add them to the entire volume of drill mud (e.g. Lever et al., 2006; Russell et al., 1992; Smith et al., 2000a). However, this method has a drawback, which is the uneven delivery of microspheres (House et al., 2003; Yanagawa et al., 2013). If the drill fluid does not form a homogenous suspension with the microspheres then chances are high that infiltration of drill fluid into the core will go unnoticed. Still, this technique has been used in many drilling operations with satisfactory results. Of course there were attempts to overcome these limitations. Juck et al. (2005) coated the inside of a liner with a microsphere suspension, so as soon as the core hits the liner it will be in contact with the tracer. They used this approach only in a rather small-scale operations and it remains to be seen whether this approach can be expanded to larger operations.

The ultimate tracer to detect infiltration of microbes into a core would be a microbe that does not occur in this environment naturally and is very easy to detect. Several studies made such attempts. In a laboratory study (Colwell et al., 1994) used the Cyanobacterium *Aphanocapsa delicatissima* to measure infiltration of water into basalt cores. Two detection methods were used to quantify infiltration of cyanobacteria in the core, cultivation and spectrophotometric chl-a measurements. Although the method proved successful for studying flow through the pore space of the basalt cores, its suitability for larger drilling operations remains questionable, as this would require larger volumes of cyanobacteria. Also, chl-a concentrations decreased by 20% over 30 days. Although the decrease is much slower as PFT, cyanobacteria might still not be the best choice when long-term stability for tracing contamination is an issue. Colwell et al. (1994) also added microspheres as an inorganic tracer and found that the cyanobacteria move faster than the microspheres, despite having a similar size.

Juck et a., (2005) used the strain *Pseudomonas Cam1-gfp2,* expressing a green fluorescent protein (GFP), as a contamination tracer for drilling in permafrost and ground ice. They painted the culture on the inside walls of the liner. Like Colwell et al. (1994) they used two detection methods, cultivation of the organism and much more sensitive PCR of the GFP-gene. Both approaches indicated good transfer of microbes from the painted walls of the liner into the core. Like in the approach discussed previously these techniques might work very well in small-scale operations but it remains questionable whether such approaches can be scaled up for deep drilling. Also, the use of genetically modified organisms in a drilling operation might also cause some major problems for permitting.

Very recently a new fluorescent particulate tracer was introduced to scientific drilling and that is an aqueous pigment solution that is normally used for paints and plastics (Friese et al., submitted). These pigment particles have the same size range as subsurface microbes (0.25 to 0.45 μm) and are available in several colours. Their biggest advantage is the price, as they are about three to four orders of magnitude cheaper than normal microspheres. At this price it is feasible to mix them into the drill mud or even use them in operations where the drill fluid is not recycled. However it should be noted that at concentrations around 10^9 particles mL^{-1} the tracer stains the drill fluid in a very bright colour, similar to dyes (fig 9). In their study, Friese et al. (subm.) also show the use of flow cytometry for quantification of microspheres, which decreases analysis time considerably. With a new generation of portable flow cytometers it is even possible to do such analyses right at the drill site. Still, fluorescence microscopy should be employed for calibration.

Dissolved chemical tracers

Different chemicals have been tested as contamination tracers: Salts like lithium bromide (Haldeman et al., 1995), potassium bromide (Phelps et al., 1989; Russell et al., 1992), barium (Chapelle and Lovley, 1990) or sodium fluoride (Hirtz et al., 1993) as well as sulfonic acids (Hirtz et al., 2001). For dissolved salt tracers the choice of a suitable compound largely depends on two factors, pore water chemistry of the formation because the natural background concentration of the ion that is used as tracer is one of the two main factors that determines the minimum detection limit, the other one is tracer concentration in the drill fluid. The former factor limits the use of dissolved salts as tracers mainly to environments with low ionic strength pore waters, i.e. freshwater lakes and terrestrial sites away from geothermal areas or brines. For a tracer study in a hypersaline brine with bromide concentrations in the 100s ppm range the initial plans for using bromide were abandoned and PFT was used instead (Hirtz et al., 2001). Also, even in environments with low pore water concentrations of the tracer the potential loss through precipitation should be considered, therefore a full pore water analysis should be available prior to drilling. For example calcium fluoride (CaF_2) has an extremely low solubility in water (15 mg L^{-1}) so fluoride might not be a suitable tracer in a lithology with high dissolved calcium concentrations.

The detection method depends on the ion of interest, ion chromatography is usually the method of choice for anions (Russell et al., 1992), whereas for cations atomic emission spectroscopy or similar techniques are being used (Chapelle and Lovley, 1990). Both techniques have a minimum detection limit between 0.01 and 1 ppm. Sulfonic acids can be detected by fluorescence or by gas or liquid chromatography. In combination with mass spectrometry the detection limit can be as low as sub-ng mL^{-1} (Serres-Piole et al., 2012). For salts the maximum tracer concentration in the drill mud is not limited by solubility issues like for PFT, moreover they do not stain the drill mud in a bright colour or cause any problems with disposal. Therefore concentrations can be set in the 100s ppm range to provide three or more orders of magnitude detection range. In closed systems where the drill mud is recycled dosing such high concentrations of salt is not much of a problem, a kilogram of salt per cubic meter of drill mud will not cause insurmountable logistical or financial challenges. In lake or ocean drilling operations however, where the drill mud is not recycled, the situation is different. As an example of a medium-sized drill rig, DOSECC's Deep Lake Drilling System uses around 20 m^3 of drilling fluid per day. This drilling system does not recirculate the drill fluid, but it exits the drill hole at the lake floor and is lost. For short operations this will not cause any problems but eventually the amount of salt that has to be purchased and brought to the drill site might become an issue.

Different techniques for extraction of tracer have been used, the choice mainly depends on the volume of available sample and its porosity. For soft and water-saturated samples centrifugation is the easiest option. Hydraulic pore-water squeezing is much more efficient but requires larger volumes of sediment, which might not be available in all cases. For very small sample volumes the only option might be slurrying the sample in deionized water, followed by centrifugation. Because this technique obviously involves dilution of the sample it causes a loss in sensitivity.

Other types of chemical tracers that are being used in hydrothermal research are various types of sulfonic acids (Hirtz et al., 1993; Hirtz et al., 2001; Serres-Piole et al., 2012). The main advantage of these compounds is their thermal stability up to several hundred °C. They are widely used in geothermal research (see (Serres-Piole et al., 2012) for an extensive review) but rarely in scientific drilling (Jackson et al., 2015).

Microbiological and molecular ecological techniques

In recent years microbiological and molecular biological techniques have gained increased attention. In most cases no tracer was deliberately added but the microbial community composition of the drill fluid, the drilling equipment and the recovered core samples were analysed and compared.

The first attempts were culture-based and used *E.coli* as tracer organisms, which were found in the drill mud as serendipitous contaminants (Beeman and Suflita, 1989). Detection was done via cultivation, which only allows for a qualitative assessment, basically providing the information whether viable *E.coli* was in the sample or not. Also, the absolute concentration of *E.coli* in the drill fluid was not determined, which also precludes the chance for a quantitative assessment. In a deep mine hosted in granite (Pedersen et al., 1997) employed a more quantitative cultivation approach by using viable counts on agar plates as well as Most Probable Number (MPN) counts of sulphate reducing bacteria to estimate microbial abundance in drill fluid and core samples. The numbers were in good agreement with total cell counts.

Using a specific strain of microbes, whether deliberately added or not as a tracer has its advantages, as they most closely resemble the indigenous microbial population. However, the results should be interpreted with caution. It remains questionable whether the distribution of *E.coli* cells provides a realistic estimate of the distribution of contaminant cells in a drill core because they are about an order of magnitude larger than subsurface microbes (Fagerbakke et al., 1996) and may therefore get trapped at pore throats where smaller indigenous cells can easily fit through. Other attempts were the addition of a phototrophic cyanobacterium followed by detection via cultivation and quantification of chl-a (Colwell et al., 1994), or a cultured GFP-expressing *Pseudomonas* strain and detection via a highly sensitive PCR (Juck et al., 2005).

The extreme sensitivity of PCR-based molecular techniques lead to their implementation as contamination tracers. The study of (Pedersen et al., 1997) was the first that used comparison of 16s-rRNA gene sequences from drill core, drill mud and from the surfaces of the tools to assess contamination. This technique was used and further refined in other studies (e.g. Davidson et al., 2011; Watanabe et al., 2000)). The enormous potential of this technique becomes clear in a recent study of 2.5 km-deep submarine coal deposits. Cell abundances were as low as <10 cells cm^{-3} and set a new record for both depth of life beneath the sea floor and low cell numbers (Inagaki et al., 2015). Although PFT was employed as a contamination tracer and all cores were checked for mechanical integrity by CT-scan, the ultra-low cell abundances make contamination control almost useless as even PFT is on the limit of sensitivity that would be required to render a sample uncontaminated if there are only 10 indigenous cells per cm^3. Because the authors were aware of this issue, they sequenced the V1 to V3 region of 16s-rRNA genes in core samples as well as the drill fluid. They then applied a probabilistic approach to estimate the likelihood that a given taxon would be consistently sampled from a group of samples, either exclusively from the sediment samples or from both drill mud and sediment sample. In this way, those taxa were identified that were either (i) exclusive to sediment samples ("most conservative") or (ii) consistently found in sediment samples in significant abundance and only occasionally found in contamination controls in low abundance. These two groups of taxa were labelled "most conservative" or "most likely", respectively. Using this approach, correction factors to the raw cell concentrations in the samples were calculated to estimate the corresponding population sizes. So for each sample there is a "raw" cell count, and a "most conservative" and "most likely" indigenous cell abundance, respectively. This massive amount of work might not be necessary for samples with higher cell abundances, but it shows very nicely the potential that molecular tools have. Given the ever-decreasing price for sequencing and the rapid advances in automatizing routine steps like DNA extraction such techniques might become routine in just a few years.

How to chose the right tracer?

Each tracer has its strengths and weaknesses and many factors have to be taken into account when deciding on a specific method.

Does a particular tracer work under the given circumstances? Not every tracer is suitable for every environment. Some tracers might be affected by the chemistry (pH, salinity) of the water used to prepare the drill. In cases where water with a chemical composition that deviates significantly from normal ocean or tap water is being used, tests prior to ordering any tracer or drill mud additives are highly recommended. As an example of what can happen when such tests were not carried out, a drilling operation was carried out in a soda lake, its waters have a pH around 10 and salinity around 20 ‰. A synthetic thickener and clay minerals had to be used to increase viscosity of the drill mud to stabilize the hole. According to the data sheets of the additives they should form a stable suspension up to pH 11. However, these tests were only made at low salinities and the suitability of the additives in a combination of high pH and high salinity was not tested. When trying to mix them with actual lake water they did not for a viscous suspension, but flocculated and settled at the bottom of the mud tank. Due to the remote location it was not possible to source other drill mud additives, so tens of thousand of litres of freshwater had to be brought from shore to the drilling platform on the lake, causing massive additional operational costs and delays.

Also, the thermal regime in the drill hole should be estimated. In deep holes or in hydrothermal settings the temperature might exceed the thermal stability of a tracer, most fluorescent tracers, either dyes or microspheres are not stable at temperatures above 100° C. In such cases PFT or sulfonic acids might be a better choice. The type of drill mud additives is an intensively discussed issue during the preparation of every drilling operation. When deciding on a specific tracer the additives have to be included in the consideration, as they can substantially alter the properties of the drill fluid and therefore of the tracer as well. For example, the addition of a polymer-based thickener at one of the drill sites of the Ketzin CO2-sequestration test site led to a decreased infiltration of drill fluid into the core, most probably by clogging the pores on the outside (Wandrey et al., 2010).

Is it technically/logistically/legally possible to use a particular tracer?

How do I get the tracer mixed into the drilling fluid? This might sound like a simple question, but PFT is highly volatile so they have to be added to the drill mud directly at the intake of the mud pump, which requires dedicated delivery system that varies the delivery rate according to the flow rate of the mud pump. As already discussed in the chapter about PFT, installation of a pump to deliver the PFT into the intake of the drill mud pump has to be discussed with the drilling team well in advance.

Another issue that becomes important is the even distribution of the tracer. In case a tracer is mixed into the drill mud tank, it is of utmost importance to ensure that the tracer becomes well homogenized (fig 10). Kallmeyer et al. (2006) provide an example of a time-course measurement of microsphere concentration in a mud pit. It took several 10s of hours before a tracer addition led to an increase in tracer concentration at the outflow of the well.

Another issue is the on-site measurement of a tracer. For fluorescent dyes only a fluorometer is required, which is available as small portable units that can easily be brought into the field. Still, in most cases the sediment has to be slurried to extract the dye and then centrifuged to remove the particles. Centrifugation is also required to measure the dye concentration in the drill fluid. This means that a centrifuge has to be brought into the field as well. Sometimes filtration will also work, but this depends on the grain size of the drill core and the composition of the drill fluid. Depending on the amount of liquid that is required for the measurement and the grain size of the material the size of the centrifuge can vary. A suitable workspace has to be allocated for this equipment. PFT analysis requires a GC-ECD, which might be difficult to operate on site because it requires a carrier gas. Also, samples have to be taken immediately after retrieval of the core, something that might be difficult on small lake drilling barges or other systems with very little working space.

Allocation of workspace becomes even more of an issue when working with fluorescent microspheres. Even when analysing them on site with a portable flow cytometer, a fluorescence microscope will be required for calibration and spot checks. Due to the relatively weak fluorescence signal a dark room for a microscope is required. If a dark room is not available then a large piece of dark cloth can be hung over the microscope and the head of the person using it, but this is not comfortable for many hours of work and should only be seen as a makeshift solution.

Legal constraints should also be taken into consideration. Depending on the location of the drill site, certain tracers might not be allowed. Also even if a tracer is absolutely non-toxic and does not cause any legal issues it might raise some attention with the local population if it stains the drill fluid in a bright colour. The impact of the local population on the success of a drilling campaign should never be underestimated and it should be a primary goal of anyone involved in the drilling campaign to maintain a good relationship with the people that live nearby the drill site. Pumping many cubic meters of a brightly coloured solution into the ground might not increase their sympathy towards the operation. Adverse reactions can range from revoking access rights over court orders to stop drilling to physical attacks. These are not theoretical considerations, all this did happen during various scientific drilling campaigns. If the choice of a less conspicuous tracer can help running a project more smoothly then it should be done that way.

Depending on the location of the drill site, disposal of the drill mud after the termination of the drilling operation might also be an issue. Again, even if the tracer is non-toxic and does not pose any harm or additional problems for its safe disposal, if it stains the drill mud in a bright colour it might cause some problems.

What is the lowest concentration of tracer that I can detect in a sample, and is that level sufficient for the material I want to retrieve? It is of no use to invest lots of resources into a contamination control method that actually works but to find out later that the minimum detection limit is not sufficient and small but in this case significant contamination passes unnoticed. Several factors have to be taken into account, concentration of cells in the drill fluid, lowest expected cell concentration in the sample, tracer concentration and minimum detection limit. Even if these calculations cannot be solved with great accuracy, they will at least provide an order-of-magnitude estimate that will indicate whether the planned approach is feasible or not. There should be a safety margin of at least one order of magnitude.

Concluding remarks

Drilling is a science in itself and no bio- or geoscientist should feel bad for not being familiar with all the technical details. For the success of every scientific drilling operation it is therefore of utmost importance to develop a drilling strategy in close collaboration with those people that will eventually run the drilling operation. Most commercial drilling companies have little to no experience working with scientist and vice versa. So it is absolutely necessary for the scientists to clearly formulate their expected goals and requirements and then discuss them with the drilling company. Many scientific drilling operations do not utilize their full potential because of insufficient planning or coordination between the science and the drilling team.

Even as someone with little or no practical experience in drilling, one should be at least familiar with the basic principles and techniques in order to be able to discuss the goals and technical strategies to achieve them. Perhaps the single most important issue that determines the success of a biogeochemical drilling campaign is to get involved as early as possible. Drilling with contamination control is different from drilling without, but most changes can easily be implemented at an early stage. Nothing is worse and more problematic than trying to implement contamination control in a project at the last minute. It is not just the technical changes; also the workflow needs to be adjusted to accommodate the additional sampling, plus the extra space for analyses and the potentially different legal situation.

A solid dataset from pilot experiments is also a prerequisite. Will the tracer work under the given conditions and will it provide sufficient sensitivity. As mentioned previously, there are many things that can go wrong, but most of them can be figured out well ahead of drilling. As a final remark, one should not underestimate the time that is necessary to organize a drilling campaign, irrespective of contamination control. The timeframe can easily be measured in years from the first draft of the project over the planning and permitting process to the actual drilling and later on the closing of the borehole and demobilization of the drill rig. Despite all the efforts that have to be put into drilling campaign, it is the only way to obtain samples from subsurface environments. Every drilling campaign provides new and exciting samples so all the hassle is well worth it.

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Figures

Fig 1. The Long Piston Corer of Woods Hole Oceanographical Institution. The thick-walled steel core barrels are held together by joints. The core shoe and the core catcher are removed to allow inserting a liner. The large weight set is in the back. (Foto: Kallmeyer, GFZ)

Fig 2a. A team of happy technicians pulls a 9 m-long liner out of the core barrel and brings it to the science area, the so-called catwalk, on board the IODP drill ship JOIDES Resolution. At the cat walk the core will be sectioned and sampled of time-sensitive parameters like PFT tracer. (Foto: Kallmeyer, GFZ)

Fig 2b. A team of even happier scientists cuts the core into 1.5-long sections and takes subsamples. Note the sets of syringes at the freshly cut ends. This way samples for PFT are taken. (Foto: Kallmeyer, GFZ)

Fig3: A selection of drill bits and core catchers used on the IODP Drill Ship JOIDES Resolution. (Foto: Anna Ling, University of Miami)

Fig4: A cutting shoe of a hydraulic piston corer (HPC). The front edge is bent and damaged from hitting harder material. (Foto: Kallmeyer, GFZ)

Fig 5: A wireline corer is about to be deployed on board JOIDES Resolution. The spearhead sits on top of the corer and the overshot assembly is hanging on chains at the left. A piece of drill string can be seen in the foreground on the right. (Foto: Kallmeyer, GFZ)

Fig 6: Drill fluid stained with fluorescein in a holding tank. (Foto: Alawi, GFZ)

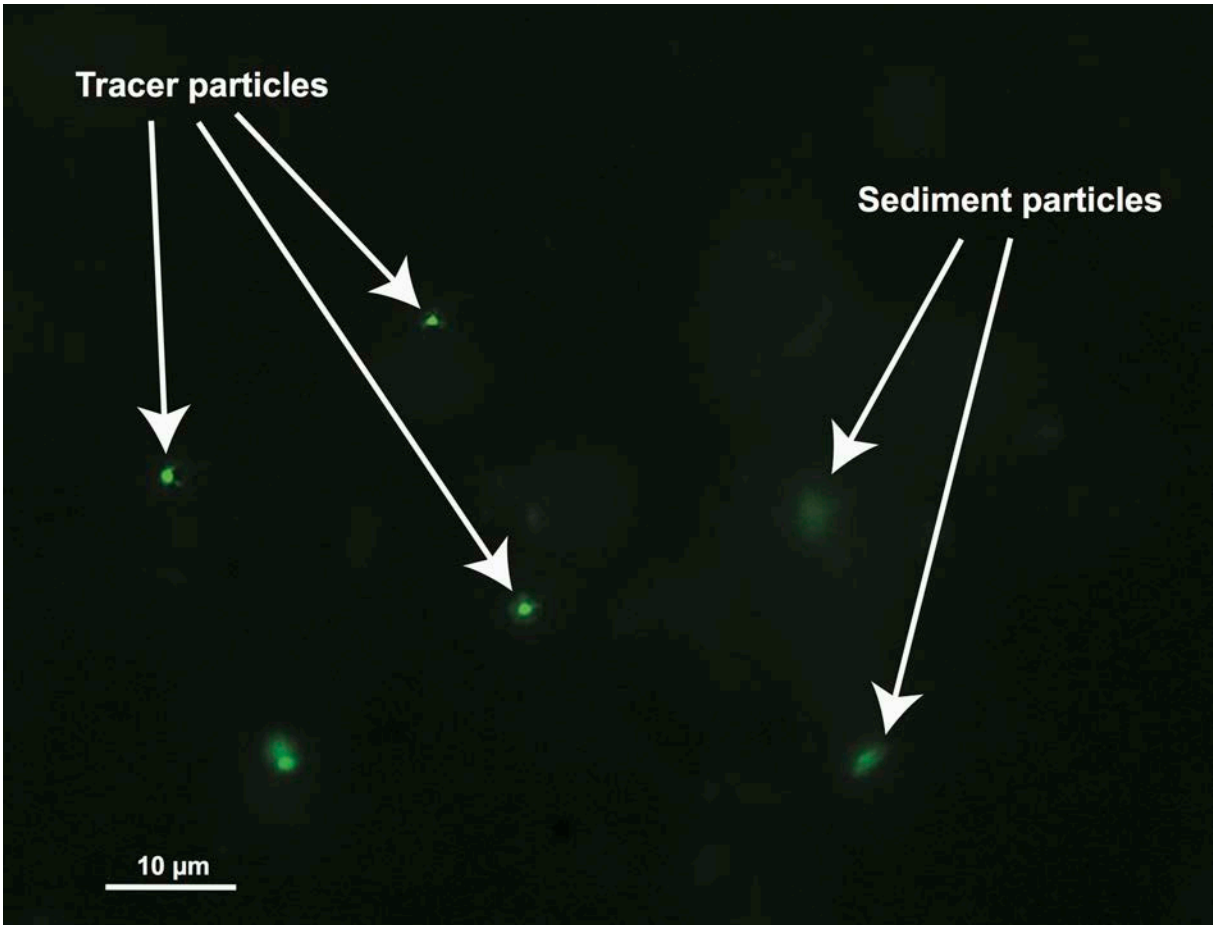

Fig 7: Microphotograph of fluorescent particles in a contaminated sediment sample. (Foto: Friese, GFZ)

Fig 8: A small plastic bag filled with microsphere tracer solution taped at the bottom of a piston corer. The bag will burst open when the corer hits the sediment and release the microspheres that will bathe the core in a highly concentrated tracer solution. (Foto: Kallmeyer, GFZ)

Fig 9: Microsphere tracer mixed into the drill fluid at a concentration of 10^9 particles ml^{-1}, which is already a 1000-fold dilution of the stock solution. (Foto: Kallmeyer, GFZ)

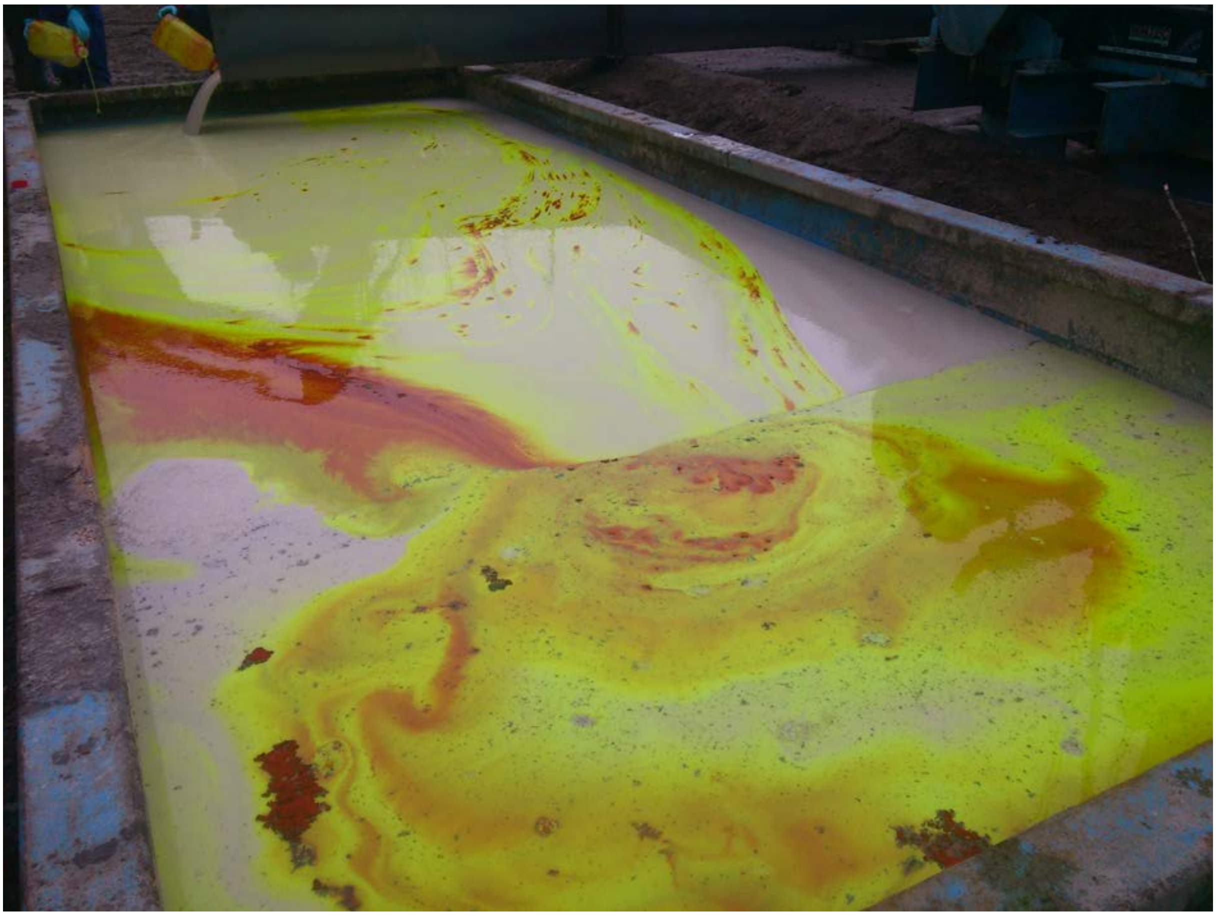

Fig 10: Mixing fresh fluorescein into a batch of drill fluid. The uneven distribution is apparent, more mixing will be required to ensure homogeneous distribution. (Foto: Alawi, GFZ)