Transport and retention of colloids are important issues when addressing the risk of contamination of the aquatic environment. A field study tracer experiment was performed allowing a quantification of solutes and colloids along macropores in a sandy loam soil with a tile drain located at 1.2 m depth. Using a field sprayer, a 6-m² plot was irrigated with 50 mm of water containing a mixture of 1-μm fluorescent microspheres (1.34 × 10¹⁰ melamine-resin microspheres [MS] L⁻¹), bromide (0.14 g Br L⁻¹), and the dye Brilliant Blue (2.2 g BB L⁻¹) during a 4-h period. Before irrigation, the groundwater table was more than 0.7 m below drain depth. The tracer concentrations were measured in drain water and in soil collected along 15 stained preferential flow paths located in the drain trench and the adjacent till. It was found that the tracer concentration along biopores do not necessarily reflect the concentration of tracers transported through the biopore. Furthermore, it was recognized that at drain depth the proportion of soil samples from the drain trench with concentrations of Br and MS greater than two times the detection limit was significantly higher than in samples from the till. This implies that substances can accumulate along the drain pipe during heavy precipitation events. Finally, water transported through a few biopores well connected to the drain pipe resulted in drainage. Compared with the concentrations in the added tracer solution, the sampled drain water showed undiluted concentrations of BB and Br, whereas MS was reduced 150 times.

**Colloid movement** is of considerable importance in arable systems as potential harmful bacteria and virus are applied to the field from manure (Gerba and Smith, 2005; DeNovio et al., 2004) and because colloids may facilitate transport of strongly sorbing pesticides (Gjettermann et al., 2009; McGechan and Lewis, 2002). Hence, when released, such colloids may be transported in macropores to deeper layers, contaminating the groundwater or surface waters.

The mobilization of colloids in soils is a very complex process, including desorption and dispersion promoted by lowering of ionic strength or changes in pH (Legedsmand et al., 1999; de Jonge et al., 1998; Kretzschmar and Sticher, 1998; Seta and Karathanasis, 1997). Furthermore, mechanical breakdown of the soil aggregates resulting from certain agricultural practices and raindrop erosion (Jarvis et al., 1999; Gao et al., 2004, 2005; Daraghmeh et al., 2009) may contribute to colloid mobilization.

When mobilized, colloids can be transported through the soil profile along preferential flow pathways (McDowell-Boyter et al., 1986; Gjettermann et al., 2009; Burkhart et al., 2008; Cey et al., 2009) such as root channels, earthworm burrows, and fractures (Beven and Germann, 1982; McGechan and Lewis, 2002; Jarvis, 2007). In numerous studies, dye tracers have been applied to natural soils to recover and describe these preferential transport pathways (e.g., Flury et al., 1994; Gjettermann et al., 1997; Petersen et al., 2001; Stamm et al., 2002; Kasteel et al., 2005; Rosenbom et al., 2008; Nielsen et al., 2010).

As for mobilization, attenuation of the colloids in the soil and along the preferential pathways is affected by both mechanical and physiochemical processes. For instance, colloids can be retained by straining. This happens when the colloids are larger than the pore diameter in which they are transported (McDowell-Boyter et al., 1986; McGechan and Lewis, 2002; DeNovio et al., 2004). Colloids smaller than the pores are influenced by physiochemical filtration mechanisms, including grain-straining, film-straining, and retention...
When evaluating transport and retention of colloids the most realistic outcome would be found by adding natural colloids as tracers. However, added natural colloids are very difficult to distinguish from colloids generated in the experimental soil. Thus, while model colloids like fluorescent microspheres (MS and polystyrene spheres [PS]) do not represent the variety in shapes, types, and surface properties of natural colloids, microspheres are a useful tool as they can be introduced in well-defined concentrations and are detectable from soil samples (Burkhardt et al., 2008). Hence, fluorescent microspheres have been used to evaluate effects of certain colloid properties in natural soils. For instance, regarding colloid size and transport depth, Cey et al. (2009) and Passmore et al. (2010) showed that it was possible to retrieve smaller PS (0.53 and 1.1 μm, respectively) from stained soil samples at greater depths than larger PS (3.75 μm PS and 1.6–16 μm, respectively) in unsaturated soils. The conclusion regarding surface charge effects are ambiguous. Harvey et al. (1989) reported that carboxylated PS were retarded more than neutral PS in a saturated sandy aquifer, whereas Passmore et al. (2010) showed carboxylated PS to be less attenuated than plain PS in an unsaturated Luvisol.

Despite the apparent usefulness of artificial microspheres as colloid tracers, and despite colloids being transported along preferential pathways only a few studies have examined the transport of colloids along preferential pathways using fluorescent microspheres along with dye tracers in the vadose zone under field conditions (Burkhardt et al., 2008; Cey et al., 2009; Passmore et al., 2010). Examination of MS in soil has been made possible by Burkhardt et al. (2008), with the establishment of a sensitive method quantifying fluorescent microspheres (MS) in soil. Furthermore, Burkhardt et al. (2008) found that dye was an essential guide when sampling for microspheres as almost no microspheres were found in unstained soil. This conclusion was strongly supported by the tension infiltration test made by Cey et al. (2009), who added that the dye tracer could act as an excellent surrogate for colloid distribution in the vadose zone. Building on these studies, Passmore et al. (2010) focused on the utility of microspheres as surrogates for the transport of the specific E. coli RS2 g. They concluded that the microspheres of similar size as the bacteria mimicked the transport of the bacteria quite well. In all three studies (Burkhardt et al., 2008; Cey et al., 2009; Passmore et al., 2010) the largest number of microspheres was measured in samples near the soil surface. Furthermore, the findings from these studies all supported that the number of microspheres did not decrease as rapidly with depth in macropores as in matrix flow domains.

Yet, compilation of further field evidence is necessary to unravel and understand the complicated colloid transport and retardation along preferential pathways in unsaturated natural soils. In the three studies (Burkhardt et al., 2008; Cey et al., 2009; Passmore et al., 2010) examination of flow patterns was limited by the irrigations areas (0.2–2 m²) and the depth of staining (0.4–1.7 m). Hence, none of these studies were set up to incorporate examinations and comparisons of differences of MS and Br distributions along macropores in soils between and in drain trenches. However, as parts of the groundwater risk assessment may be based on monitoring contaminants in the drain water (Kjær et al., 2009) and the link between macroporosity and tile drains is only beginning to be understood, further information on transport and retention of contaminants in soil with adjacent drain trenches is important.

The goal of this study was to increase the knowledge about the distribution of retained solutes and microspheres transported along individual macropores in a sandy loam till between drains and within a drain trench. Hence, we wanted to examine 1) whether there are differences in distribution patterns between the three added tracers (Brilliant Blue, bromide, and MS) and 2) whether there are different retention patterns in the soil between drains and within a backfilled drain trench. Finally, we wanted to examine 3) the amount of drain water and tracer concentrations in the drain water generated in conjunction with the irrigation (if any). The findings of this study are intended to help and improve model conceptualization regarding transport in and between drain trenches and thereby achieving a better representation of peak concentrations reaching surface water bodies, in turn affecting the ecosystem functions.

**Materials and Methods**

**Study Site**

The 6-m² study site (Fig. 1) was situated at a field in Taastrup near Copenhagen, Denmark (55°40’47”N, 12°17’37”E) having udic and mesic soil moisture and temperature regimes, respectively (Soil Survey Staff, 2010). The dominating soil type at the field is classified as a Luvic Phaeozem according to World Reference Base for Soil Resources (IUSS Working Group WRB, 2006). The soil has developed in a glacial till of Weichselian (Wisconsinan/ Würmian) age. Data on the pedological horizons presented in the till along with texture and organic matter properties are given in Table 1. Furthermore, a sketch of the horizons along...
with a picture of the soil can be seen in Fig. 2A. Since 1965 the field has been tile drained at a depth of about 1.2 m, ensuring that the groundwater table was generally kept at or below this level. Drain flow occurs in the period from October to March when the groundwater level occasionally rises above the drain level (Petersen et al., 2004). When establishing the drain system in 1965, 0.5-m-wide and 1.2-m-deep drain trenches were excavated and tile pipes were placed at the bottom of the trenches. The trenches were backfilled with lumps of A-, E- and B-horizon material. The drain pipes were placed with a horizontal distance of 16 m. Each tile drain pipe consists of 33 cm long waterproof sections. Hence, water can only enter the drain via joints between these sections.

The soil outside the drain trench (referred to as “the till”) was generally free of calcium carbonate down to a depth of 1.4 m. In this calcareous free till numerous single channeled biopores (root channels and earthworms burrows) and desiccation fractures (aperatures in micrometer scale) were observed. The number of biopores and desiccation fractures is in the same range in the drain trench as in the calcareous free till (Nielsen et al., 2010). Furthermore, Nielsen et al. (2010) reported that small voids between the lumps of backfill material were present in the drain trench. Below 1.4 m depth in the calcareous till few biopores were observed and almost exclusively found in conjunction with desiccation fractures and glaciotectonic fractures caused by subglacial deformation (Nielsen et al., 2010).

Table 1. Soil textures in four horizons from the examined till. The texture in the drain trench was a mix of these textures as lumps of A, E, and B horizon material was used to backfill the trench after the drain was placed.

<table>
<thead>
<tr>
<th>Horizon</th>
<th>Avg. depths</th>
<th>Texture</th>
<th>Clay (&lt;2 μm)</th>
<th>Silt (2–20 μm)</th>
<th>Sand (20–2000 μm)</th>
<th>Organic matter kg kg⁻¹</th>
<th>Clays kg kg⁻¹</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0–30</td>
<td></td>
<td>0.09</td>
<td>0.29</td>
<td>0.59</td>
<td>0.03</td>
<td></td>
</tr>
<tr>
<td>E</td>
<td>30–55</td>
<td></td>
<td>0.13</td>
<td>0.30</td>
<td>0.56</td>
<td>0.01</td>
<td></td>
</tr>
<tr>
<td>B/C</td>
<td>55–140</td>
<td></td>
<td>0.19</td>
<td>0.21</td>
<td>0.59</td>
<td>0.01</td>
<td></td>
</tr>
<tr>
<td>CCaCO₃</td>
<td>&gt;140</td>
<td></td>
<td>0.18</td>
<td>0.30</td>
<td>0.51</td>
<td>0.01</td>
<td></td>
</tr>
</tbody>
</table>

**Preparation and Soil Conditions before Tracer Application**

Before the experiment, the field was used for conventionally grown wheat (*Triticum aestivum* L.); it was plowed and sowed in...
The experimental plot was covered with a tarpaulin immediately after stubble cultivation to minimize evaporation. One week before tracer application, the site was watered with 20 mm tap water using a field sprayer to moisten the soil to field capacity. Just before tracer application (3 June 2008) the volumetric soil water content was measured with a neutron probe (503DR Hydroprobe Moisture Gauge, InstroTek Inc., CA) in two tubes located within the experimental plot. The average soil water content was 0.32 and 0.35 m$^3$ m$^{-3}$ at 15 and 105 cm depth, respectively. On the day when the tracer application took place, the groundwater table was located more than 70 cm below drain depth, that is, deeper than 1.9 m below surface.

**Tracer Application and Sampling**

The tracers used in this experiment are the anionic dye Brilliant Blue (BB C.I. 42090), a reactive bromide tracer (KBr, henceforth referred to as Br) and fluorescent melamine-resin microspheres (MS) (Microparticles GmbH, Germany). Brilliant Blue is a reactive tracer that undergoes nonlinear sorption when the concentration is below 10 g L$^{-1}$ (Ketelsen and Meyer-Windel, 1999; Germán-Heins and Flury, 2000; Kasteel et al., 2002) resulting in some retardation by Cey et al. (2009), compounds undergoing low to moderated sorption can be represented by the dye BB.

The MS used are spheres with a diameter of 0.98 ± 0.06 μm and a density of 1.51 g cm$^{-3}$ colored with the dye Sulforhodamine B monosodium salt, having a negative surface charge of 265 μmol g$^{-1}$ due to a carboxylated surface. These MS were chosen because their density is more like natural mineral colloids (Burkhardt et al., 2008) than the 1.05 g cm$^{-3}$ PS used by, e.g., Cey et al. (2009) and Passmore et al. (2010). In this context it can be noted that Harvey et al. (1989) supposed that the PS mimicked abiotic colloids better than the bacteria tracer. However, the size and the negative surface charge correspond to the ones found reflecting E. coli RS2 g best (Passmore et al., 2010). Finally, Burkhardt et al. (2008) used the same type of MS and succeeded in quantifying them in soil samples.

Using a moving field sprayer, 50 mm of tap water with 2.2 g BB L$^{-1}$, 0.14 g Br L$^{-1}$, and $1.34 \times 10^{10}$ MS L$^{-1}$ was added during a 4-h period to an effective study area of 6 m$^2$ centered within a 14-m$^2$ irrigated area intersecting the drain pipe at a 90° angle (Fig. 1).

Water application alternated between irrigation and short pauses as irrigation was halted whenever there was a risk of overland flow away from the treated area. The length of pauses was determined by the time needed for the ponded water to infiltrate. Following irrigation, the area was covered with a tarpaulin.

One week after irrigation, the plot was excavated to a depth of 1.25 m in the drain trench and to 1.8 m outside the trench. Afterward, the first of three vertical profiles in the till was cleaned. The surface of the vertical profile was chipped with a knife to expose the stained flow paths. The stained pathways were mainly vertical macropores such as earthworm burrows. The dyed wall material was sampled along each stained macropore. The vertical extent of each sample was 10 to 15 cm, whereas the horizontal sampling extent was restricted to the dyed soil within one millimeter of the macropore wall. The sampling strategy differed in parts of the E horizon as the dye did not always follow distinct pores here. In such cases, the samples were obtained at the center of the dyed soil material. After soil sampling along the first profile in which soil along two macropores was sampled (Fig. 1), the A horizon was removed, exposing the stained pattern below. Based on this pattern two additional vertical profiles were excavated. The profiles were cleaned as the first profile and soil along a further 10 stained macropores was sampled (Fig. 1).

The drain trench was explored by gradually chipping off small portions of soil using a knife. The dimensions of the drain trench examinations were: 1.2 m long, 0.5 m wide, and 1.2 m deep. Soil sampling followed the same procedure in the drain trench as in the till.

Furthermore, 100-cm$^3$ cylindrical ring samples (height: 3.6 cm; diam.: 6 cm) were collected at the surface and sliced. On average, the upper and the lower slices were 3 and 14 mm thick, respectively.

To reveal the background concentrations of the tracers, a number of 14 unstained samples were collected from the till matrix in six different depths (0.3–1.7 m) at the site. The samples were stored at 5°C until they were air dried at room temperature, crushed in a mortar, and stored in glass containers.

A container was placed under a drain outlet 10 m downstream from the irrigated area to collect all drain water. Hence, tracer concentrations measurable in the drainage water represented an average concentration for the entire event. The drain water sample was stored at 5°C until analysis.

**Tracer Quantification Procedures**

The quantification procedure for MS in soil samples followed that of Burkhardt et al. (2008). In short, a sample of 100 mg of dried soil was suspended in 100 mL of deionized water. The suspension was treated in an ultrasonic bath for 15 min to disperse the MS from the soil particles. An aliquot of 10 mL of the suspension was then filtered using a black polycarbonate membrane filter (pore
size: 0.22 μm, diam.: 47 mm, nonfluorescent; GE Water & Process Technologies, MN) in a glass suction filtration system (Toyo Roshi Kaisha Ltd., Japan). A 1-mL subsample of the drainage water was filtered as well. The effective diameter of the filter was 34 mm, yielding an effective surface area of 907 mm².

The number of MS on the membrane filters was then examined by fluorescent microscopy (Zeiss III Rs, Germany). The ultra-high-pressure Hg-lamp used had an excitation wavelength of 436 nm and a suppression-filter (LP 520) which excited the fluorescent dye in the MS (λ<sub>emission</sub> = 585 nm). The magnification was 125 times altogether (10× by the ocular, 10× by the objective, and 1.25× by a tubusfactor in the Zeiss III Rs microscope). At each cover slip a drop of Zeiss Immersol 518 Z was added to increase the contrast. Twenty pictures were taken in a grid pattern for each filter (using a Nikon Coolpix 3.34 mega pixel, no-blitz, closure time 0.5 s and shutter f4). The microspheres were counted manually on photos with less than 5 MS. The pictures with more than 5 MS were examined using the image analysis program ImageJ (Rasband, 2008). When using ImageJ, the yellow color was adjusted to 100 (range 0–255), as this was found to minimize color interference from other sources. The picture was then transformed to an 8-bit picture, with a threshold of 15 in the gray scale ranging from 0 to 255. Because there were varied size halos around the MS, the number of circular particles ranging from 5 to 50 pixels was counted. The length of 5 pixels corresponded to 1.8 μm. The total area of the 20 recorded images was 7.8 mm² corresponding to 0.9% of the effective filter area. In a preliminary test, the MS recovery rate was 37% ± 3% (avg. ± SE) for a range of 10<sup>6</sup> to 10<sup>9</sup> MS g<sup>−1</sup> soil. Based on this test, a correction factor was used to correct the measured number of MS in the sampled soil. Compared to Burkhardt et al. (2008), who recovered 87 and 105% in their test with soil, the recovery rate in this study is low. The difference in recovery rates may be due to differences in soil texture (silt loam versus sandy loam) and a slightly modified recovering protocol. In the modified protocol the effective filter area and the area of the image analyzed has been changed and another image processing program was used.

When quantifying Br and BB, the soil was shaken in deionized water (1:1 w/w) for 16 h and then centrifuged for 30 min at 1341.5 g. Subsamples of the supernatant and subsamples of the untreated drain water sample were analyzed for Br using a Dionex Ion Chromatography (CX 500 HPLC, CA) while other subsamples were analyzed for BB concentrations, measured by spectrophotometry at 630 nm (Shimadzu UV 106, MD). In some cases a limited sample size precluded the measurement of BB.

### Results and Discussion

When looking at the excavated till profile, it is notable that staining almost exclusively occurred along a few vertical biopores (Fig. 2A). The horizontal extent of staining along these biopores was only up to a few millimeters thick, whereas larger horizontal staining (up to 30 cm) occurred in the E horizon and at walls of desiccation fractures. Horizontal staining along the drain trench was associated with the till–drain trench interface (1 m below surface, Fig. 2B). Furthermore, dye was distributed continuously along the bottom of the drain trench at the interface between the drain pipe and the soil (Fig. 2C and 2D). Moreover, the visual interpretation of the excavated areas indicated that the largest extent of staining by BB was found around the drain pipe. Contrarily to findings in Nielsen et al. (2010) no staining was observed at desiccation fractures or between lumps of backfill material in the drain trench. Finally, more than 2.0 L of water was transported via the drain to the container at the drain outlet (Fig. 2E).

### Tracers in the Drain Water Sample

The 2.0 L of drain water sampled corresponds 2 to ~1% of the water added at the net irrigated surface area (Fig. 1), and to ~8% of the water added at the net drain trench surface area (0.5 × 1 m). It was noted that some blue water had escaped from joints between drain pipes downgrade, and the drain pipe was not isolated from the soil. Hence, even more than 2.0 L of water had probably entered the drain. When comparing the tracer concentrations in the cumulative sampled drainage water with the applied concentrations, it turned out that the concentration of the conservative tracer Br (0.12 g L<sup>−1</sup>) and the weakly to moderately retarded dye BB (2.1 g L<sup>−1</sup>) in the drainage water were almost equal to the applied concentrations, whereas the number of MS (8.7 × 10<sup>7</sup> MS L<sup>−1</sup>) was found to be 150 times lower than in the added tracer solution. The quantity of BB, Br, and MS in the drain water container corresponds to ~1% of BB, ~1% of Br, and ~0.04‰ of MS of the tracer solution added to the net irrigated surface area and to ~8% of BB, ~8% of Br, and ~0.5‰ of MS in the tracer solution added to the net drain trench surface area.

This implies that solutes and weakly to moderately retarded species may potentially reach the drain pipe undiluted during a heavy precipitation event. Although some filtration does occur for colloids, transporting strongly sorbing materials or harmful bacteria, colloids may also move from the surface to drainage water in high concentrations when a continuum of macropores between soil surface and drain are present. A previous study at the same field has observed that earthworms can be flushed through the drain pipes (Petersen et al., 2004), indicating that biopores with even better connection between drain and surface than those observed in this study can be present along the drain line. Hence, it is expectable that the concentration of colloids such as MS would be less diluted when transported through such direct connected pores than through the ones in this study. The observations from this study imply that a high concentration of substances may reach surface water bodies in peak events, influencing the ecosystem functions.
Tracers in Samples from the Till

No tracers (BB, Br, or MS) were detectable in samples from the unstained till. This corresponds with the findings of Cey et al. (2009) and Burkhardt et al. (2008). Conversely, all three tracers were measurable in the sliced surface ring samples with average concentration in the upper slice of MS: $2.2 \times 10^7 \mu g g^{-1}$, Br: $40 \mu g g^{-1}$, and BB: $190 \mu g g^{-1}$. Furthermore, from these samples it was recognized that MS counts decreased more strongly from the upper (0–3 mm) to the lower slice (22–36 mm) (15 and 55 times in the two rings, respectively) than the concentration of Br (3–4 times) and BB (4–20 times) which probably is due to straining and filtration at the uppermost mm just at and below the surface.

Along the 12 examined stained macropores in the till, the distribution patterns varied both among the pores and with depth. In Fig. 3, four representative depth distribution patterns from the site are illustrated (A–D). The distribution pattern A (representing 3 macropores) clearly illustrates stained pores with low contents of MS and Br along the macropore walls. In parts of B (representing 4 macropores) the retention of MS is greater than in A, whereas the concentration of Br is still very low. In C (representing 3 macropores) all three tracers are found in considerable amounts. The distribution pattern in D (representing 2 macropores) is a mix of B and C and differs mainly from these types by including additional samples from the A horizon and by having multiple local maxima in tracer concentrations. When looking at the macropores from which the distribution patterns are derived it was observed that Type A, B, and D all were stained biopores which distinctly traversed the E horizon. In Type C it was difficult to see whether the horizontal staining was associated with one distinct biopore or with a terminating biopore having an internal catchment in the E horizon. From such internal catchments new biopores may have been activated.

The highest concentration of MS along the four presented macropores was measured in the soil sample collected along the wall of biopore D, just below the soil surface. However, it has to be noted that the concentration was almost the same as at 0.35 m depth in the E horizon (B). Furthermore, the concentration was less than what has been found in other areas with root networks and in one sample collected along a distinct biopore. Since the A horizon was removed after sampling along two biopores, it was impossible to sample the upper 30 cm of stained biopore wall material in the remaining macropores examined in the till. Hence, it cannot be determined whether there was a higher concentration of MS in the upper 30 cm than further downward in individual macropores. Due to straining and filtration at the surface, such a trend is expected and would be in accordance with the findings in Burkhardt et al. (2008), Cey et al. (2009), and Passmore et al. (2010).

The staining patterns and the distribution of the chemical constituents in individual biopores appear to be related. When staining was restricted to the walls of a biopore, the MS and Br contents were mostly low. This finding can be interpreted as the walls in such cases were of low permeability. Such low permeability would reduce the exchange of tracer solution between the macropore and the matrix, although still allowing sorption of BB to the wall material. In contrast, the highest concentration of MS was generally found in areas where staining extended horizontally, as in the E horizon. This implies that the biopore wall was more permeable here, allowing water to migrate from the biopore into the matrix. Burkhardt et al. (2008) previously reported that tracer hot spots can occur along macropores.

The examination of the individual biopores in the till suggests that the three tracers often followed similar distribution patterns within an individual pore. This likely reflects that the distribution along the individual macropores is affected by the pore chemistry, permeability, and tortuosity. The concentration of MS often mimicked the variation in the concentration of BB. However, BB could be present without a corresponding measurable concentration of

![Fig. 3. A selection of four typical distribution patterns in stained macropores from the examined till (out of a total of 12 sampled macropores). Type A shows a single channel biopore where sampling starts in the E horizon and continues in a distinct biopore. In Type B sampling starts in the E horizon as well, but continues along a slightly more meandering biopore compared to Type A. Sampling in Type C starts in a biopore just above the E horizon (0.5–0.7 m) and continues into a distinct biopore below. The staining terminates in a desiccation fracture below 1.3 m. Type D represents a macropore with multiple local maxima in tracers associated to different parts of the E horizon (0.35–0.45 and 0.60–0.65 m).](www.VadoseZoneJournal.org | 350)
Tracers in Samples from the Drain Trench

In the drain trench three stained biopore flow paths (two individual biopores and one cluster of less than 10 biopores within a cylinder with a diameter of 10 cm) were found at depths greater than 0.35 m. In the two individual biopores, staining terminated above a depth of 0.75 m, which was more than 0.25 m above the drain pipe. One biopore had a distribution pattern similar to the one in Fig. 3A, while the other was more like the one in Fig. 3D, with a local tracer concentration maximum at a depth of 0.6 m in a stained root channel network.

A cluster of biopores was situated at the boundary between the drain trench and the adjacent till at the outer wall of the excavation (Fig. 2B). They fed the drain pipe through a continuum of macropores even though an identification of the exact pore feeding was impossible. However, the examination of the drain trench revealed that these stained biopores were the only pores contributing to staining at the till–drain trench interface (Fig. 2B) and along the drain pipe (Fig. 2C and 2D). The distribution patterns in Fig. 4 are based on samples from two biopores in the cluster. In the drain trench, the MS concentration exceeded the detection limit in one biopore sample only; this sample was collected at drain depth. The concentration of Br was low and decreased from the top of the soil and downward, whereas the concentration of BB was almost constant, except for one biopore sample at a depth of 0.75 m. However, at the depth of the drain pipe the tracer concentrations increased. Depending on the tracer type, the tracer concentration measured in samples from the depth interval of 1 to 1.25 m were at least 1.5 to 10 times larger than the concentrations measured in the biopores feeding the drain trench. This indicated that the concentrations of tracers in the soil samples from the walls of biopore were weakly related to the amount of substances transported through them. The samples with the increased level of tracers were samples from the till–drain trench interface flow path (Fig. 2B) and around the drain pipe (Fig. 2C and D).

Drainage was interpreted as being a result of local water saturation. Areas with local saturation could be related to the macropores at the till–drain interface and the macropores around the drain pipe fed by the water-conducting cluster of biopores. As the drain pipe consists of 33 cm long waterproof tile drain sections, water can only enter or leach from the drain at the joints. Water leaching from the joints may have added further tracers to the amount which was presumably filtered and adsorbed before the pressure threshold was reached. Furthermore, tracers could still be retained along the drain pipe when the hydraulic pressure fell below the threshold for water entering into the drain. Additionally, as mentioned above, some tracers were lost through joints further downgradient, not reaching the container at the outlet.

Comparison of Microspheres and Bromide Distribution in Samples from Drain Trench and Till

Based on the total dataset for Br and MS (BB was excluded due to sparse data) the general tracer concentration decreased along macropores from the top of the soil down to depths about 1 m (Fig. 5). These observations are in agreement with Cey et al. (2009). The decrease in concentrations is more distinct for Br than for MS. At depths below 1 m, the magnitude of concentrations is affected by the sampling site (macropores in the till versus macropores in the drain trench).

Furthermore, from Fig. 5 it is indicated that the concentrations of tracers in samples from BB-stained material in the drain trench and the till differ below the uppermost 25 cm. For comparison, the samples were divided into four groups: (i) samples from the till with tracer concentrations exceeding two times the detection limit (2×DL: 2 μg Br g⁻¹ soil, 1 × 10⁶ MS g⁻¹ soil), (ii) samples from the till with tracer concentrations below 2×DL, (iii) samples from the drain trench with tracer concentrations above 2×DL, and (iv) samples from the drain trench with tracer concentrations below 2×DL. The threshold of 2×DL was used because the number of measurements above and below this limit was almost equal. When comparing the percentages of samples where the Br and MS contents exceeded 2×DL at different depths, it turned out that the tracer retention differed among stained samples from the trench.
and the till (Table 2). At the depth interval of 0.25 to 1 m, more stained samples from the till fulfilled the criterion (>2×DL) for MS compared to samples from the drain trench. The percentage of the samples with higher concentrations of Br than 2×DL is only larger in the till at the depth interval of 0.25 to 0.5 m. Below the 0.5-m depth, the percentage of samples in the drain trench with more than 2×DL is larger than the corresponding percentage in the till. A Fisher’s exact test, used for comparing proportions for small sample sizes that are not normally distributed (Agresti and Finlay, 2009), was conducted to test the hypothesis that the proportion of samples with higher concentration of tracers than 2×DL sampled in the depth interval of 0.25 to 0.50 m was unaffected by the sampling location (till or drain trench). The hypothesis was rejected for both tracers (p = 0.01 for Br and p = 0.007 for MS). However, drawing definite conclusions from these results and the corresponding p values is difficult since the number of samples in the drain trench is relatively low (5). If only one sample had contained more MS than 2×DL, no statistical significance would have been found. Bearing this observation in mind, there might be a tendency toward a higher content of Br and MS in the stained macropore material sampled at the depth interval of 0.25 to 0.5 m in the till compared to the content in the drain trench macropores.

A similar test on samples from the depth interval 1–1.25 m resulted in rejection as well (p = 0.005 for Br and 0.01 for MS), strongly supporting that the sample proportion having more than 2×DL was higher for samples collected in the drain trench than for those collected in the till. This, along with the visual interpretation indicating that the largest extent of staining by BB was found around the drain pipe, suggests that substances may accumulate along the drain pipe during heavy precipitation events. This interpretation is supported by the stained pattern evaluated in Nielsen et al. (2010). The influence of such accumulation along with the linkage between macropores and drain pipes should be addressed thoroughly when interpreting the occurrence of contaminants in drain water samples.

In total, 70% of the samples in dyed structures in the drain trench contained concentrations of Br above 2×DL, while the corresponding figure in the till was only about 50% (Table 2). In total, the MS appeared to be more evenly distributed, as 43 and 38% of the samples from the till and the drain trench, respectively, contained more than 2×DL. However, although the proportion of samples with MS is roughly equal in the till and the drain trench, the retained amounts appear to be a poor indication of the transport through the pores. A significantly larger proportion of samples with a high amount of tracers was found at depth in the drain trench than at similar depth in the till. Because the texture in the drain trench corresponds with the till texture as it is backfilled, and the number of biopores are in the same range in and between drain trenches at the field (Nielsen et al., 2010), the difference in tracer concentrations is regarded as an effect from the presence and location of the drain pipe. Hence, the risk of contamination of drain water during intense precipitation events with groundwater initially below drain depth mainly stems from pores leading to the drain trench and from the possibility of remobilization of substances in the drain trench. These findings should be regarded as relevant when monitoring of contaminants in drain water is used as part of the groundwater risk assessment.

### Conclusions

This tracer study has revealed that the retention and thereby the distribution patterns of Brilliant Blue (BB), bromide (Br), and fluorescent microspheres (MS) differ between and along macropores. An interpretation of the distribution patterns along macropores indicated that the interaction and mobility of water between the macropore and matrix was important. Moreover, it was recognized that the concentration of tracer along the biopore walls examined in the drain trench did not necessarily reflect the concentration of substances transported through the biopores.

Distinct differences in transport and retention between the till and the drain trench was observed when comparing the bromide and microspheres distributions quantitatively. It was notable that the proportion of samples with measurable concentrations of MS and Br in the drain trench was significantly higher than the proportion of samples from the till at the depth of the drain pipe (1–1.25 m). This implies that substances can be accumulated along the drain pipe.

### Table 2. Total number of dyed samples from the till and the drain trench at depths down to 1.25 m. The number of samples as well as the percentages of all samples with concentrations of Br and melamine-resin microspheres (MS) exceeding twice the detection limit (2×DL) is given for samples from the till and from the drain trench.

<table>
<thead>
<tr>
<th>Depth</th>
<th>All samples</th>
<th>Br &gt; 2×DL</th>
<th>MS &gt; 2×DL</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Till</td>
<td>Drain</td>
<td>Till</td>
</tr>
<tr>
<td></td>
<td>n</td>
<td>n</td>
<td>n</td>
</tr>
<tr>
<td>m</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0–0.25</td>
<td>10</td>
<td>5</td>
<td>8</td>
</tr>
<tr>
<td>0.25–0.50</td>
<td>18</td>
<td>5</td>
<td>14</td>
</tr>
<tr>
<td>0.50–0.75</td>
<td>24</td>
<td>4</td>
<td>11</td>
</tr>
<tr>
<td>0.75–1.00</td>
<td>16</td>
<td>3</td>
<td>2</td>
</tr>
<tr>
<td>1.00–1.25</td>
<td>9</td>
<td>20</td>
<td>1</td>
</tr>
<tr>
<td>Total</td>
<td>77</td>
<td>37</td>
<td>36</td>
</tr>
</tbody>
</table>

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during heavy precipitation events. Hence, these findings suggest that it would be valuable to assess the remobilization potential of retained substances along the drain pipes in future studies.

In relation to the samples from the drain trench, it was found that solutes such as Br and weakly to moderately retarded substances represented by the dye BB can enter the drain pipe and thereby surface water in an undisturbed state, while colloids entering the drain pipe, as represented by MS, may be filtered.

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