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Tracing the spatial propagation of river inlet water into an agricultural polder area using anthropogenic gadolinium

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Abstract

The inlet of diverted river water into agricultural areas or nature reserves is a frequently applied management strategy to prevent fresh water shortage. However, the inlet water might have negative consequences for water quality in the receiving water bodies. This

study aimed to obtain a spatial image of the inlet water propagation into a hydrological complex polder area. We used anthropogenic gadolinium (Gd-anomaly) as a tracer for diverted river water. A clear reduction in the river water contribution was found from very dry conditions on 5 August 2010 to very wet conditions on 22 October. Despite the large inlet water impact on 5 August, the diverted river water did not propagate
 up into the small agricultural headwater ditches. Gadolinium proved to be an effective tracer for diverted river water in a polder system. We applied our results to upgrade the interpretation of water quality monitoring data and to validate our integrated nutrient transport models.

1 Introduction

¹⁵ Local governments invest heavily in the identification and mitigation of water quality problems within their districts. However, in many agricultural areas and nature reserves worldwide, the inlet of diverted river water from upstream areas also affects the local water composition. Agricultural areas often depend on river water inlet for irrigation and groundwater recharge (e.g. FAO, 2003; Siebert et al., 2007). In wetland nature reserves, the introduction of diverted river water is an increasingly applied management tool to compensate for water shortage in summer or to counteract salinization (e.g. Roelofs, 1991; Runhaar et al., 1996; Delaune et al, 2005). In the Netherlands, for example, water inlet from the Meuse and Rhine rivers influences more than 60 % of the surface water system in dry periods (Bloemendaal and Roelofs, 1988; op cit. Roelofs, 1991).



In both agricultural areas and nature reserves, the inlet of river water might have major negative consequences for water quality. In many agricultural areas in arid, semiarid, and Mediterranean climates, good quality water resources are becoming scarce and are primarily allocated to industrial and drinking water purposes. This causes an

- ⁵ increase in the application of poor quality surface water in agricultural watersheds. In some arid areas, the river discharge that is available for agricultural irrigation in dry periods largely consists of untreated urban sewage effluent (Ryan et al., 2006; Srinivasan and Reddy, 2009). This obviously brings risks for human health through infection by pathogens and through accumulation of heavy metals in the food chain. For exam-
- ple, Srinivasan and Reddy (2009) reported higher morbidity rates in communities that use wastewater for irrigation in India. In more temperate climates, pollutant concentrations in river discharge may also increase in dry periods in summer due to less dilution of industrial spills and (partially treated or untreated) sewage effluent. Van Vliet and Zwolsman (2008) reported a general deterioration of the water quality of the Meuse
 river (France, Belgium, The Netherlands) during droughts with respect to many ele
 - ments including nutrients and heavy metals.

Diverted river water often causes direct or indirect eutrophication in the receiving surface waters. For example, Roelofs (1991) reported the disappearance of *Stratiotes aloides* L. (Water soldier) from a mire complex in The Netherlands, which resulted from

- internal eutrophication caused by the inlet of alkaline (HCO₃ and SO₄-rich) water. In the coastal wetlands of Louisiana (US), water inlet from the Mississippi is used to counteract the increasing salt-water intrusion due to ongoing land subsidence (Delaune et al., 2005). Shenyu Miao et al. (2011) reported the possible negative consequences of introducing Nitrogen-rich Mississippi water into an N-limited wetland ecosystem. Run-
- haar et al. (1996) gave several other examples of negative ecological consequences of water inlet into nature reserves.

The principles of detecting and mitigating water quality problems have been established in statutory legislations, such as the European Water Framework Directive (WFD) (EU, 2000) and the US Clean Water Restoration Act (CWRA) (EPA, 2009).



These policies stimulate an integrated approach of the complete hydrological system in river basins. This includes the link between upstream pollution sources and their effects on water quality in downstream agricultural areas and nature reserves that depend on water inlet and irrigation. The WFD for example states that a water body is "at risk"

- when it causes downstream dependent water bodies to fail to meet their good status objectives. In addition, the WFD water quality standards for nutrients and other biological parameters are based on average concentrations in summer (April–September). Water shortage and the application of inlet water are normally concentrated in these summer months. Consequently, the use of summer concentrations for testing the compliance with maximum allowable concentrations leads to a relatively large impact of
 - inlet water on the resulting water quality status.

The spatial propagation of inlet water is important for water quality and nature reserve management, especially when the inlet water quality differs from locally discharged water. For example, when the inlet water carries contaminants or pathogens,

- ¹⁵ avoiding direct use of this water for agricultural irrigation may be desirable (e.g. Srinivasan and Reddy, 2009). In nature reserves, the spatial distribution of plant communities may be constrained by the spatial impact of inlet water (e.g. Runhaar et al., 1996). Furthermore, water managers invest heavily in surface water quality monitoring networks. Whether the sampled water consists of locally discharged water or distant inlet
- ²⁰ water is important for the interpretation of the point-scale concentration measurements. For example, monitoring the effects of changes in land management, such as the reduction of fertilizer use, should focus on locally discharged water at sampling locations unaffected by distant (water inlet) sources of pollution.

The impact of inlet water relative to other sources of water and contaminants is often assessed through water and mass balance studies (e.g. Van den Eertwegh et al., 2006; Kieckbusch and Schrautzer, 2007; Antonopoulos, 2008). This approach deals with the watershed as one "black box" reservoir with several water and contaminant inputs and outputs. Water and mass balance studies do not provide any insight into the spatial propagation of the inlet water within the surface water system of an agricultural area or



nature reserve. Whether the inlet water remains within the main channels or whether it also penetrates into the smaller ditches remains unknown. This information could be obtained from detailed hydrological water and solute transport modeling. However, these models are typically calibrated towards an optimal representation of water levels or fluxes at specific monitoring locations (Gallart et al., 2007). Their performance in

reproducing correct spatial patterns of inlet water proportions is not known, due to the absence of appropriate measurements for calibration and validation.

The objective of this study was to obtain a spatial image of the contribution of river inlet water to the water composition in the channels and ditches of a hydrologically complex palder system. We introduce cadelinium as an effective tracer for the dates

- ¹⁰ complex polder system. We introduce gadolinium as an effective tracer for the detection of areas affected by inlet water. This first time application of gadolinium for this purpose was a part of an extensive monitoring and modeling study of the agricultural polder "Quarles van Ufford" in The Netherlands. In addition to the monthly and weekly nutrient concentration measurements, we sampled 22 monitoring locations twice for
- analysis of gadolinium and other Rare Earth Elements (REE). One sampling run was conducted after a dry period in summer on 5 August 2010 and the second run took place after a wet period in autumn on 22 October 2010. Through these measurements, we successfully measured the impact of inlet water at the sampled locations during distinct hydrological conditions. These results enabled us to improve our inter pretation of the water quality monitoring data and to evaluate the spatial performance
 - of our water and solute transport models.

2 Methods

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2.1 Study area

This study focuses on the 120 km² polder area "Quarles van Ufford" in The Netherlands (Fig. 1) (51°50′ N, 5°35′ E). The polder is located between the rivers Meuse (south) and the main Rhine branch called "Waal" (north). Surface elevations in the relatively



flat area range from 3 to 7 m above sea level from west to east. About 80% of the area is in agricultural land use, of which 71% is grassland, 15% is maize, 7% is fruit orchard, and 7% is arable land. 16% of the total area is urban and some small patches of forest cover 4% of the polder. The shallow subsurface mainly consists of Holocene fluvial floodbasin deposits (clay and silt) with an average thickness of 3–6 m. Locally, sandy channel bed deposits and inland aeolian dune sand reach the surface.

The deeper subsurface consists of Pleistocene fluvial and aeolian deposits (sand and gravel).

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The polder Quarles van Ufford has a semi-humid sea climate with an average yearly precipitation of 800 mm and an average yearly estimated evaporation of 550 mm, resulting in an average estimated yearly recharge of 250 mm. The surface water system consists of a dense artificial network of channels and ditches with a total length of 850 km (Fig. 1). The spacing between the ditches averages 200 m. Inlet of water is possible through five human controlled inlet locations, as indicated in Fig. 1. The two inlets in the south directly divert water from the river Meuse into the polder. The other inlets in the east bring water from the neighboring polder "Bloemers". This water is a

- inlets in the east bring water from the neighboring polder "Bloemers". This water is a mixture of inlet water from the Meuse and local agricultural and urban drainage water from polder "Bloemers". There are no inlet structures that make it possible to divert river water from the Waal into the polder.
- Excess water from Quarles van Ufford drains towards the river Meuse at one outlet location at the western side of the polder (Fig. 1). The overall surface water flow direction is from east to west. Locally, the flow directions are complex and only marginally known. The local flow directions depend on the precedent weather conditions, weir crest levels, and on the intermittent influence of several small-scale pumping stations.
- ²⁵ In a discharge situation, the flow is directed from the smaller secondary and tertiary headwaters towards the first order main channels. During dry periods opposite flow directions may occur, as inlet water is distributed through the first order channels and possibly penetrates into the smaller secondary and tertiary ditches.



The nutrients in the surface water system mainly originate from inlet water and agricultural drainage. Most urban sewage water is treated and discharged directly to the Meuse and the Waal. One small-scale sewage treatment plant in the south discharges its effluent to the local surface water system, which can influence the local water quality

⁵ (Fig. 1). Several sewer overflows located in the urban areas may also have local impact on water quality.

2.2 Research activities

The study area Quarles van Ufford is one of four pilot catchments in the research project "Catchment Monitoring". This project aims to monitor the impact of the Dutch manure legislation on the nutrient concentrations at the catchment-scale (Woestenburg and Van Tol-Leenders, 2011). We studied the nutrient concentrations and fluxes in the pilot catchments by combining detailed water quality monitoring with process-based models of catchment-scale nutrient transport.

In Quarles van Ufford, 23 water quality monitoring locations were sampled from
 2003–2010 (Fig. 2). The monitoring network was designed to provide a good spatial coverage over the polder and to include main channels, smaller ditches, the inlet locations, and the catchment outlet. 16 locations were sampled monthly and 6 locations were sampled weekly. A continuous flow proportional sampler was situated at the catchment outlet. The samples were analyzed for PO₄, NH₄, NO₃, SO₄, N_{kj}, N_{tot}, P_{tot},
 O₂, pH, EC, CI, alkalinity, and Chlorofyl-a.

The integrated water and nutrient transport model consists of four coupled submodels for water flow in the subsurface (SWAP, Kroes et al., 2008), nutrient transport in the subsurface (ANIMO, Groenendijk et al., 2005), water flow in the surface water system (SWQN, Smit et al., 2009), and nutrient transport in the surface water system (NUSWAlite, Siderius et al., 2009). Siderius et al. (2011) describe the integrated model calibration and validation for Quarles van Ufford.



2.3 Tracer method: gadolinium

We used the Rare Earth Element (REE) gadolinium (Gd) as a tracer to assess the inlet water influence at the monitoring locations. Gd has been used in several studies to trace urban wastewater through rivers, lakes, estuaries, and coastal seawaters

- ⁵ (Möller et al., 2000; Kulaksiz and Bau, 2007; Rabiet et al., 2009; Petelet-Giraud et al., 2009; Lawrence and Bariel, 2010). The widespread occurrence of elevated Gd concentrations results from the use of a very stable Gd complex (Gd-DTPA) as a contrast agent in magnetic resonance imaging (MRI) in hospitals. Patients are injected with the Gd complex prior to their MRI-scan. After the research, the complex is excreted and
- transported conservatively through sewage systems and wastewater treatment plants towards the surface water system. As a consequence, rivers and other surface water bodies downstream of populated areas generally show REE patterns with positive Gd anomalies (Möller et al., 2000). This study is the first to apply REE patterns and Gd anomalies to trace the propagation of inlet water into a polder area.
- To quantify the Gd-anomaly, it is common practice to first normalize the measured REE concentrations against the average natural REE composition of the earth's upper crust (e.g. Rabiet et al., 2009). Hereto, the REE composition of the North American Shale Composite (NASC, Hannigan and Sholkovitz, 2001) is used as a reference standard. The Gd-anomaly is calculated from the shale-normalized concentrations of Gd and its neighboring REE elements Samarium (Sm) and Terbium (Tb):

$$Gd_{ano} = \frac{Gd_{N}}{(0.33 \cdot Sm_{N} + 0.67 \cdot Tb_{N})}$$

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with Gd_N is the shale-normalized Gd concentration, Sm_N is the shale-normalized Sm concentration, and Tb_N is the shale-normalized Tb concentration (e.g. Petelet-Giraud et al., 2009). Gd-anomalies larger than 1 suggest elevated Gd concentrations relative to the other REE elements. The reproducibility of the calculated gadolinium anomalies is approximately 0.1.



(1)

2.4 Sampling timing, locations, and procedures

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We sampled the surface water monitoring locations for REE analysis on 5 August and 22 October 2010. These two sampling moments represented two extremes in the hydrological situation. On 5 August, hydrological conditions were very dry and we
anticipated for a relatively large impact of river water inlet on the local surface water composition throughout the polder. The total precipitation deficit during the preceding two months was 118 mm (101 mm precipitation and 219 mm evapotranspiration). The volume of water inlet during these months was 5.3 × 10⁶ m³ (50 mm). On 22 October, on the other hand, hydrological conditions were very wet. We expected local drainage
of excess rainfall water to dominate the local surface water composition and a much lower impact of inlet water compared to 5 August. The total precipitation excess during the two months before 22 October was 184 mm (275 mm precipitation and 91 mm

evapotranspiration). Water inlet during these months was 2.7 × 10⁶ m³ (25 mm). Both on 5 August and 22 October 2010, surface water was sampled at 22 locations
¹⁵ in the polder Quarles van Ufford. The locations were selected from the regular water quality sampling program. We included the five inlet locations, the catchment outlet, some main channels, and some small agricultural ditches. In addition, we selected a sampling location where we expected the minimum influence of inlet water. This location was chosen in the first part of a ditch draining a slightly elevated field, situated on an old river sand dune.

The samples were taken using a peristaltic pump and filtered through $0.45 \mu m$ polyethersulfon filters. We collected the samples in pre-washed 100 ml HDPE bottles. The samples were acidified with 1 % ultra pure HNO₃ and stored at 4 °C until analysis. The samples were analyzed using an innovative ICP-MS setup (Inductively Coupled Plasma Mass Spectrometry, Thermo Fisher Scientific, X-serie 2, Waltman, MA, USA). Very accurate measurements of low REE-concentrations (<0.001 ppb) were pro-

duced by coupling the ICP-MS with an MP2 micro-peripump, a FAST precision pump, and an APEX desolvation nebulizer (all Elemental Scientific, Omaha, NE, USA), which



improved the ionization efficiency of the pre-dry particles. The approximate accuracy of this ICP-MS setup is 10%. We did not use the frequently applied pre-concentration procedure to increase the REE concentrations before analysis (Hennebrüder et al., 2004).

5 3 Results

3.1 Gd-anomalies

The Gd-anomalies that were calculated from the REE measurements of 5 August and 22 October 2010 are shown in Fig. 3. On 5 August, the Gd-anomalies of the diverted Meuse water at the two direct inlet locations were 8.6 and 9.5 (Fig. 3a). The other inlets
that bring water from the neighboring polder Bloemers showed lower Gd-anomalies of 4.2, 5.8, and 7.9. This water partly consists of inlet water from the Meuse and partly of local agricultural and urban drainage water. The relatively high Gd-anomaly of 7.9 is probably caused by the short and direct flow route from the Meuse water inlet of polder Bloemers towards this location. Less mixing with locally drained water occurred to the two other inlet locations from polder Bloemers.

Despite the positive Gd-anomalies at the inlet locations, some of the measurements show very low Gd-anomalies of 1.2 and 1.3 on 5 August (a value of 1 means no Gd-anomaly). The lowest Gd-anomaly (1.2) was measured at the specially selected location where a minimum influence of inlet water was expected. The two locations with

²⁰ Gd-anomalies of 1.3 are relatively small agricultural headwater ditches. These negligible values indicate that no inlet water penetrates up into these small ditches.

In most larger channels, Gd-anomalies on 5 August ranged between 3.9 and 6.8. These values indicate that this water consist of a mixture of inlet water and locally drained water. We found one very high Gd-anomaly of 30.3 in the south. This location is in a small ditch near the small-scale sewage treatment plant that discharges its

²⁵ is in a small ditch near the small-scale sewage treatment plant that discharges its effluent to the local surface water system (Fig. 1). In dry periods, the water managers



block the discharge from this area to prevent water shortage and the sewage treatment plant effluent spreads out over the sub-catchment. The extremely high Gd-anomaly at this monitoring location indicates that the effluent influenced local surface water quality on 5 August. In the northeast, we found another high Gd-anomaly of 10.2 with an unknown source. A possible explanation of this high value is the influence of a nearby sewer overflow.

On 22 October, the Gd-anomalies for the two direct Meuse water inlet locations of 8.6 and 8.4 were similar to those measured on 5 August. However, the Gd-anomalies of the inlet locations from polder Bloemers were lower on 22 October (2.4, 2.1, and 3.5). In addition, all other monitoring locations within Quarles van Ufford also show much lower Gd-anomalies compared to 5 August. These results indicate a decrease in the proportion of diverted Meuse water in the polder and a larger influence of locally drained water.

At the monitoring location near the sewage treatment plant, the Gd-anomaly de-¹⁵ creased from 30.3 on 5 August to 1.1 on 22 October. Possibly, the effluent of the treatment plant does not spread out through the sub-catchment during wet conditions and therefore does not influence the water quality at this location on 22 October. Another possible explanation is that no MRI-patients have contributed Gd to this smallscale sewage treatment plant at the time of the second measurement round. The

Gd-anomaly at the location in the northeast decreased from 10.2 on 5 August to 4.2 on 22 October. Still, the relatively high value relative to the other locations indicates that the unknown local source of Gd is also active during wet conditions.

3.2 Surface water quality measurements

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The consequence of the spatial propagation of inlet water within the polder depends on the difference between the composition of the diverted river and the composition of the locally drained water. Therefore, we compared the water quality data from the 2003–2010 regular monitoring program of the locations that were most affected by inlet water with the locations that were not or only marginally affected. In Fig. 4, the



left-side boxplots summarize the measured concentrations at the two monitoring locations with the lowest Gd-anomalies (\leq 1.3). The right-side boxplots give the results for the locations near the direct inlets with Gd-anomalies ranging from 7.9 to 9.5.

Figure 4 shows clear differences in water composition between the locations that
⁵ were most affected by diverted river water and those that represent locally drained water. P_{tot} concentrations were significantly higher in the locally drained water dominated locations, while N_{tot} concentrations were higher in the inlet water dominated locations. The higher N_{tot} concentrations at the locations that were mainly influenced by inlet of diverted river water can be attributed to the higher NO₃ concentrations. The low NO₃
¹⁰ concentrations in locally drained water are probably caused by denitrification and uptake of NO₃ in biomass. The SO₄ concentrations were higher in the inlet dominated locations. The EC was highest in the local drainage locations, while the highest pH-values occurred at the inlet water dominated locations.

3.3 Modeled proportions of diverted river water

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The proportions of diverted river water within the surface water system of Quarles van Ufford were further analyzed through the integrated water and nutrient transport model. Figure 5 gives the modeled fractions of inlet water in the main channels and ditches at the sampling dates 5 August and 22 October 2010. The modeled fractions confirm the large difference in the proportions of inlet water between the two sampling rounds that was also found from the REE-measurements. On 5 August, during very dry hydrological conditions, the inlet water from the Meuse and the neighboring polder "Bloemers"

- controls the water composition in most main ditches and channels. The low inlet water proportions in the northern part of the polder are probably caused by an incorrect
- parameterization of a small-scale pumping station or an incorrect weir crest level. The model results for 22 October show much lower inlet water proportions. During very wet conditions, the large amounts of locally drained water control the chemical composition of most channels. Near the active inlet locations, some channels still have a large proportion of inlet water on 22 October.



The small agricultural headwater ditches were not explicitly incorporated in the model, but were represented as a lumped storage. The fraction of inlet water in this lumped storage decreased from 51 % on 5 August to 5.1 % on 22 October.

4 Discussion and conclusions

In this study, we aimed to obtain a spatial image of the propagation of diverted river water into the channels and ditches of a hydrologically complex polder system. Our measured REE patterns and Gd-anomalies revealed a large influence of inlet water at most water quality monitoring locations during very dry conditions on 5 August. However, the diverted river water did not reach the locations in the small agricultural head water ditches. During very wet conditions on 22 October, the impact of diverted river water vas very low, except for the measurement locations near the direct inlets.

We illustrated the significance of the spatial propagation of diverted river water for water quality patterns in the area by relating our REE measurements to the results of the 2003–2010 water quality monitoring program. A clear difference was shown between the water composition at inlet water dominated monitoring locations with high Gd-anomalies and local drainage water dominated monitoring locations with low Gdanomalies. The locally drained water of Quarles van Ufford is characterized by a relatively high P_{tot} concentration and EC and by relatively low pH and low concentrations N_{tot}, NO₃, and SO₄ (Fig. 4). The water composition at the inlet water dominated locations is characterized by a relatively low P_{tot} concentration and EC and by relatively high pH and high concentrations N_{tot}, NO₃, and SO₄. These results illustrate the value of knowing the inlet water fractions for the interpretation of water quality monitoring data. In addition, the design of a water quality monitoring network could benefit from a spatial image of the propagation of inlet water.



We also applied the REE measurements from this study to validate the results of the integrated nutrient transport modeling. We showed that the modeled fractions of inlet water on 5 August and 22 October 2010 were consistent with the measured Gd-anomaly patterns. Especially in complex systems such as the Quarles van Ufford polder, hydrological modeling can benefit from using tracers to verify flow directions and mixing proportions of distinct water sources.

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Although several studies reported on the adverse effects of the inlet of diverted river water on surface water quality (e.g. Roelofs, 1991; Runhaar et al., 1996; Delaune et al, 2005), we did not find any international scientific literature on the spatial propagation of inlet water within a surface water bady. Before this study, our accumption was that the

- inlet water within a surface water body. Before this study, our assumption was that the diverted river water would propagate up into all small headwater ditches in dry periods. Figure 6a visualizes this concept. To compensate for the water loss through evapotranspiration, ditch water will infiltrate from the ditches into the agricultural fields. The headwater ditches will be replenished with diverted river water distributed through the
- ¹⁵ main channels and ditches. Our REE measurement results, however, contradicted our initial conceptual model as they proved that the diverted river water did not penetrate into the headwater ditches. We developed an alternative conceptual model, which is shown in Fig. 6b. In this concept, the locally drained surface water is "pushed back" into the headwater ditches by the water pressure from the main ditches and channels
- that are filled with inlet water. Instead of a gradual decline of the proportion of inlet water (Fig. 6a), a relatively sharp transition from inlet water dominated surface water to locally drained surface water exists (Fig. 6b). This concept is supported by previous research in a similar polder (Hendriks, 1990) and corresponds with experiences of the local water managers. Still, the spatial propagation of inlet water also depends on local hydrological conditions.

The improved understanding of water and nutrient sources in Quarles van Ufford helps to identify appropriate water quality mitigation options. A reduction of N_{tot} concentrations in the polder could be established by reducing the river water inlet volumes or by reducing the inlet water N_{tot} concentrations. A reduction of P_{tot} concentrations



can be obtained by reducing the leaching from the agricultural fields or by increasing the water inlet which enhances the "flushing" of the polder. A complicating aspect of changing the inlet volumes is that this may affect the residence time of water within the polder system. Stopping the water inlet would lead to a dominance of locally drained water and to longer residence times, especially during dry periods. It is unclear whether the increased residence time will lead to a building up of even higher concentrations of P_{tot} and other agrochemicals or that a longer residence time will increase the effect of biochemical processes that reduce the concentrations.

Gd proved to be an excellent tracer to study the spatial propagation of diverted river ¹⁰ water into a hydrological complex polder system. As suggested before by Möller et al. (2000), the occurrence of very stable Gd-DTPA complex in the effluent of wastewater treatment plants brings new opportunities for hydrological research. The Gd-DTPA complex shows minimal chemical interaction with its surroundings under natural conditions and has a high residence time in the water system, which makes it a good tracer ¹⁵ to study the mixing of water from different sources. Gd-anomalies can also be used as an indication for urban wastewater influence and the possible occurrence of pollutants and pathogens.

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Fig. 2. Surface water monitoring locations in Quarles van Ufford.

















Fig. 5. Modeled proportions of inlet water in the main ditches and channels on 5 August and 22 October 2010.





Fig. 6. Visualization of the two conceptual models of the spatial propagation of inlet water into a polder system.

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