

General comments for the reviewers

We wish to thank anonymous reviewer #1 and Saif Rafi for their constructive and useful comments. Their reviews helped improve the manuscript.

On behalf of all co-authors,
Floris Naus

Author's response to anonymous referee #1

1. Reviewer's comment: This study explores groundwater salinity variations in a village in southwestern Bangladesh using (1) geological evolution, (2) groundwater and lithological sampling, and (3) modeling. The paper is well written and logically describes the aim, methods, and findings of the groundwater investigation.

Author response: We thank the referee for his time to review our paper.

2. Reviewer's comment: Figure 1 seems insufficient for the reader to fully understand the field site. An additional satellite image would give the reader a better sense of the region. Perhaps consider a two-paneled figure with a satellite image adjacent to the site diagram. An additional figure in an appendix would also suffice.

Author response: We agree that we can be more clear.

Changes in manuscript: We will add a figure with satellite imagery, either in the appendix or as a two-paneled figure.

3. Reviewer's comment: To connect back to the first paragraph of your introduction, I would be interested in how you could relate your findings back to drinking water availability. Is there enough freshwater to sustain the villages with drinking water? Is it likely that the freshwater recharge in high elevation regions will recharge at a pace similar to the salinization from the nearby low-lying regions and aquaculture? See line 520.

Author response: This is indeed an important question. Unfortunately, the exact fluxes of water are hard to estimate. It is difficult to assess the exact amount of water that is being extracted and used, as the fresh water is used without regulation for both drinking/household water and irrigation water. Additionally, it is difficult to determine or estimate the recharge of fresh water, because of the (varying) clay cover thickness and the uncertain amount of recharge from fresh water ponds (article line 486-494).

We state that salinization from the aquaculture ponds could be a threat to the fresh groundwater, following the observation of salinization in the groundwater in the southern part of the village (Figure 7). The exact impact of salinization of low-lying regions on the fresh groundwater requires temporal measurements and modelling of groundwater flow.

For these reasons, we didn't aim to make an estimate of the fluxes and the sustainability of the fresh groundwater resource under present-day circumstances, which we acknowledge as a shortcoming in line 550. We see this as beyond the scope of this work, and as a topic for further research.

Changes in manuscript: No change

4. Reviewer's comment: One of the main conclusions is that elevation is correlated with groundwater salinity, where higher elevations are fresher than lower areas. This appears to be a poldered region surrounded by tidal creeks (as noted by the embankments in Fig. 2). The polder regions in SW Bangladesh are highly altered. The inhabitants have cleared forests, built embankments, compacted the landscaped by limiting flooding, dug ponds, and raised areas around their villages. There is a massive amount of sediment movement at the surface. Does this study area have ~2 m natural variation in topography (partial answer in 4.1.3 b)? Or is the difference in elevation observed across the transect a recent development due to anthropogenic activity? If so, does that change how you would interpret the correlation between groundwater salinity and elevation? Could the salinity variations be more closely related to clay cap thickness?

Author response: We found a natural variation in topography of approximately 1.5m, based on our elevation measurements (par. 2.2.2).

It is true that a large anthropogenic influence is typical for the topography in the polders in Bangladesh. In our specific study area, we think that the main elevation differences are natural, for multiple reasons:

- The high area is very large, with a width of approximately 2 km (Figure 1), which is much larger than the anthropogenically created narrow high ridges next to creeks or next to roads that are present throughout most of the polders.
- The high area has a fluvial soil type, which suggests natural formation (FAO, 1959).
- This high lying area meanders naturally through the landscape, which also suggests a fluvial, natural origin.

“Could the salinity variations be more closely related to clay cap thickness?” When looking at Figure 3, we observe that the salinity of the groundwater is not only related to the clay cap thickness. In the low-lying areas directly surrounding the high-lying village (near N1, near P8 and near P14), the clay cap is similarly thin as below the high-area, but the groundwater salinity is saline instead of fresh. This indicates that clay thickness alone can't explain the salinity differences.

Changes in manuscript: No change

5. Reviewer's comment: What are the coordinates for the study area? This should be included in Fig. 1 or line 76.

Changes in manuscript: We added coordinates to Figure 1.

6. Reviewer's comment: I'm a bit unsure how the “groundwater observation well” and “groundwater sample” data are different. Why include P16 and P17?

Author response: ‘Groundwater observation well’ indicates a groundwater sample taken from a newly placed well (with a filter of 5 ft). A groundwater sample refers to any sample we took from the groundwater, including samples from household tubewells.

The installation of P16 and P17 had two goals:

1. They were used to better assess the lithological situation between P11 and P10.
2. They are used to determine whether density driven flow could lead to salinization of the water below the thick clay layer (see line 453).

Changes in manuscript: Clarified the type of tubewell: Added ‘household’ in line 124 and in Table 1.

Clarified the use of P16 and P17: added '*to get more detailed lithological information between P11 and P10*' in line 100.

7. Reviewer's comment: Some of your groundwater tubes appear to be installed in the clay. In similar field environments, I have had difficulty extracting groundwater samples from clay layers, especially using hand-flapper drilling methods. It is always important to acknowledge the potential inaccuracies associated with this type of field environment. This type of tube installation can be a major source of error when dating or looking at cation exchange. Are you confident that you were able to evacuate the installation water, pump the tube, and extract groundwater despite the low permeability of the clay layer? It may be worth noting the potential limitations of the hand-flapper installation method.

Author response: We are indeed not very clear in this regard. We didn't extract water from the clay by groundwater tubes, but we used the open auger boring method (De Goffau et al., 2012, see pictures below). We drilled a hole with a hand auger, without any drilling fluid. Then we waited for the hole to fill with water, which we sampled by inserting a sample bottle into the hole with a stick, which we pulled back out using a rope that was attached to the bottle. We called these samples retrieved from 'Hand-auger borings' in Table 1, but we didn't specifically describe the field procedure of these samples.



Changes in the manuscript: Added a sentence in line 126 '*To sample porewater from the clay, we used the open auger boring method (De Goffau et al., 2012). We drilled a hole with a hand auger, without any drilling fluid. Then we waited for the hole to fill with water, which we sampled by inserting a sample bottle into the hole with a stick, which we pulled back out using a rope that was attached to the bottle.*' Changed the name of these samples from 'hand-auger borings' to 'open auger borings'

8. Reviewer's comment: The variation in the clay cap thickness is substantial. Figure 3 shows the thickness varying from 5 to 35 m. The thickness of the cap has to play a major role in present-day surface water influence on groundwater salinity. Yet Figure 7 shows both freshening and salinization under the thickest region of the clay cap (near P9, P10, and P15). Can you better explain how the freshening and salinization stages can differentiate between current day processes (e.g. salinization due to aquaculture) and depositional processes (e.g. salinization due to evaporation of pore water during deposition of sediments 10 kyr BP)? Can you discuss any

uncertainties associated with this methodology (without the use of dating), especially in an area with highly variable connate water? Most of these questions arose from the section 3.8.2. You do a nice job of answering some of these questions in your phased subsurface evolution in section 4.

Author response: The stage of freshening or salinization alone cannot be used to differentiate between salinization by aquaculture or salinization by depositional processes. The salinization stages were used to determine the direction of salinization (see section 3.8.2).

The difference between present-day and depositional processes are, instead, postulated after combining the field results with the geological history described in literature, which we describe in section 4.1. In section 4.2, we acknowledge that our postulated evolution of the groundwater salinity is uncertain, and that it should be seen as a conceptual model.

Changes in the manuscript: Added a sentence in line 550: *‘Without age dating, we can’t determine the exact moment of salinization or freshening that has occurred.’*

9. Referee comment: Do you see any seasonal changes in groundwater salinity associated with exchange with the tidal channels? If you see signatures of salinization and freshwater recharge in the thinned cap areas, how do you expect the groundwater signatures to compare near the tidal channels?

Author response: We didn’t look in detail at interaction of the aquifer with the tidal river, nor did we study temporal variety in salinity in the groundwater in great detail. As a consequence, we have no evidence for seasonal groundwater salinity near the tidal channel.

Without detailed and temporal measurements of water levels/heads in both the river and the aquifer, it is difficult to say what we expect near the tidal rivers. There are many factors at play that control exchange between the tidal channel and the aquifer, such as the depth of the tidal river, the thickness of the clay layer below the tidal river, possible clogging layers, changing water and salinity levels due to seasonality and tides (Bhuyian et al., 2012), possible salinity gradients in the tidal rivers and possible density driven flow.

At the northern tidal channel, not much exchange between the aquifer and the channel is expected, because of how narrow and shallow the channel has become.

Changes in the manuscript: No change

Author’s response to the short comment by Saif Rafi

1. Reviewer’s comment: This manuscript analyzes ground water salinity variations in southwestern Bangladesh using geological reconstruction, lithological sampling and modeling. The manuscript addresses a very important issue of water resource problem that the southwestern Bangladesh currently facing. According to the script ground water salinity mostly depends on the surface elevation. In the higher lying area with a thin surface clay layer always have a clear pattern of fresh ground water which stored by directly infiltration or via rainfed pond. The fresh ground water is bordered by brackish-saline water at greater depth under the higher area. On the other hand, lower area often flooded by tides and tidal surges and in the direction of the adjacent lower area saline ground water is found only at relatively shallow depth below aqua cultural ponds. So, here we can find that it is possible to clear the pattern in ground water salinity by using salinity data, PHREEQC interpreted cation

exchange data and to identify the hydrological process and geographical and geological control. Thickness, relative elevation and land used are the most common geographical controlling factor in which the ground water salinity depends.

However, there are some concern summarized below and I hope these comments could help the authors to improve the manuscript.

Author's response: We thank Saif Rafi for his time to review our paper.

2. Reviewer's comment: I am concerned about the sample preservation process use in the study since nothing was described in the paper. How were the sample preserved since you are running IC and ICP-MS back to the Netherland. I am thinking about NaCl concentration.

Author's response: After sampling, the samples were sealed tightly with the cap of the tube. In the field, it was not possible to actively cool the samples, but we kept the samples out of the sun. As soon as we got back to our overnight stay in the late afternoon, the samples were actively cooled, except when there was a power cut. During the transport to the Netherlands (by plane), the samples were put in a dark place, but they were not actively cooled. After arrival in the Netherlands, they were again cooled until analysis. The concentrations of major cations and chloride are not expected to have changed, as the cap was sealed tightly, and no leakage has been detected at all. **Changes in manuscript:** We added a sentence: *"The samples were kept out of the sun and cooled as much as possible."* (line 127)

3. Reviewer's comment: See line 520. To connect back to the first paragraph of your introduction, I would be interested in how you could relate your findings back to drinking water availability. Is there enough freshwater to sustain the villages with drinking water? Is it likely that the freshwater recharge in high elevation regions will recharge at a pace similar to the salinization from the nearby low-lying regions and aquaculture?

Author's response: (same answer as to comment 3 of referee 1) This is indeed an important question. Unfortunately, the exact fluxes of water are hard to estimate. It is difficult to assess the exact amount of water that is being extracted and used, as the fresh water is used without regulation for both drinking/household water and irrigation water. Additionally, it is difficult to determine or estimate the recharge of fresh water, because of the (varying) clay cover thickness and the uncertain amount of recharge from fresh water ponds (article line 486-494).

We state that salinization from the aquaculture ponds could be a threat to the fresh groundwater, following the observation of salinization in the groundwater in the southern part of the village (Figure 7). The exact impact of salinization of low-lying regions on the fresh groundwater requires temporal measurements and modelling of groundwater flow.

For these reasons, we didn't aim to make an estimate of the fluxes and the sustainability of the fresh groundwater resource under present-day circumstances, which we acknowledge as a shortcoming in line 550. We see this as beyond the scope of this work, and as a topic for further research.

Changes in manuscript: No change

4. Reviewer's comment: Your whole paper is based on the correlation between surface elevation and ground water salinity. Ground water salinity is higher when the elevation is lower and ground water salinity is lower

when the elevation is higher (Fig 2). Thickness, relative elevation and land used are the most common geographical controlling factor in which the ground water salinity depends (line 546 and 546). But as far I know that 80 percent of the land mass of Bangladesh is made up of fertile alluvial lowland called Bangladesh plain. In plain area salinity basically depend on precipitation, infiltration and evapotranspiration (Yan,2015). So, from my point of view you should think about this too.

Author's response: Yan et al. (2015) researched the temporal groundwater salinity variation between the dry and wet season of shallow groundwater (5 m depth) in a very dynamic system, having a highly permeable soil and a high amount of infiltration. In Southwestern Bangladesh, the soil consists of clayey material with a much lower soil permeability, which leads to a much lower amount of infiltration. As a consequence, the system in Southwestern Bangladesh is much more stagnant than the system researched by Yan et al. (2015). The seasonal variation in precipitation and evapotranspiration, therefore, do not lead to changes in groundwater salinity. Additionally, we research the groundwater salinity at greater depths than Yan et al. (2015). The deeper groundwater is also less influenced by dynamics throughout the seasons. Instead, we describe that the differences in groundwater salinity form due to differences in surface conditions over longer time periods, which we describe in our article to depend on relative elevation and land use. We also acknowledge the importance of differences in infiltration: When there is a thick surface clay layer, the influences of the salinity conditions at the surface do not impact the groundwater salinity at larger depth.

Changes in manuscript: No change

5. Reviewer's comment: Line 63: Use it as a simple sentence because you are using too much “and” in one sentence and there should be no comma (and,) after and.

Author's response: The first ‘and’ separates ‘freshening and salinization processes’, while the second ‘and’ separates ‘processes’ from ‘salinity’. We cannot remove either of them. The comma after the second ‘and’ is there because of the word ‘therefore’. We think the sentence is grammatically correct.

Changes in manuscript: No change

6. Reviewer's comment: Line 100: Two groundwater observation wells P16 and P17 were installed a year later but the sample campaign ended so no water is available for them. So, I don't understand why you installed P16 and P17.

Author's response: *(same answer as to comment 6 of referee 1)*. The installation of P16 and P17 had two goals:
1. They were used to better assess the lithological situation between P11 and P10.
2. They are used to determine whether density driven flow could lead to salinization of the water below the thick clay layer (see line 453).

Changes in the manuscript: added ‘*to get more detailed lithological information between P11 and P10*’ in line 100.

7. Reviewer's comment: What is the difference between “ground water observation well” and “groundwater sample”?

Author's response: *(same answer as to comment 6 of referee 1)*

‘Groundwater observation well’ is a newly placed well (with a filter of 5 ft). A groundwater sample refers to any sample we took from the groundwater, including samples from groundwater observation wells and household tubewells.

Changes in manuscript: Clarified the type of tubewell: Added ‘household’ in line 124 and in Table 1.

8. Reviewer’s comment: It is always important to acknowledge inaccuracy and error in the field work. Some of your tube is installed in the clay and it is very difficult to extract water from this condition. So, are you confident enough that there is no error in your field work? Because you didn’t mention anything about that.

Author’s response: (same answer as to comment 7 of referee 1) We are indeed not very clear in this regard. We didn’t extract water from the clay by groundwater tubes, but we used the open auger boring method (De Goffau et al., 2012, see pictures below). We drilled a hole with a hand auger, without any drilling fluid. Then we waited for the hole to fill with water, which we sampled by inserting a sample bottle into the hole with a stick, which we pulled back out using a rope that was attached to the bottle. We called these samples retrieved from ‘Hand-auger borings’ in Table 1, but we didn’t specifically described the field procedure of these samples.



Changes in the manuscript: Added a sentence in line 126 ‘To sample porewater from the clay, we used the open auger boring method (De Goffau et al., 2012). We drilled a hole with a hand auger, without any drilling fluid. Then we waited for the hole to fill with water, which we sampled by inserting a sample bottle into the hole with a stick, which we pulled back out using a rope that was attached to the bottle.’ Changed the name of these samples from ‘hand-auger borings’ to ‘open auger borings’

9. Reviewer’s comment: In all over the manuscript there is lots of spacing problem. I can give you some example. In line 55: “Boun daries” should be “boundaries”. In line 66: “Re constructed” should be “Reconstructed”. And you can also check Line 216,217,232,244.

Author’s response: Remarkably, we didn’t see these spacing problems when we downloaded and viewed the pdf document on our computers, so we couldn’t change anything. We have no explanation for the errors found in lay-out.

10. Reviewer's comment: Do you see any seasonal changes in groundwater salinity associated with exchange with the tidal channels? If you see signatures of salinization and freshwater recharge in the thinned cap areas, how do you expect the groundwater signatures to compare near the tidal channels?

Author's response: (*same answer as to comment 9 of referee 1*) We did neither look in detail at interaction of the aquifer with the tidal river nor did we study temporal variety in salinity in the groundwater in great detail. As a consequence, we have no evidence for seasonal groundwater salinity near the tidal channel.

Without detailed and temporal measurements of water levels/heads in both the river and the aquifer, it is difficult to say what one may expect near the tidal rivers. There are many factors at play that control exchange between the tidal channel and the aquifer, such as the depth of the tidal river, the thickness of the clay layer below the tidal river, possible clogging layers, changing water and salinity levels due to seasonality and tides (Bhuyian et al., 2012), possible salinity gradients in the tidal rivers and possible density driven flow.

At the northern tidal channel, not much exchange between the aquifer and the channel is expected, because of how narrow and shallow the channel has become.

Changes in the manuscript: No change

11. Reviewer's comment: In Figure 1, there is no coordinates of the study area.

Changes in manuscript: We added coordinates to Figure 1.

12. Reviewer's comment: Figure 3 shows the thickness varying from 5 to 35 m. The thickness of the cap has to play a major role in present-day surface water influence on groundwater salinity. Yet Figure 7 shows both freshening and salinization under the thickest region of the clay cap (near P9, P10, and P15). Can you better explain how the freshening and salinization stages can differentiate between current day processes (e.g. salinization due to aquaculture) and depositional processes (e.g. salinization due to evaporation of pore water during deposition of sediments 10 kyr BP)?

Author's response: (*Same answer as to comment 8 of referee 1*). The stage of freshening or salinization alone cannot be used to differentiate between salinization by aquaculture or salinization by depositional processes. The salinization stages were used to determine the direction of salinization (see section 3.8.2).

The difference between present-day and depositional processes are, instead, postulated after combining the field results with the geological history described in literature, which we describe in section 4.1. In section 4.2, we acknowledge that our postulated evolution of the groundwater salinity is uncertain, and that it should be seen as a conceptual model.

Changes in the manuscript: Added a sentence in line 550: '*Without age dating, we can't determine the exact moment of salinization or freshening that has occurred.*'

13. Reviewer's comment: "Figure 1" seems like insufficient and difficult to understand. If you can add an additional satellite image, it will be helpful.

Author's response: (*Same answer as to comment 2 of referee 1*) We agree that we can be more clear.

Changes in manuscript: We will add a figure with satellite imagery, either in the appendix or as a two-paneled figure.

14. Reviewer's comment: In Figure 6, what is that dot line indicated.

Author's response: The dotted line indicates where Z-values are 0 (neutral).

15. Reviewer's comment: In Table 1, you should add more result for ICP-MS and IC test. Example, percentage of Al, As, Be, B etc.

Author's response: We will include a table with the relevant compounds for this study as supplementary material (Cl, Na, Ca, Mg, K, HCO₃, SO₄, Tritium, $\delta^{18}\text{O}\%$, $\delta^2\text{H}\%$)

Changes in manuscript: We will add a table as supplementary material.

List of changes in manuscript

Line numbers refer to the lines in the new manuscript.

1. We added coordinates to Figure 1.
2. We added a side panel with satellite imagery in Figure 1.
3. Changed the caption of Figure 1, to explain the side panel: '*(b) Satellite imagery of the study area*'
4. Clarified the use of P16 and P17: added '*to get more detailed lithological information between P11 and P10*' in line 104-105.
5. Clarified the type of tubewell: Added '*household*' in line 129 and in Table 1.
6. Added a few sentences at lines 131-134: '*To sample porewater from the clay, we used the open auger boring method (De Goffau et al., 2012). We drilled a hole with a hand auger, without any drilling fluid. Then we waited for the hole to fill with water, which we sampled by inserting a sample bottle into the hole with a stick, which we pulled back out using a rope that was attached to the bottle.*'
7. Changed the name of the '*hand-auger borings*' to '*open auger borings*' in Table 1.
8. We added a sentence: "*The samples were kept out of the sun and cooled as much as possible.*" (line 136)
9. Changed 'sulfate' to 'sulphate' in line 182 to keep in line with spelling style throughout the document
10. Added a sentence in line 570: '*Without age dating, we can't determine the exact moment of salinization or freshening that has occurred.*'
11. We added a table of the water composition of the samples as supplementary material.
12. We added that the data used in this study is available in the appendix and supplementary material (line 587).
13. We changed Figure to Fig. when referring a figure throughout the document.
14. We added a reference:

De Goffau, A., Van Leeuwen, T.C., Van den Ham, A., Doornewaard, G.J., Fraters, B.: Minerals Policy Monitoring Programme Report 2007–2010. Methods and Procedures. National Institute for Public Health and the Environment, Bilthoven, The Netherlands, RIVM Report 680717018, 2012

Groundwater salinity variation in Upazila Assasuni (southwestern Bangladesh), as steered by surface clay layer thickness, relative elevation and present-day land use

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Abstract. In the southwestern coastal region of Bangladesh, options for drinking water are limited by groundwater salinity. To protect and improve the drinking water supply, the large variation in groundwater salinity needs to be better understood. This study identifies the palaeo and present-day hydrological processes and their geographical or geological controls that determine variation in groundwater salinity in Upazila Assasuni in southwestern Bangladesh. Our approach involved three steps: a geological reconstruction, based on the literature; fieldwork to collect high density hydrological and lithological data; and data processing to link the collected data to the geological reconstruction in order to infer the evolution of the groundwater salinity in the study area. Groundwater freshening and salinization patterns were deduced using PHREEQC cation exchange simulations and isotope data was used to derive relevant hydrological processes and water sources. We found that the factor steering the relative importance of palaeo and present-day hydrogeological conditions was the thickness of the Holocene surface clay layer. The groundwater in aquifers under thick surface clay layers is controlled by the palaeohydrological conditions prevailing when the aquifers were buried. The groundwater in aquifers under thin surface clay layers is affected by present-day processes, which vary depending on present-day surface elevation. Slightly higher-lying areas are recharged by rain and rainfed ponds and therefore have fresh groundwater at shallow depth. In contrast, the lower-lying areas with a thin surface clay layer have brackish–saline groundwater at shallow depth because of flooding by marine-influenced water, subsequent infiltration and salinization. Recently, aquaculture ponds in areas with a thin surface clay layer have increased the salinity in the underlying shallow aquifers. We hypothesize that to understand and predict shallow groundwater salinity variation in southwestern Bangladesh, the relative elevation and land use can be used as a first estimate in areas with a thin surface clay layer, while knowledge of palaeohydrogeological conditions is needed in areas with a thick surface clay layer.

1 Introduction

In the Ganges–Brahmaputra–Meghna (GBM) river delta, home to 170 million people, availability of safe drinking water is problematic because of the very seasonal rainfall, the likelihood of arsenic occurrence in the shallow groundwater and the pollution of surface water bodies (Harvey et al., 2002; Ravenscroft et al., 2005; Chowdhury, 2010; Sharma et al., 2010; Bhuiyan et al., 2011). In the southwestern coastal region of Bangladesh, suitable drinking water options are even more limited, as here the groundwater is largely brackish to saline

(Bahar and Reza, 2010; George, 2013; Fakhruddin & Rahman, 2014; Worland et al., 2015; Ayers et al., 2016). Consequently, the people in this region are at high risk of preeclampsia, eclampsia and gestational hypertension from drinking groundwater, and at increased risk of ingesting pathogens from water from the traditional ponds (Kräzlin, 2000; Khan et al., 2014). Stress on the limited reserves of fresh groundwater is expected to rise in the future through a combination of climate change, sea level rise, increased abstraction for irrigation and industry, and population growth (Shameem et al., 2014; Auerbach et al., 2015). To protect and improve the drinking water supply in the coastal region it is therefore important to understanding the present-day spatial variation and formation processes of the groundwater salinity.

Previous studies have found great variation in the groundwater salinity in southwestern Bangladesh (BGS and DHPE, 2001; George, 2013; Worland et al., 2015; Ayers et al., 2016). Several explanations for this large variation have been proposed. One is present-day saline water recharge from the tidal rivers and creeks and the aquaculture ponds that cover much of the region (Rahman et al., 2000; Bahar et al., 2010; Paul et al., 2011; Ayers et al., 2016). Another is freshwater recharge where the clay cover is relatively thin and from rainfed inland water bodies (George, 2013; Worland et al., 2015; Ayers et al., 2016). Finally, some of the variation in the salinity of the groundwater is thought to reflect historical conditions prevailing when the aquifer was buried (George, 2013; Worland et al., 2015; Ayers et al., 2016). Studies in coastal deltas elsewhere, where a higher head due to freshwater infiltration in the higher areas leads to the formation of freshwater lenses, have identified elevation differences as being important factors controlling groundwater salinity (Stuyfzand, 1993; Walraevens et al., 2007; Goes et al., 2009; de Louw et al., 2011; Santos et al. 2012). It has been suggested that both present-day and palaeohydrological processes are important, as deltas are almost never in equilibrium with present-day boundary conditions (Sukhija et al., 1996; Groen et al., 2000; Post and Kooi, 2003; Sivan et al., 2005; Delsman et al., 2014).

It remains unclear how each of the proposed processes influences groundwater salinity variation in southwestern Bangladesh. Previous studies found no spatial autocorrelation in groundwater salinity, presumably because the sampling distances were larger than the expected variation in groundwater salinity (Ayers et al., 2016). In our study, we set out to elucidate the hydrological processes that determine the salinity variation in the groundwater by using high density sampling in a case study area with large variation in land use, surface water bodies and surface elevation. In addition, we aimed to identify geographical or geological factors controlling the dominant salinization and freshening processes and, therefore, the groundwater salinity.

2 Methods

2.1 Methodological approach

Our approach consisted of three steps. First, based on the literature, we reconstructed the geological evolution of southwestern Bangladesh. Second, in the field we carried out high density hydrological and lithological data collection to capture the large expected variation in groundwater salinity. Third, we inferred the evolution of the groundwater salinity by interpreting the field data in the light of the regional geological reconstruction and present-day surface conditions, to determine the dominant processes responsible for the variation in groundwater salinity.

2.2 Fieldwork

To research all the proposed processes, the study area needed to have much variation in land use and surface water bodies, and appreciable variation in surface elevation. We selected an area in southwest Bangladesh by analysing satellite imagery (World Imagery, ESRI, Redlands, CA, USA) to distinguish land use and surface water bodies, Shuttle Radar Topography Mission data (SRTM) (Farr et al., 2007) to analyse elevation patterns and a soil map (FAO, 1959) to ascertain surface geology and geomorphology. The case study area, which is in the Assasuni Upazila (FigureFig. 1), comprises settlements on slightly higher land, surrounded by lower-lying agricultural fields and aquaculture ponds (FigureFig. 1). There are freshwater ponds in the settlements. The soil in the study area is composed of fluvial silts in the higher areas and tidal flat clays in the surrounding lower-lying areas (FAO, 1959). The study was conducted along a crooked 6.3 km long transect oriented approximately north–south (FigureFig. 1) running through several settlements. At the north and south ends are tidal creeks (FigureFig. 1) whose salinity varies seasonally. They are fresh in the monsoon period, but the salinity slowly rises during the dry season and by April and May the creek water contains up to two thirds seawater (Bhuiyan et al., 2012).

In 2017, hydrological and lithological data were collected along the transect at high density (see Sect. 2.2.1) to a depth of 50 m in two field campaigns: one in the dry season (January–February) and one in the wet season (July–August). To do so, groundwater observation wells were constructed, the ground was levelled, and surface and groundwater were sampled. The hydrological data were used to establish (1) the present-day variation in groundwater salinity (2) whether groundwater is affected by freshening or salinization (this entailed analysing the cation exchange) and (3) the source water type recharging the groundwater (for this we analysed the isotopic data). The lithological data were linked with the reconstructed geological history to determine the palaeohydrogeological conditions in the study area from the Last Glacial Maximum (LGM) until the present day.



Legend

- ▲ Groundwater observation well
- DPHE borehole
- Groundwater sample
- Transect with buffer
- Channel
- Channel under tidal influence
- High-lying treed settlements
- Low-lying aquaculture.
- Low-lying agriculture

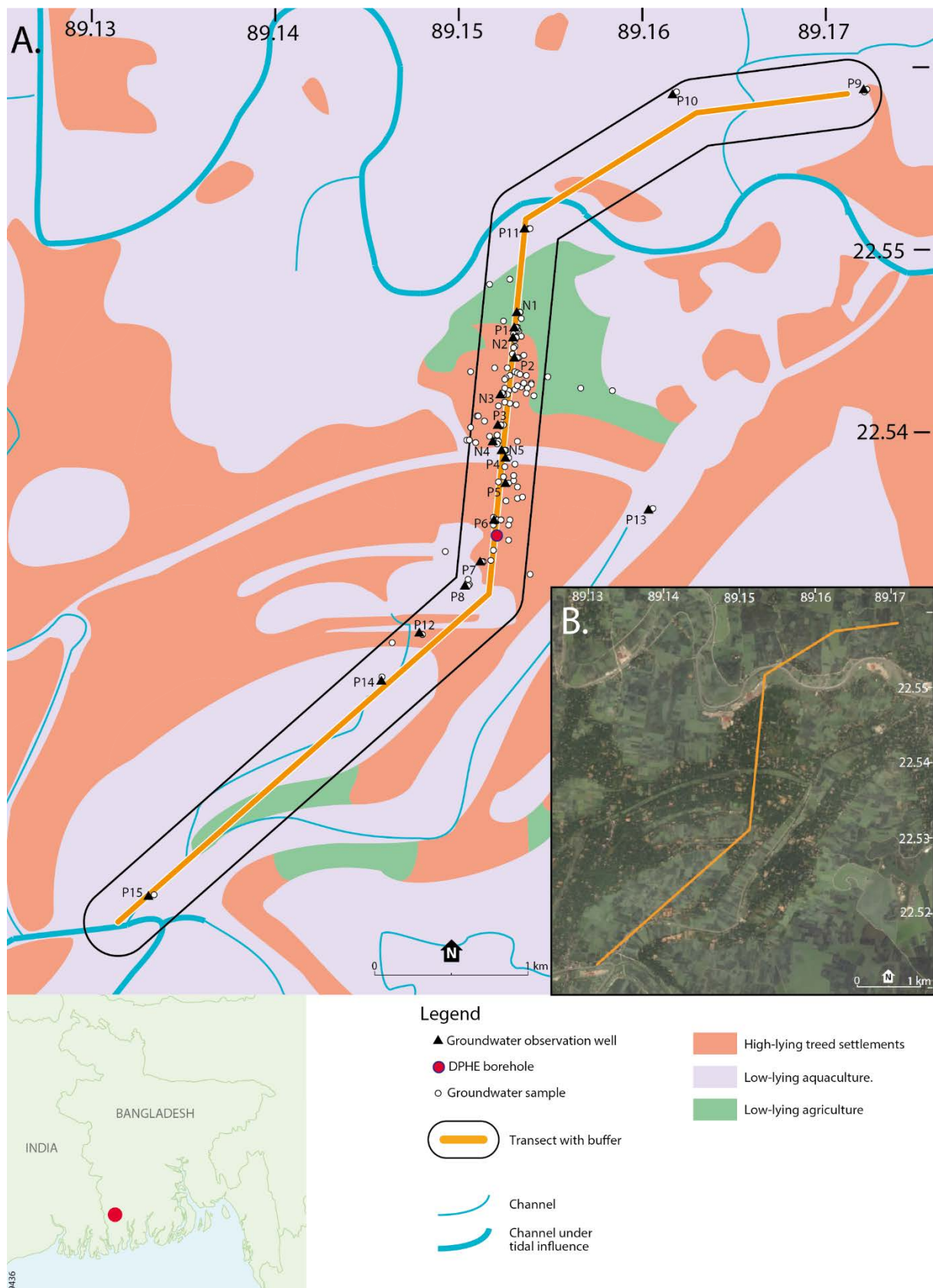


Figure 1. (a) Overview of the study area, indicating the transect and groundwater observation points, land use and current surface water channels. The land use types are based on satellite imagery (b), SRTM data, a soil map (FAO, 1959) and field observations. The location of the Department of Public Health Engineering (DPHE) borehole is indicated in the figure. (b) Satellite imagery of the study area

2.2.1 Lithological drillings and groundwater observation wells

Groundwater observation wells were installed to collect lithological information and groundwater samples. In 2017, 34 tubes with filters at depths of between 6 and 46 m deep were installed at 20 locations. At 14 locations, a single tube was installed, at four other locations, a nest of two tubes was installed, and at the last four locations, a nest of three tubes was installed. Two groundwater observation wells (P16 and P17) were installed a year later [to get more detailed lithological information between P11 and P10](#), but as the sampling campaign had ended, no water samples are available for them.

The first drilling at each location was used to collect the lithological data to approximately 46 m depth (150 feet). The traditional “sludger” or “hand-flapper” method was used as drilling technique (Horneman et al., 2004). The drilling fluid was water from nearby surface water or tube wells, which was pumped out directly after installation by pumping the tube wells for at least 30 minutes or until EC (electrical conductivity) and temperature had stabilized. During the first drilling at each location, the sediment slurry was interpreted in the field every 1.524 m (5 feet). Additional lab analyses were performed on 47 sediment samples from the surface clay layer and at the filter depths. These samples were analysed for their grain size distribution with a Malvern Scirocco 2000, after pre-treatment to remove organic matter and carbonates and after peptizing the mud particles using a peptization fluid and ultrasound. The particles less than 8 μm were classed as clay, those between 8 and 63 μm as silt and those over 63 μm as sand (Konert and Vandenberghe, 1997).

The carbonate and organic matter contents of the samples were quantified by thermogravimetric analysis (TGA) using a Leco TGA-601. The percentage of organic matter was defined as the weight loss percentage between 150–550°C, corrected for structural water loss from clay by a factor of 0.07 times the fraction smaller than 8 μm (van Gaans et al., 2010; Hoogsteen et al., 2015). The carbonate content was determined as the percentage weight loss between 550 °C and 850 °C.

2.2.2 Elevation

Using a Topcon ES series total station (Topcon, Japan), surface elevation at the installed groundwater observation wells was measured relative to a zero benchmark (a concrete slab at nest 1). The elevation data were used to correlate the wells in terms of their water levels as measured at least two days after installation.

2.2.3 Hydrochemistry and isotopes

During two sampling campaigns, water samples were taken and analysed for anions (IC) and cations (ICP-MS); samples were also taken for tritium analysis, as well as for $\delta^2\text{H}$ and $\delta^{18}\text{O}$ analyses. For details, see Table 1. The groundwater samples in the groundwater observation wells was sampled at least a week after installation. The [household](#) tube wells and groundwater observation wells were purged by pumping approximately three times the volume inside the tube. [To sample porewater from the clay, we used the open auger boring method \(De Goffau et al., 2012\). We drilled a hole with a hand auger, without any drilling fluid. Then we waited for the hole to fill with water, which we sampled by inserting a sample bottle into the hole with a stick, which we pulled back out using a rope that was attached to the bottle.](#) EC, temperature and pH were measured directly in the field using a HANNA HI 9829 (Hanna Instruments, USA). Alkalinity was determined by titration within 36 hours of sampling (Hach Company, USA). [The samples were kept out of the sun and cooled as much as possible.](#)

Table 1. Overview of chemical and isotope samplings.

	Samples for IC and ICP-MS (N = 129)	Samples for $\delta^2\text{H}$ and $\delta^{18}\text{O}$ (N = 45)	Samples for tritium (N = 23)
Dry season sampling campaign (January and February 2017)	26 groundwater observation wells, 68 <u>household</u> tube wells, 10 freshwater ponds, 2 aquaculture ponds, 8 <u>hand-open</u> auger borings	-	-
Wet season sampling campaign (July and August 2017)	6 groundwater observation wells, 2 freshwater ponds, 3 aquaculture ponds, 3 <u>hand-open</u> auger borings, 1 inundated field	27 groundwater observation wells, 9 <u>household</u> tube wells, 2 freshwater ponds, 3 aquaculture ponds, 3 <u>hand-open</u> auger boring, 1 inundated field	14 groundwater observation wells, 6 <u>household</u> tube wells, 3 <u>hand-open</u> auger borings

For the IC and ICP-MS analyses, the water samples were stored in a 15 ml polyethylene tube after filtering through a 0.45 μm membrane. Back in the Netherlands, aliquots were transferred to 1.5 ml glass vials with septum caps for IC analysis. For the IC, the aliquots were diluted in accordance with their EC, which was used as an approximation of their salinity. Below 2000 $\mu\text{S}/\text{cm}$ the aliquots were not diluted (1:0), between 2000 and 4000 $\mu\text{S}/\text{cm}$ the aliquots were diluted two times (1:1), between 4000 and 10000 $\mu\text{S}/\text{cm}$ the aliquots were diluted five times (1:4), and above 10000 $\mu\text{S}/\text{cm}$ the aliquots were diluted ten times (1:9). The remaining sample was spiked by adding 100 μl of nitric acid (HNO_3), put on a shaker for 72 hours, and used for ICP-MS. For the ICP-MS, the chloride concentrations were low enough to allow for direct measurement. However, samples 34, 128, 149, 150, 151, 164, 167, 169, 176, 178 showed matrix effects, so were remeasured after diluting five times. The samples for isotope analysis ($\delta^2\text{H}$ and $\delta^{18}\text{O}$) were stored in 15 ml polyethylene tubes and analysed on a Thermo GasBench-II coupled to a Delta-V advantage (Thermo Fisher Scientific, USA). Samples for tritium analysis were stored in polyethylene 1 litre bottles and analysed according to NEN-EN-ISO 9698.

2.3 Calculations and modelling

2.3.1 Variations in groundwater salinity

To study salinity variation the water samples were classified on the basis of chloride concentration into four classes (adjusted from Stuyfzand, 1993): fresh (chloride concentration <150 mg/l) brackish (chloride concentration 150–1000 mg/l), brackish–saline (chloride concentration 1000–2500 mg/l) and saline (chloride

concentration >2500 mg/l). We estimated the chloride values of observation wells P16 and P17 from their EC values: we assigned them the chloride values of samples with similar EC values.

2.3.2 Interpretation of isotopic composition

The stable isotopic composition was interpreted using the mixing line between rainwater and seawater and the Meteoric Water Lines of the two closest meteorological stations with isotopic data: Dhaka and Barisal (respectively 185 km and 125 km from the study area) (IAEA, 2017). For the mixing line between rain and seawater, the weighted average rainwater composition was based on data from the Barisal meteorological station (IAEA, 2017) and the seawater isotopic composition was based on Vienna Standard Mean Ocean Water (VSMOW).

2.3.3 Cation exchange

Evidence of cation exchange was assessed by calculating the amount of enrichment or depletion of the cations compared to conservative mixing for each sample. Chloride was used as an indicator of the degree of conservative mixing. The deviation from conservative mixing for compound i (in meq/l) was calculated using the following formulae, based on Griffioen (2003):

$$iZ = i_{\text{sample}} - i_{\text{conservative}} \quad (1)$$

with:

$$i_{\text{conservative}} = i_{\text{fresh}} + (i_{\text{sea}} - i_{\text{fresh}}) \cdot \frac{(Cl_{\text{sample}} - Cl_{\text{fresh}})}{(Cl_{\text{sea}} - Cl_{\text{fresh}})} \quad (2)$$

where i refers to the concentration in meq/l. Seawater is used for the saline end member i_{sea} . To calculate the Z-values we used values from Ganges water (Sarin et al., 1989) and from pond water (this study) for the freshwater end member i_{fresh} . We assumed that Ganges water has had a large influence on the study area for most of the Holocene and that pond water might have influenced the groundwater recently. Z-values had to be negative or positive for both freshwater end members to be accepted as being truly affected by hydrogeochemical processes. To account for false positive or false negative Z-value due to errors in analysis, the Z-values also had to be larger than the expected error for them to be interpreted as affected by hydrogeochemical processes. Like Griffioen (2003), we assumed the expected error in the amount of exchange was 2.8%, based on a standard error in analysis of 2% and standard propagation of error. The same formula was used to indicate whether sulfatesulphate had been depleted by reduction or enriched by other sources.

2.3.4 PHREEQC simulations of cation exchange

For the interpretation of the hydrogeochemical processes that have occurred in each of the groundwater samples, we needed to take account of site-specific conditions and site-specific hydrochemical processes; to do so, we used the PHREEQC model code (Parkhurst and Appelo, 2013). Possible dissolution or precipitation of minerals was assessed by calculating saturation indices for calcite, dolomite and gypsum, and the partial pressure for CO₂. Additionally, cation exchange during salinization or freshening was simulated, to interpret the stage of salinization or freshening for the samples. For salinization, a scenario was simulated in which seawater diluted 10 times displaces Ganges water (Sarin et al., 1989). For freshening, two scenarios were simulated, because different cation exchange patterns were expected for a) a scenario in which Ganges water displaces 10 times

diluted sea water and b) a scenario in which Ganges water displaces 100 times diluted sea water. The salinities assigned to the saline water end members were based on the salinity levels of mostly less than 1/10th seawater detected in the groundwater.

The Z-values of the samples were compared to the Z-values calculated in the PHREEQC scenarios. Freshening or salinization was determined based on the NaZ value of the samples, with a positive NaZ value indicating freshening and a negative NaZ value indicating salinization. Next, the simulated MgZ patterns in the three PHREEQC scenarios were used to differentiate between the stages of freshening or salinization in the groundwater samples.

The cation exchange processes were simulated using 1D reactive, advective/dispersive transport. The time steps were 1 year and the groundwater velocity was exactly one cell per time step, which makes the Courant number 1. The dispersivity was taken as half of the velocity, which resulted in a Peclet number of 2. The Courant and Peclet numbers were both within the boundaries of a stable model (Steefel & MacQuarrie 1996). The Cation Exchange Capacity (CEC) used in the model was based on the value calculated from the empirical variables for marine soils given by Van der Molen (1958):

$$CEC = 6.8 * A + 20.4 * B \quad (3)$$

where *A* is the percentage of the particles smaller than 8 µm and *B* is the organic matter percentage, based on values for the 26 sand samples taken in our study. Sulfate reduction and methane production were simulated by introducing CH₂O in a zero-order reaction. For the salinizing scenario, CH₂O was introduced at 0.1 millimoles per year. For the freshening scenario, 0.05 mmol per year were introduced. Based on the calculated saturation indices, calcite dissolution was simulated by keeping the calcite saturation index at 0.25 throughout the run, which is representative for marine water (Griffioen 2017).

3 Results

3.1 Regional hydrogeological reconstruction

The relevant Holocene sedimentary history of the researched upper 50 m of the subsurface starts in a landscape determined by conditions under the LGM. During the LGM, the sea level was much lower than it is today, leading to fresh conditions. The freshwater rivers eroded deeply incised valleys down to 120 m below the present-day land surface (BGS & DPHE, 2001; Hoque et al., 2014; Mukherjee et al., 2009). In the interfluvial areas, a Pleistocene palaeosol formed, characterized by oxidized sands (Umitsu, 1993; Burgess, 2010; Hoque et al., 2014). The LGM conditions in the study area are uncertain, as the area is near the edge of a possible palaeo-channel (Hoque et al., 2014; Goodbred et al., 2014), making it possible that the starting conditions for the Holocene sedimentation could be either a Pleistocene incised valley or a palaeosol.

The Holocene sedimentary history in southwestern Bangladesh can be divided into three distinct periods. First, at around 10–11 kyr BP (kilo years before present), a transgressive period started, when the sea level started to rise rapidly (Islam and Tooley, 1999). In combination with an increase in monsoon intensity (Goodbred and Kuehl, 2000b), this marks the start of a period with very rapid sedimentation (Goodbred and Kuehl, 2000a), leading to a transgressive sediment thickness of 20 to 50 m in nearby study sites (Sarkar et al., 2009; Ayers et al., 2016). The maximum inland location of the shoreline was either slightly south or slightly the north of our study area (Goodbred and Kuehl, 2000a; Shamsudduha and Uddin, 2007). During the transgression, sedimentary conditions are expected to have become more under the influence of marine salinity. In the second

period – from 8 kyr BP – sea level rise slowed down, and at ~7 kyr BP the coast started to prograde (Goodbred and Kuehl, 2000a; Sarkar et al., 2009; Goodbred et al., 2014), which probably reduced the influence of marine water. Concomitantly, the monsoon intensity decreased, which caused the sedimentation rate to decline (Goodbred and Kuehl, 2000b; Sarkar et al., 2009). Finally, between 5 kyr and 2.5 kyr BP the Ganges moved eastwards (Allison et al., 2003; Goodbred and Kuehl, 2000a; Goodbred et al., 2003; Goodbred et al., 2014; Morgan and McIntire, 1959; Sarkar et al., 2009). The probable cause of the migration was a topographical gradient resulting from disproportional sedimentation by the Ganges in the west part of the delta (Goodbred et al., 2014), which reduced the supply of sand to the study area, resulting in smaller channels and a larger area of floodplain. In these tidal floodplains, silts and clays were deposited during high water events (Allison et al., 2001), which is why clay overlies all of southwest Bangladesh (BGS and DPHE, 2001; Sarkar et al., 2009; Ayers et al., 2016). Some small late Holocene channels depositing fine sand were still present; sediments from such channels have been found in nearby study areas (Sarkar et al., 2009; Ayers et al., 2016).

Even though the general trend since approximately 7 kyr BP has been progradation, there was a period between 4.5 kyr and 2 kyr BP in which the sea level was higher than it is today (Gupta et al., 1974; Mathur et al., 2004; Sarkar et al., 2009), which might have led to marine deposition at an elevation above present-day sea level (Goodbred et al., 2003; Sarkar et al., 2009).

3.2 Lithology

The collected lithological data reveals a large variation in the thickness and organic matter content of the surface clay layer. In the floodplains in the north at P9, P10, P16 and P17, and in the south at P15, the clay cover is approximately 35 m thick and rich in organic matter (Figure 2, Table 3), whereas around the settlement in the centre of the transect (henceforth referred to as the central settlement) it is 3–10 m thick and less rich in organic matter (Figure 2, Table 3). From the middle of the transect towards the north and south, the clay cover becomes gradually thicker (Figure 2). Under the clay cover is an aquifer composed of grey sands with carbonates, which extends down to the end of all the drillings at 46 m depth (Figure 2, Table 3). This main aquifer contains some small discontinuous organic-matter-rich clay layers (Figure 2, Table 3). Extrapolating from a log of the Department of Public Health Engineering (DPHE) for a borehole located between P6 and P7 (Figure 2), it seems likely that this sand layer extends to 110 m depth and is followed by a clay layer from 110 to 128 m depth and a second aquifer down to a depth of 152 m. At N5, the surface clay layer was succeeded by a gravel bed at 10 m depth.

3.3 Elevation

The villages are at a different elevation than the rest of the study area. Compared with the benchmark, the elevation of the groundwater observation wells in the villages (N2, P2, N3, P3, P5, P6, P7, P11, P12, P15) is between 0.5 to 1.8 m higher, while the elevation in the agricultural fields (N1, P1, N5) and aquaculture ponds (P8, P14) is -0.6 to 0 m (Figure 2). The elevation was not measured at P9, P10, P16 or P17 but data from the SRTM and field observations suggest that these areas are also relatively low.

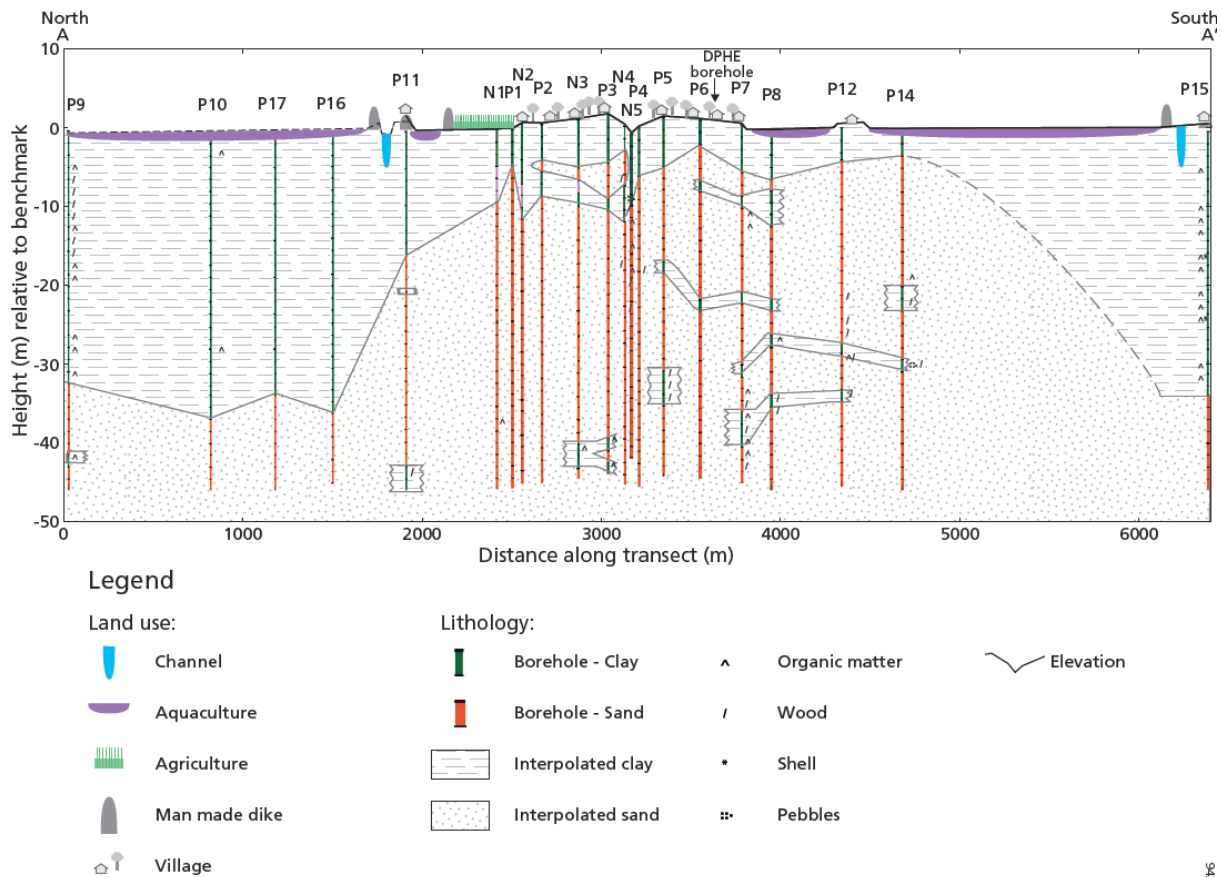


Figure 2. Lithological data from the boreholes drilled in this study, and the interpolated sand and clay layers. A clear difference in thickness of the clay layer is visible between the palaeo floodplains at the north and south sides of the transect, and the palaeo channel in the middle of the transect.

3.4 Salinity

The variation in surface water and groundwater salinity is shown in [FigureFig. 3](#). Surface and groundwater salinity are discussed separately below.

3.4.1 Surface water salinity

The sampled surface water ponds can be divided into fresh and saline ponds. The ponds used for aquaculture contain slightly less than 25% seawater and have a chloride concentration of around 4000 mg/l, and they are saline throughout the year. The rainwater ponds in the settlements on higher land have a chloride concentration below 50 mg/l and are fresh throughout the year. In the wet season, additional surface water bodies are formed, when many of the agricultural fields become flooded by the large amount of rain. The water in the flooded agricultural field near N1 and P1 had a chloride concentration around 200 mg/l in July 2017 ([FigureFig. 3](#)), which may be caused by the dissolution of salts from the saline topsoil, as salt deposits are visible on the surface after the fields dry out again in the post-monsoon period.

3.4.2 Groundwater salinity

The chloride concentrations of the groundwater samples vary between 18 mg/l and 4545 mg/l, which indicates that the most saline groundwater samples contain somewhat less than 25% seawater. The salinity of the

groundwater in the first aquifer correlates well with surface elevation: higher areas are fresher than lower areas (FigureFig. 3). At the slightly higher central settlements, the groundwater is fresh to a depth of approximately 30 m. Below that depth, the groundwater is brackish or brackish-saline. In the lower areas with a thick clayey top layer at the northern and southern ends of the transect, the groundwater is brackish to brackish-saline (FigureFig. 3). This was also the case at P16 and P17, where the EC values were respectively 2.49 and 3.4 mS/cm, which – based on the chloride concentration of samples with similar EC values – corresponds with chloride values between 500 and 1000 mg/l.

In low areas with a thin clay cover, groundwater is saline-brackish under the agricultural areas and saline under the aquaculture ponds. The groundwater salinity difference between these two land use types is substantial: groundwater chloride concentration under the aquaculture ponds exceeds 4000 mg/l, which is more than double the groundwater chloride concentration under the agricultural areas (1000–2000 mg/l). The few samples taken to the side of the transect show the same differences in groundwater salinity between these land use types. The salinity of water samples taken a few metres below the surface from the clay is generally similar to that of the groundwater in the sand aquifer immediately below, except for two clay water samples near the former creek in the middle of the central settlement and one clay water sample from the agricultural fields north of the central settlement (FigureFig. 3).

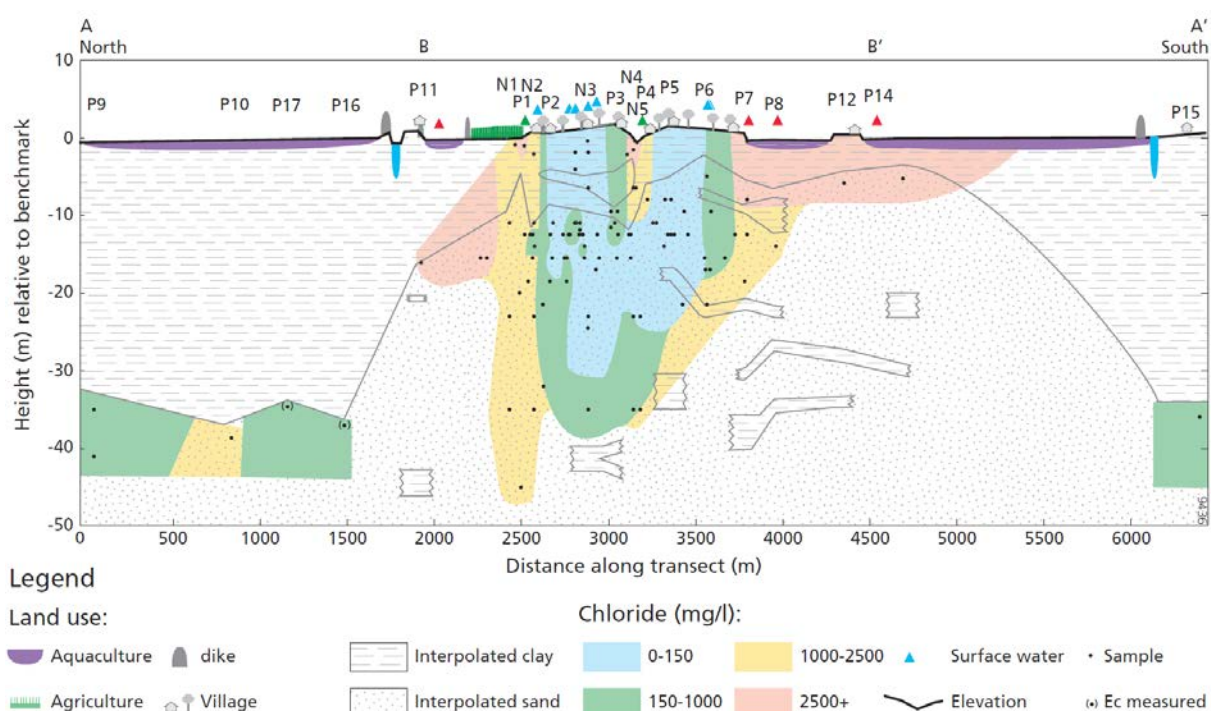


Figure 3. Salinity of the sampled groundwater, of phreatic water sampled in clay just below the surface and of sampled surface water bodies.

3.5 Stable isotopes

The samples were divided into four source water classes (FigureFig. 4). The first class consisted of samples with a relatively light isotopic composition, similar to the weighted average rainwater. This indicates that direct rain infiltration is the dominant source of this water. These samples were taken from surface water bodies in the wet season. The shallow groundwater sample from the clay layer near N3 also falls in this class, revealing that direct infiltration of rain occurs to some extent in the higher-lying area. Lastly, the groundwater at P7 and P8, and at

P9, P10 and P15 falls in this class, but since the samples were taken at great depths, or the isotopic composition of overlying groundwater is different, it is unlikely this groundwater formed under present-day conditions (FigureFig. 5).

The second class contains samples with a relatively heavy isotopic composition skewed to the right of the MWLs (FigureFig. 4). This indicates an effect of evaporation and mixing with seawater. Samples of this class were taken at relatively shallow depths and close to surface water bodies (FigureFig. 5), which suggests that the main source of this water is water infiltrating from stagnant surface water.

The third class comprises samples with a relatively heavy isotopic composition located close to the MWLs in FigureFig. 4. In the study area, this class is visible in five groundwater samples taken just north of the central settlements (FigureFig. 5). The main water source of this class is unclear, but seems to be relatively heavy rain, with limited evaporation or mixing with seawater.

The last and largest group is made up of intermediate weight samples without one clear water source type. These samples could be a mix of different sources of water, such as rainwater, water from surface water bodies and seawater. Some influence of surface water bodies is indicated by the small skew to the right from the MWLs. Samples from this class were collected from both fresh and saline water under the thin clay layer (FigureFig. 5).

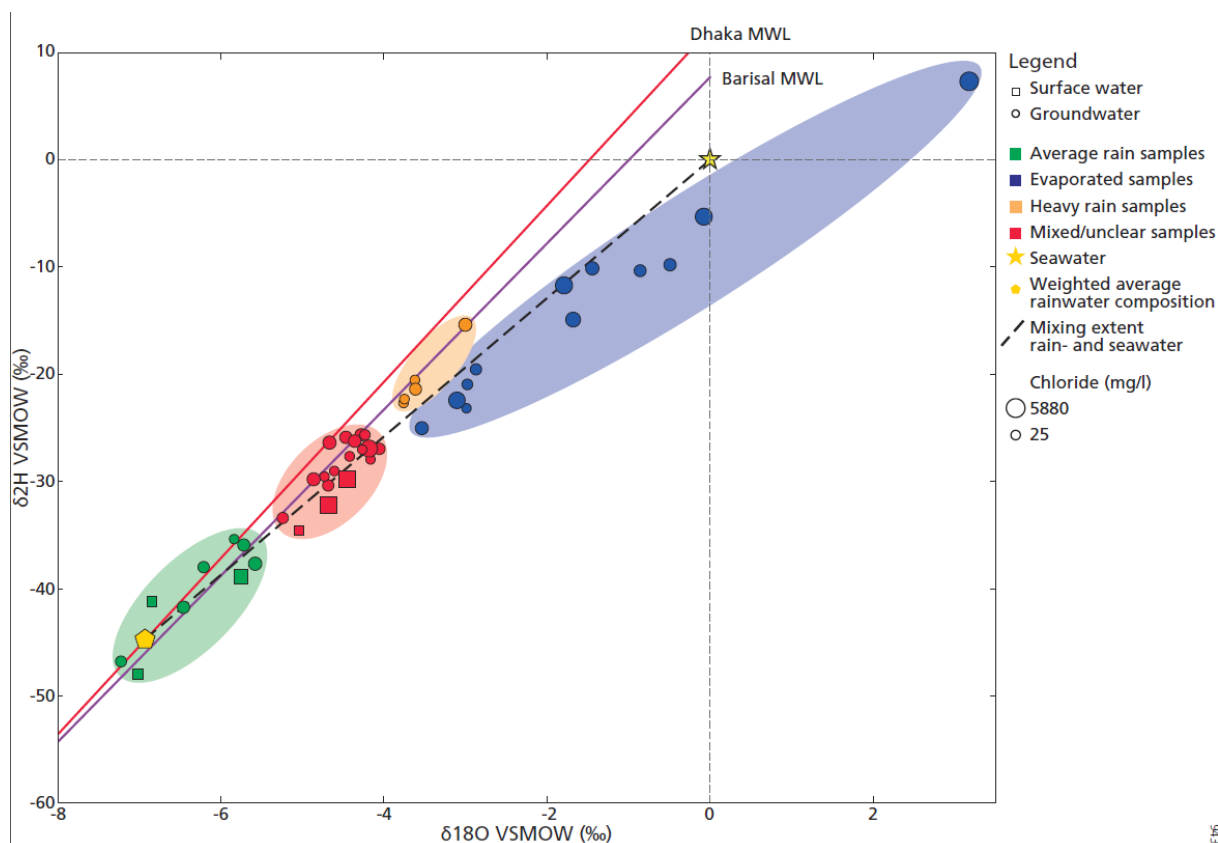


Figure 4. Stable isotope content in the study area. Local Meteoric Water Lines based on monthly samples from the two closest stations Dhaka and Barisal at respectively 185 km and 125 km from the study area (IAEA, 2017). The mixing line between rainwater and seawater is based on the weighted average rainwater composition from Barisal (IAEA, 2017) and seawater VSMOW.

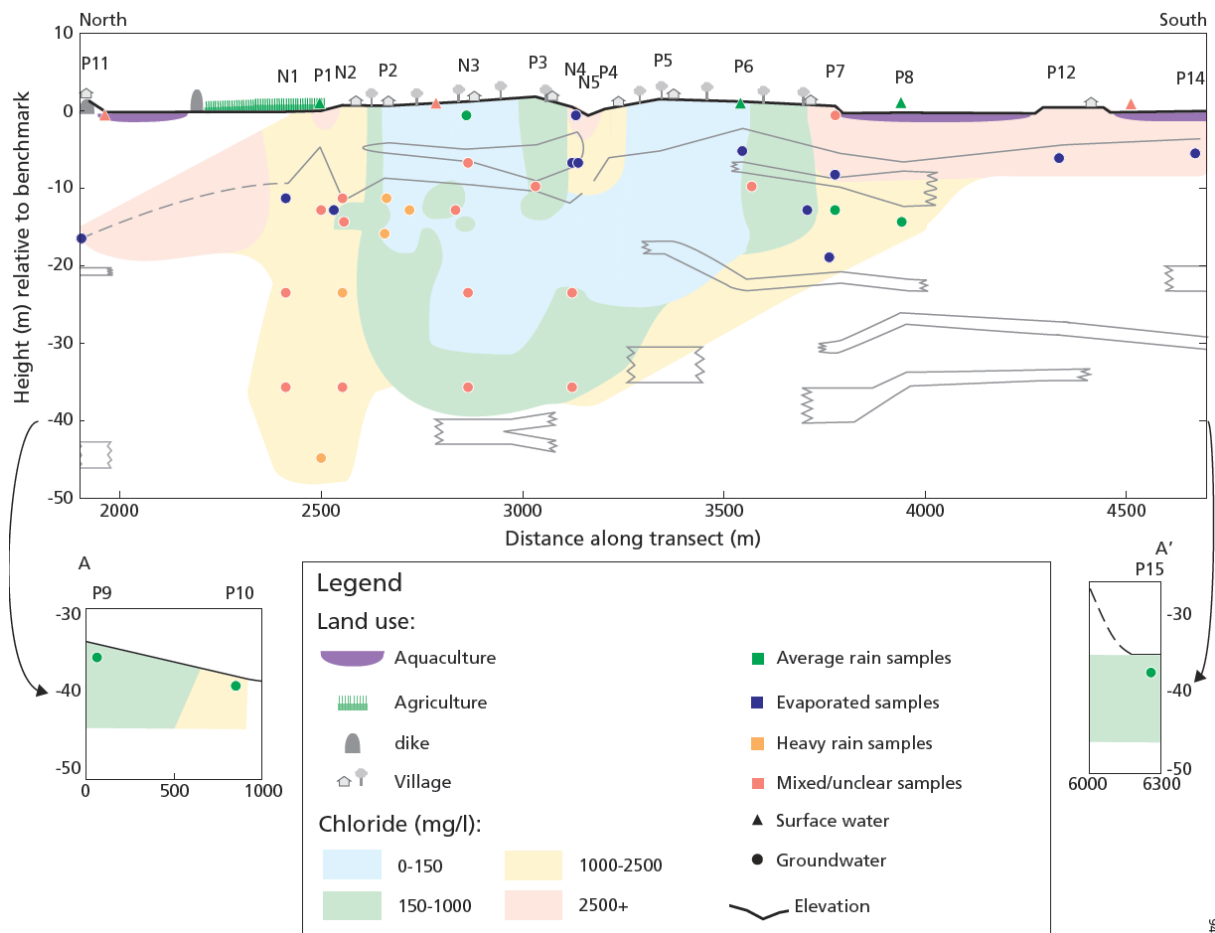


Figure 5. Isotope group of the groundwater sampled in the cross section.

3.6 Tritium

All 23 samples had a tritium concentration below the detection threshold of 1.64 Bq/l, and therefore tritium content could not be used to date the water. Seawater (<0.4 Bq/l), recent rainwater (0.5 Bq/l), or water older than 70 years would all have a tritium concentration below detection threshold, and might therefore be the source of the groundwater (IAEA, 2017).

3.7 Redox conditions and saturation indices

The conditions in the groundwater are reduced, with sulfate reduction and organic matter decay. Sulfate reduction is indicated by depleted sulfate concentrations compared to conservative mixing. Enrichment of SO_4 is observed only in the shallow saline clay near N4 and N5, possibly due to pyrite oxidation. Organic matter decay can be inferred from the partial pressure of CO_2 varying between $10^{-0.4}$ and $10^{-1.6}$ atm in most of the groundwater samples, which is high even for tropical conditions, but not unusual in Holocene coastal regions (Appelo and Postma, 2005; Griffioen et al., 2013).

Most of the groundwater samples from between 10 and 25 m deep are somewhat supersaturated for calcite, with saturation indices between 0 and 0.7, which is common for seawater-derived groundwater in coastal aquifers (Rezaei et al., 2005; Griffioen et al., 2013). Since the sediments contain carbonates (Appendix; Table 3), calcite, aragonite or dolomite is available for dissolution. The samples that were subsaturated for calcite were taken from the clay cover and the shallow aquifer (<10 m deep).

3.8 PHREEQC simulations

The Z-values of the groundwater samples were compared with the patterns of the Z-values during the salinization and freshening scenarios (Figure 6). The patterns of the Z-values were used for the interpretation, as the exact Z-values of the samples were expected to be lower than the Z-values in the model scenarios, since each sample is the result of a specific, less extreme mix of end members. The CaZ, MgZ and NaZ of the groundwater samples were plotted as points in the scenario whose Z-value patterns they best matched, with the X location determined by the chloride concentration matching the values of the chloride in the model scenario. Samples with a chloride concentration exceeding the chloride values in the scenario were plotted at the saline sides of the figures. Six cation exchange (CE) groups were identified (Table 2).

Table 2. Cation exchange groups identified.

Symbol CE group	NaZ value	MgZ value	Description
+	Positive	Positive	Freshening fresh (<200 mg Cl/l)
*	Positive	Negative	Freshening saline (<2000 mg Cl/l)
.	Neutral	Negative, neutral or positive	No cation exchange
-	Negative	Positive	Initial salinization
~	Negative	Neutral	Intermediate salinization
—	Negative	Negative	Late-stage salinization

3.8.1 Freshening

The two freshening scenarios show different patterns for MgZ, because the saline water had a larger percentage of Mg on the cation exchange complex than the fresh water (Appelo et al., 1987; Beekman and Appelo, 1991; Griffioen, 2003). Consequently, the MgZ remained positive during the freshening in the freshwater freshening scenario, while the MgZ became negative during the saline water freshening scenario. We therefore used the MgZ values of the samples to differentiate between freshening freshwater samples (MgZ+) and freshening saline water samples (MgZ-).

Freshening freshwater samples came from multiple locations in the shallow (<20 m deep) fresh groundwater in the central settlement, which indicates that this fresh groundwater is likely formed by water infiltrating from the surface. The freshening detected in the deep brackish samples at P9 and P15 is unlikely to be caused by infiltrating fresh surface water, because of the thick impervious clay layer underlying the saline aquaculture water (Figure 7). Like their isotopic composition, this suggests that this groundwater formed under palaeohydrological conditions.

Freshening saline water was detected in three saline samples taken under the agricultural field north of the central village (Figure 7). The freshening results either from infiltrating surface water, or from fresh water flowing north, as suggested by head data measured in the groundwater observation wells.

3.8.2 Salinization

In the salinization scenario, the MgZ follows a clear sequence. The MgZ value rises initially, then falls until it becomes negative (Figure 6). The samples could therefore be divided into three salinization stages: initial salinization with a positive MgZ (- in Figure 7), intermediate salinization with a neutral MgZ (~ in Figure 7), and late-stage salinization with a negative MgZ (— in Figure 7). When initial, intermediate and late-stage salinizing groundwater is found in sequence, the direction of salinization can be interpreted. Groundwater at the salinization front is in the initial stage, behind it is intermediate salinizing groundwater, and finally late-stage salinizing groundwater is close to the source of the saline water.

Two clear salinization sequences are observed in the study area. One is south of the central settlement, with late-stage salinization close to the aquaculture ponds followed by intermediate salinization and initial salinization towards the fresh groundwater under the central village (Figure 7). This sequence indicates that the aquifer has been salinized from the surface of the lower areas. Another salinization sequence is visible at a former creek near N4 and N5 (Figure 7). Although the clay at the very top is freshening due to recent freshwater recharge, late-stage salinization is visible in the shallow aquifer, followed by intermediate and initial salinization in the brackish and fresh samples at around 12 m depth (Figure 7). This indicates that some of the groundwater has been salinized by water infiltrating from this former creek.

Just north of the central settlement, the salinizing samples lack a clear sequence, but the different salinization stages can still be compared with each other to reveal differences in salinization processes. Under the agricultural fields near N1 and P1 the shallow brackish to fresh-brackish samples display initial salinization, whereas there is late-stage salinization in the samples from depths of 36.5 and 45.7 m. The samples from 36.5 m below the village on higher land again display initial salinization. This suggests that the deeper subsurface under the lower areas has been salinized for longer than the shallower subsurface.

The samples taken below the aquaculture ponds at P11, P12 and P14 display intermediate or late-stage salinization, suggesting that they have been salinized by water infiltrating from the aquaculture ponds above. However, as there are no samples close by with different salinization characteristics, it was not possible to determine the direction of this salinization front.

Some of the fresh and brackish water samples taken in the central settlement also display salinization. They have probably been salinized by some limited local saline water recharge, as the cation exchange characteristic in the fresh samples is sensitive to small changes in salinity. Near N3, the source of this local salinization is probably water infiltrating from the surrounding low areas, as the salinizing samples were taken close to the edge of the higher-lying area (Figure 1).

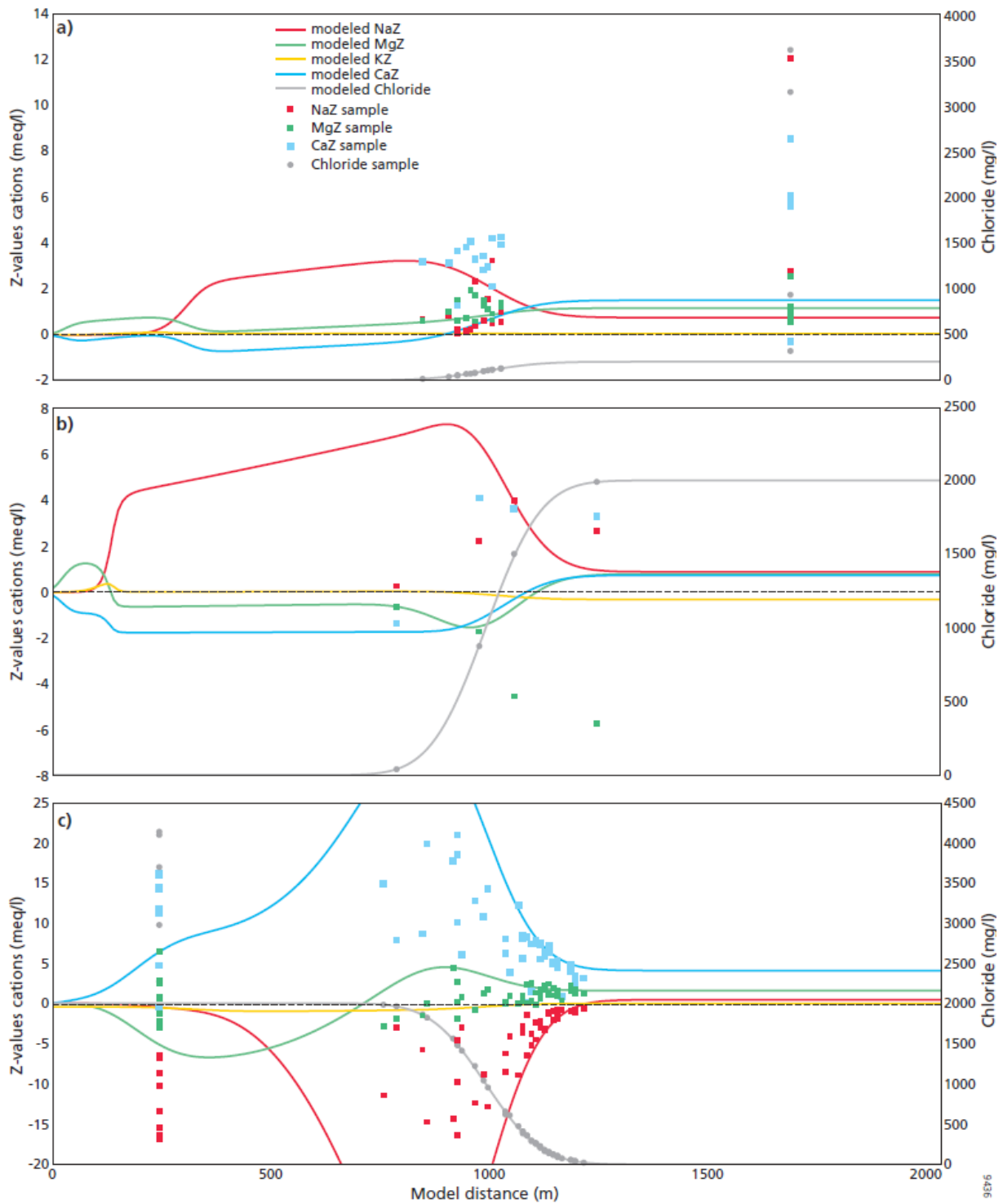


Figure 6. Results of the freshwater freshening scenario (A), the saline water freshening scenario (B) and the salinization scenario (C), together with the samples that match each scenario according to the Z-values of their cations. The X location of the samples is determined by the chloride concentration matching the values of the chloride in the scenario. Samples with a chloride concentration exceeding the chloride values in the scenario are plotted at x= 250 or x=1750.

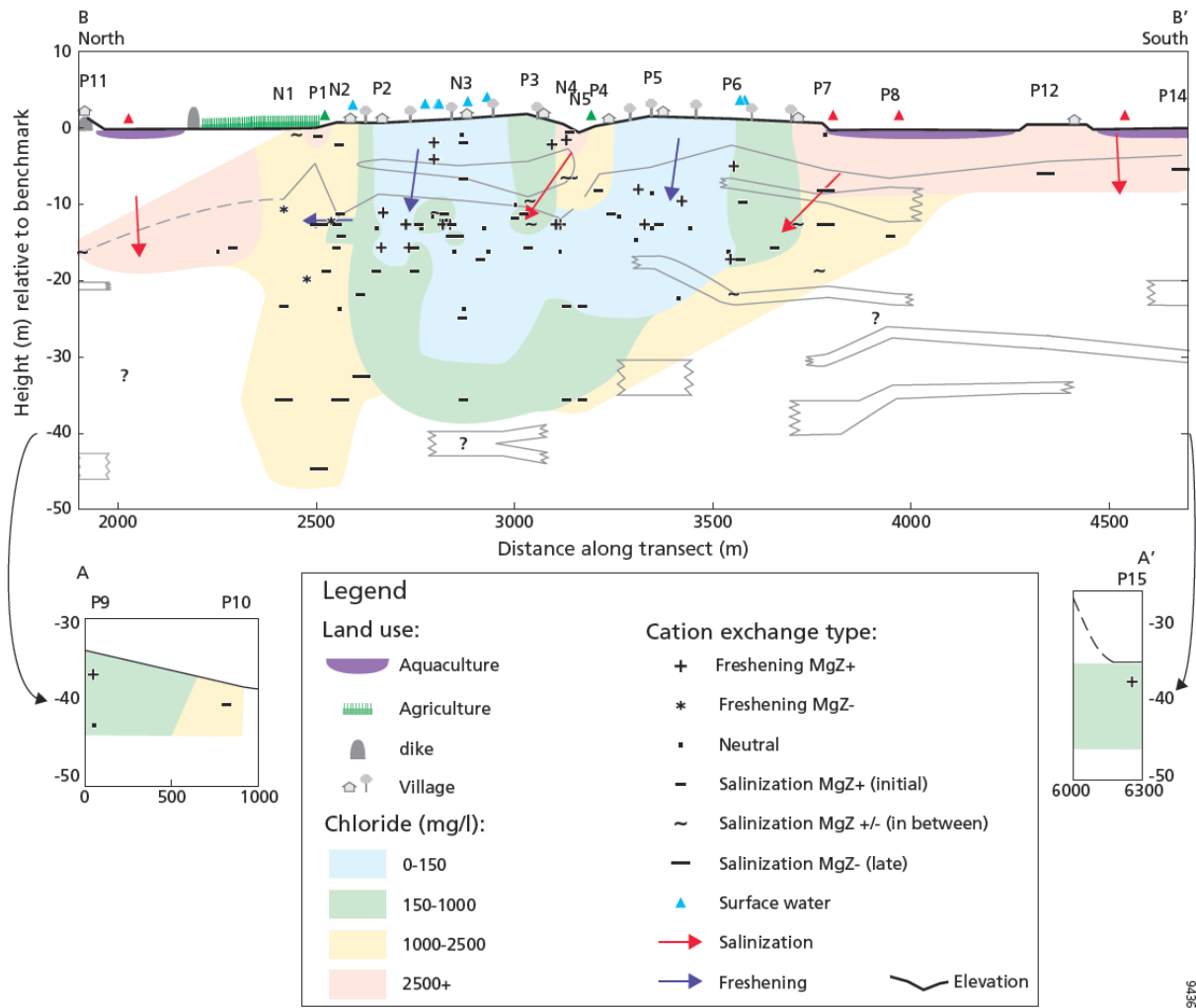


Figure 7. Cation exchange types of the sampled groundwater and the phreatic water from the surface clay layer

4 Discussion

4.1 Inferred hydrogeological evolution of the study area

Combining the field study results with the geological reconstruction based on literature enabled us to postulate a reconstruction of the evolution of the lithology and salinity distribution in the groundwater. This evolution is discussed below, considering the three main Holocene sedimentation phases described earlier plus an additional phase describing present-day processes.

4.1.1 Phase 1: Filling of an incised valley during Holocene transgression (10ka–7ka BP)

The Pleistocene palaeosol was not observed in the study area, which indicates either that a large incised valley was present in the Pleistocene, or that a Holocene channel later truncated the palaeosol (Hoque et al. 2014, Goodbred et al. 2014, Sarkar et al. 2009). This incised valley or truncated Holocene channel filled up rapidly during the transgression, when a rapid sea level rise combined with an increase in monsoon intensity led to accelerated sedimentation of channel sands (Figure 8 (a)) (Islam and Tooley, 1999; Goodbred and Kuehl, 2000a; Goodbred and Kuehl, 2000b; Sarkar et al., 2009; Ayers et al., 2016). Therefore, the deeper part of the first aquifer sands must have been during the transgression.

4.1.2 Phase 2: Emergence of large lithological differences (c. 7 kyr– 5/2.5 kyr BP)

At some point in the early Holocene, channel meandering and associated sand deposition became limited to the middle of the transect (Figure 8(b)). This is indicated by the switch in sedimentary conditions from sand to clay deposition at approximately 30–35 m depth in the lower-lying areas at P9, P10 in the north, and P15 in the south. The unimodal, poorly sorted grain size distribution of this clay indicates that this switch probably occurred during the progradation (Sarkar et al., 2009). Throughout most of the rest of the Holocene, the areas near P9, P10 and P15 remained mangrove-forested tidal delta floodplains, while sand continued to be deposited by the Holocene channel around the present-day central village. The large difference in lithology indicates that the location of the channel was stable throughout the progradation. Possibly, mangrove vegetation in the floodplains prevented the channels from meandering due its ability to capture sediments up to the mean high water level (Furukawa et al., 1996; Auerbach et al., 2015) and to protect land against erosion (Van Santen et al., 2007; Kirwan et al., 2013).

This difference in lithology steered the influence of surface water on groundwater during the rest of the Holocene. The groundwater in the sandy aquifer in the middle of the transect continued to be influenced by the fresh surface water conditions, while the aquifers below the floodplains were much more isolated from surface influences by the thick clay layer. The groundwater under the thick clay layer must therefore be controlled by the hydrological conditions at the time of burial. Consequently, the thickness of the clay is the factor controlling the relative importance of palaeohydrological conditions for present-day groundwater salinity. We assume that during the progradation, the salinity at the surface decreased, as evidenced by the freshening cation exchange observed in the brackish groundwater near P9 and P15 (Figure 3). Additionally, the notion that this groundwater formed under different circumstances than the water close to the present-day central village is reinforced by the different isotopic composition of the groundwater below the thick clay layer compared to the isotopic compositions of the groundwater in the middle of the transect. This process of connate water sealing with subsequent limited influence has also been proposed by George (2013), Worland et al. (2015) and Ayers et al. (2016).

4.1.3 Phase 3: Clay deposition and formation of elevation differences (5/2.5 kyr–present)

a) Clay deposition

After the Ganges migrated eastwards between 5 kyr and 2.5 kyr BP, the areas that contained large sandy Holocene channels during the progradation also started to develop a clay cover (Figure 8(c)) (Goodbred and Kuehl, 2000a; Allison et al., 2003; Goodbred et al., 2003; Sarkar et al., 2009; Goodbred et al., 2014). The salinity during the deposition of the clay cover is not known, as it has been greatly affected by more recent freshening and salinization processes (see phase 4). Overall, however, more brackish conditions are likely to have prevailed from the moment the Ganges migrated eastwards, because the upstream supply of fresh water decreased. Possibly, the deeper groundwater at N1, P1 and N2 has been affected by salinization from this period, since its cation exchange characteristic indicates salinization at a later stage than the shallower samples (Figure 3).

Possibly, starting during the clay deposition in phase 3, salinization of the edges of the aquifer under the thick clay cover has occurred, due to density-driven flow from saline water infiltration in adjacent areas with a thin clay cover (Kooi et al., 2000). In the study area, however, brackish conditions are present at P16 and P17,

indicating that density-driven flow has not affected the groundwater immediately below the thick clay layer. This does not mean that density-driven flow is not relevant in the study area. As salinization from density-driven flow mainly consists of vertical convection cells, the resulting salinization can be expected to occur deeper than immediately below the thick clay cover (Kooi et al., 2000; Smith and Turner, 2001).

b) Formation of elevation differences

While the clay cover was being deposited in phase 3, there was probably little difference in elevation. The observed differences in elevation are thought to have come about after the clay deposition, as a result of differences in autocompaction (Allen, 2000; Bird et al., 2004; Tornquist et al., 2008), which can lead to an inversion of surface elevation (Vlam, 1942; van der Sluijs et al., 1965). The thick, organic-matter-rich clays under the floodplains are likely to have been compacted more than the thinner clay cover on the former sand channels, which would account for the present-day elevation differences of up to 1.5 m between the floodplains and the central former channel area in the village on higher land. These elevation differences are similar to those observed in a comparable delta areas in the Netherlands (Vlam, 1942; van der Sluijs et al., 1965).

The elevation differences in the middle part of the transect cannot be explained by autocompaction, as here only small changes in lithology are observed. Instead, we hypothesize that they may result from erosion by creeks at the edge of the higher areas. Evidence for this is provided by landforms that look like pathways of erosion caused by meandering tidal creeks north of the central settlement, the tidal creek soils in the areas hypothesized to be affected by erosion, and the distribution of tidal creeks and former tidal creeks in lower-lying areas overlain by thin clay (FAO, 1959). Erosion by tidal creeks implies that the clays originally lay above the average tidal river water level. A possible explanation is clay deposition during the higher sea level between 4.5 kyr and 2 kyr BP (Gupta et al. 1974, Mathur et al. 2004, Sarkar et al. 2009). In a subsequent stage, the average water level of the nearby tidal creeks dropped again, resulting in erosive channels.

4.1.4 Phase 4: Emergence of groundwater salinity differences (present-day processes)

a) Higher areas: Freshening by rain and rainfed ponds

The present-day small differences in elevation result in large differences in groundwater salinity, as the surface elevation has determined whether freshening or salinization has occurred in the groundwater. In the higher-lying areas, the conditions at the surface have mostly been fresh since the elevation differences came about, as the slight elevation has prevented saline water flooding from tides and tidal surges. The fresh groundwater is recharged either by direct infiltration of rainwater, and by infiltration of rainwater stored in man-made ponds. Direct rainwater infiltration, which is a common formation process of freshwater lenses in elevated zones within saline areas (Goes et al., 2009; de Louw et al., 2011; Stuyfzand, 1993; Walraevens et al., 2007), is indicated by the light isotopic composition in the phreatic groundwater from the clay at N3.

Infiltration from the rainfed man-made ponds by humans could be a source of fresh groundwater, since they contain fresh water year-round (Harvey et al., 2006; Sengupta et al., 2008). This enables infiltration of fresh water in the dry season when the hydraulic head between the ponds and the groundwater is expected to be larger than in the wet season. The evaporated isotopic composition of the groundwater at P6 suggests such infiltration of pond water (FigureFig. 5). However, the isotopic composition of the deeper fresh groundwater shows only a small amount of evaporation (FigureFig. 4, FigureFig. 5), indicating that infiltration from the freshwater ponds is

not the main process responsible for the fresh groundwater – a conclusion reinforced by the usually very low permeability of the pond bottoms (Sengupta et al., 2008), and the fact that construction of freshwater ponds occurred relatively recently in geological terms (Kräzlin, 2000).

b) Low areas: salinization by marine-influenced water

Unlike the higher areas, the lower areas have often been flooded by tides or tidal surges. Recharge of this saline surface water has been possible in the lower areas with a thin clay cover, where salinizing saline groundwater was observed. This difference between salinization in the lower areas and freshening in the higher areas causes the surface elevation to be the most important factor controlling the salinity of the groundwater in areas with a thin clay cover. Since the salinization originates from the surface, there is already a large difference between higher and lower areas in terms of salinity in the uppermost part of the subsurface, similar to the finding reported by Fernández et al. (2010) for a delta area in Spain. This differs from the situation in Zeeland, the Netherlands, where small fresh groundwater lenses are also present in low-lying areas (Goes et al., 2009; de Louw et al., 2011).

Erosion by tidal creeks also occurred in the middle of the central settlement, as visible at the former creek location near N4, N5 and P4, which has salinized the existing fresh groundwater body, as evidenced by the chloride concentrations (FigureFig. 3) and the sequential salinization patterns (FigureFig. 7) recharging on top of the fresh groundwater.

Recently, aquifers under thin surface clay layers have become salinized by water infiltrating from the overlying aquaculture ponds. This is observed at P11, P7 and P14, where the groundwater has a similar salinity to the overlying aquaculture ponds and an isotopic composition that indicates large evaporation and mixing effects. The salinization by saline aquaculture was detected only in the shallow groundwater underneath saline aquaculture ponds in lower areas with thin surface clay layers. Slightly deeper, the salinization from the shrimp farms was no longer observed; the samples from approximately 12 m deep at P7 and P8 are already much less saline (respectively 1230 and 1370 mg/l), and a totally different isotopic composition (FigureFig. 3, FigureFig. 5). Furthermore, no effect was observed in aquifers under thick clay layers. This rather limited influence of saline aquaculture is not unexpected, as saline aquaculture was only introduced in the study area approximately 30 years ago (Azad et al., 2009). It does indicate that land use has become a controlling factor for the shallow groundwater salinity in areas with thin surface clay layers. In the future, salinization from aquaculture ponds is expected to continue, and hence the extent of salinization to increase. Since the low-lying aquaculture areas with thin surface clay layers are adjacent to the higher areas, continued salinization from the aquaculture ponds could be a threat for the fresh groundwater under the higher area.

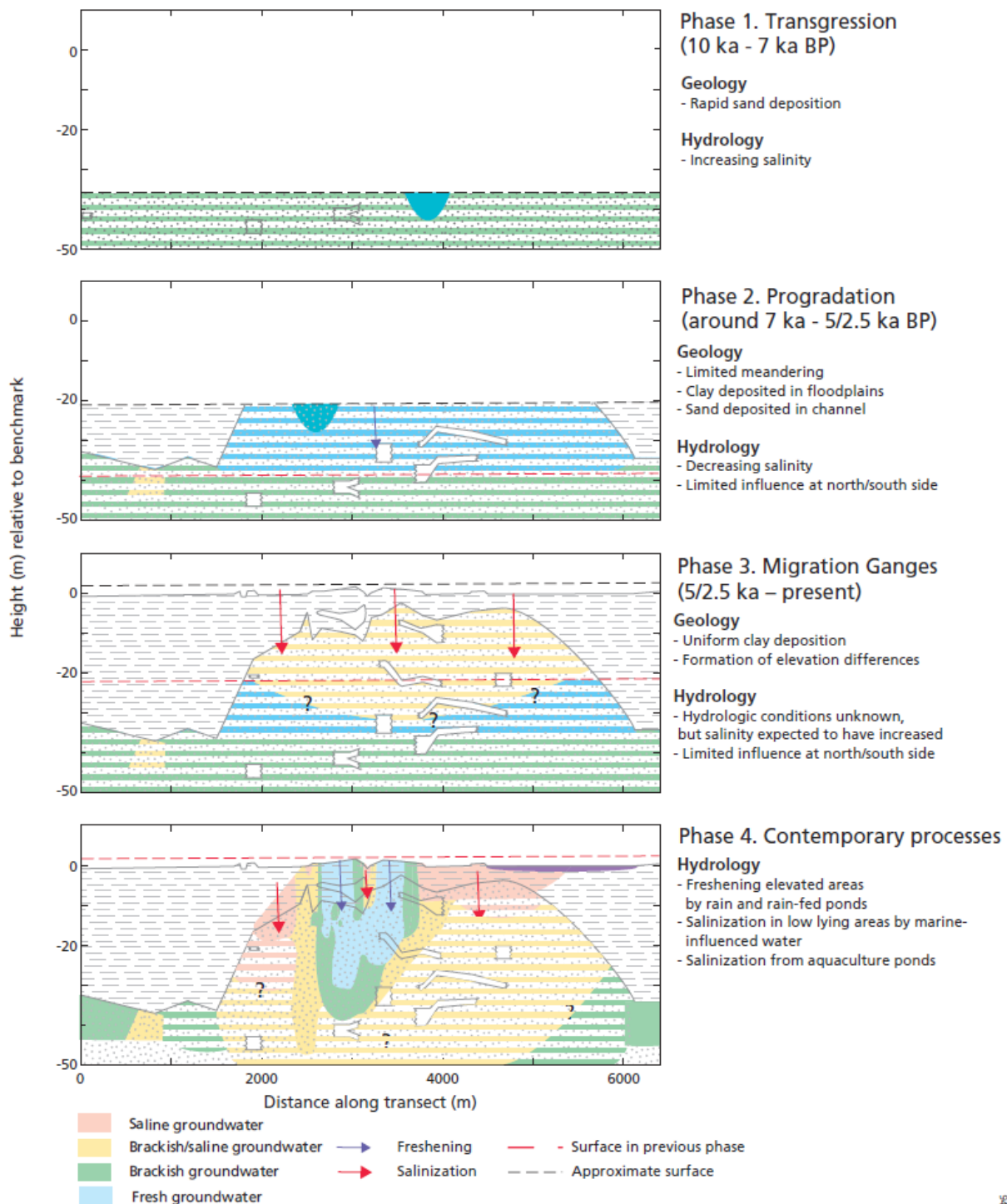


Figure 8. Sediment build-up and associated freshening and salinization processes in the study area.

4.2 Reflection

Above, we derived a hydrogeological evolution of a small area in southwestern Bangladesh, focusing on explaining the large variation in lithology and groundwater salinity which has often been reported in southwestern Bangladesh (BGS and DHPE, 2001; George, 2013; Worland et al., 2015; Ayers et al., 2016). Thanks to the high density of the sampling and the combination of salinity data with isotopic data and PHREEQC-interpreted cation exchange data, it was possible to establish clear patterns in groundwater salinity and to identify relevant hydrological processes and geographical and geological controls. Under the slightly

higher area with a thin surface clay layer, a clear pattern of fresh groundwater was identified, which is attributable to recharge by direct infiltration of rain or via rainfed ponds. The presence of such fresh groundwater lenses in this region was postulated by Worland et al. (2015) and Ayers et al. (2016), and the occurrence of fresh groundwater in elevated areas has been described in other brackish or saline deltas (Stuyfzand, 1993; Walraevens et al., 2007; Goes et al., 2009; de Louw et al., 2011; Santos et al. 2012), but to the best of our knowledge, these phenomena in southwestern Bangladesh have never previously been reported in such detail. The fresh groundwater is bordered by brackish and brackish–saline groundwater at greater depth under the higher area and in the direction of the adjacent lower areas, which probably indicates mixing of fresh groundwater with recharged saline flood waters from the lower areas. Saline groundwater is found only at relatively shallow depths below aquaculture ponds in areas with a thin surface clay layer and is attributed to recharge from these present-day aquaculture ponds. Under thick surface clay layers in the lower areas, brackish water is found; it is postulated to be controlled by palaeo salinity conditions at the time of sealing of the sand aquifer. The importance of palaeo conditions for groundwater salinity in isolated parts of the subsurface in this region has been mentioned by others (George, 2013; Worland et al., 2015; Ayers et al., 2016) and has been described in other coastal zones (Sukhija et al., 1996; Groen et al., 2000; Post and Kooi, 2003; Sivan et al., 2005). We postulate the hydrological processes described above and the resulting observed groundwater salinity variation to be primarily steered by three geological and geographical controlling factors: clay cover thickness, relative elevation and present-day land use.

We acknowledge our study has several limitations, and our interpretations should be seen as a conceptual model to explain the observed spatial patterns of clay and sand deposits and of groundwater salinity. We did not focus on quantifying recharge, discharge and flow rates, or the exact time scales of the hydrological processes. Without age dating, we can't determine the exact moment of salinization or freshening that has occurred. We have been unable to discern comprehensive groundwater flow directions, aside from sketching some indicative flow directions that would account for recharge, as we could not find evidence for locations and patterns of upward groundwater flow and discharge. These upward groundwater flows are expected to be present in convection cells caused by density-driven salinization (Kooi et al., 2000; Smith and Turner, 2001), and discharge is anticipated at drainage points in the landscape which are thought to be present at the edge of higher areas and at the lowest points in the landscape, i.e. the tidal rivers (Tóth, 1963). A possible next step would be to develop a numerical model to further elucidate these flow processes, as well as estimates of recharge and discharge rates and time scales of the described hydrological processes.

Despite these limitations, we contend that the identified controlling factors (clay cover thickness, relative elevation and present-day land use) satisfactorily explain an appreciable part of the observed variation in groundwater salinity variation in the larger southwestern Bangladesh region. Relative elevation and land use data could provide a first estimate of the groundwater salinity in areas with a thin surface clay layer, while knowledge of the palaeohydrogeological conditions seems to be necessary to understand and predict the groundwater salinity in areas with a thick surface clay layer. A next step would be to test the validity of this hypothesis at regional scale.

Data availability. The data used in this study in the appendix and supplementary material, and may be obtained by contacting the corresponding author.

Appendix A

Table 3. The results of the sediment sample analyses. For the red TGA values no clear peak was identified, indicating influence of TGA noise. For the orange TGA values the peak overlapped the border between the organic matter and the carbonate temperatures.

Sample number	Sample location	Depth (m)	Grain size percentage						Thermogravimetric analysis	
			% <8 µm	% 8– 63 µm	% 63– 126 µm	% 126– 252 µm	% 252– 502 µm	% >502 µm	% Organic matter (weight loss 150– 550 °C)	% carbonates (weight loss 550– 850 °C)
1	N1	5	15	57	21	5	2	1	1.51	2.43
2	N1	12	2	11	19	52	14	1	0.49	2.02
3	N1	24	0	7	18	51	23	2	0.45	2.01
4	N1	37	16	18	13	30	19	4	1.34	1.90
5	P1	5	40	56	2	0	0	2	0.40	1.13
6	P1	14	1	9	39	44	6	1	0.33	2.58
7	N2	5	14	63	18	4	1	0	1.26	2.79
8	N2	12	5	18	32	40	5	0	0.57	2.33
9	N2	24	1	10	15	48	19	7	0.41	2.04
10	N2	37	0	10	19	46	20	5	0.55	2.15
11	P2	8	6	28	33	24	6	2	0.76	2.69
12	P2	12	0	9	18	54	16	2	0.56	2.15
13	N3	5	41	56	2	0	0	2	0.29	1.84
14	N3	8	2	12	27	48	10	1	0.58	2.02
15	N3	11	15	49	17	14	4	2	2.81	2.57
16	N3	24	1	8	13	44	29	5	0.30	1.87
17	N3	37	0	6	8	32	40	14	0.30	1.96
18	P3	5	30	63	4	1	0	2	0.51	1.63
19	P3	9	0	9	29	44	14	3	0.56	2.23
20	P3	24	0	11	21	35	25	9	1.05	1.97
21	N4	3	22	68	8	1	0	1	0.98	3.23
22	N4	8	1	9	16	40	27	7	4.86	1.56
23	N4	9	11	46	22	14	6	1	1.75	3.37
24	N4	24	0	9	27	48	13	3	0.39	2.50
25	N4	37	0	6	15	50	25	3	0.32	2.14
26	N5	9	15	43	16	17	7	2	1.86	2.99
27	N5	12	0	2	5	30	55	9	0.41	1.33
28	N5	24	0	6	17	58	17	2	0.36	2.18

29	N5	37	0	6	15	44	26	8	0.39	2.14
30	P4	3	24	67	6	1	1	2	1.18	3.63
31	P4	9	0	10	22	48	17	3	0.46	1.70
32	P5	5	22	65	9	2	1	2	1.18	4.26
33	P5	9	2	16	26	38	16	3	0.36	1.82
34	P6	3	14	55	22	6	2	1	0.42	3.73
35	P6	6	0	5	20	49	21	4	0.44	1.97
36	P6	9	7	23	25	36	8	2	1.48	3.00
37	P6	23	0	7	16	52	24	2	0.31	1.82
38	P6	24	17	55	17	6	3	1	3.07	2.63
39	P7	9	2	25	44	20	7	3	0.83	2.89
40	P7	14	1	11	28	39	16	5	1.55	2.07
41	P8	5	12	53	26	7	2	0	1.56	3.07
42	P8	15	0	7	31	43	16	3	0.54	2.15
43	P8	37	1	5	7	33	49	5	4.65	1.50
44	P9	3	32	61	4	0	0	2	1.94	1.73
45	P9	15	26	65	7	1	1	1	3.03	3.53
46	P9	32	23	65	10	1	1	0	4.86	1.65
47	P9	37	1	13	39	39	6	2	0.54	2.92

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Competing interests. The authors declare that they have no conflict of interest.

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