

1 Dear Editor,

2 We would like to thank you for your effort reviewing our manuscript and your valuable comments
3 which we have taken in full consideration to improve our manuscript.

4 Below we will explain point by point how we dealt with your comment.

5 Beside the changes suggested we improved our paper on a number of other topics. The marked up
6 manuscript version is added after our response below.

7 Our revised manuscript can be found in a separate document. We hope our improvements will
8 satisfy your demands and lead to acceptance of our paper.

9 Sincerely,

10 Bas van de Grift, on behalf of all authors

11

12 **Main comments:**

13

14 1. You need to improve your Fig. 2 for the reader to be able to see the results. I propose to make
15 4 panels numbered also A, B, C and D for use as reference in the main body of text. A should be a
16 new panel including flow and precipitation, and the other three panels should then show your N, P
17 and turbidity data.

18 Agreed and changed accordingly.

19

20 2. You should leave out all discussions from your Results sections - still you have some embedded
21 e.g. p. 25, L. 12-18. Also avoid having references to other sections in your result section - e.g. p.
22 27, l. 18 and 19.

23 We checked the results section and agree with your comment that it contains some embedded
24 discussions. Therefore we removed:

- 25
- 26 • p. 25, L 12-23
 - 27 • p. 25 L32 – p.26 L3
 - 28 • p. 26 L 22-32
 - p. 27 L13-20

29 3. You should reduce the length of your Result sections. Delete the introduction under Results, p.
30 23, l. 11-16 (not needed). Reduce the section 3.2 with 50% - so avoid giving all details on the
31 dynamics - the reader can see for themselves in the Fig. 2A, B, C and D. Please refer to each
32 panel or panels using the letters. Leave out of the manuscript the section 3.6 - is really not needed
33 for the presentation of your results and tests of your hypothesis - instead it is diluting your nice
34 high frequency monitoring results and analysis of these data. You can include general things from
35 these observations in the discussion with references instead - if needed for support.

- 36
- 37 • We deleted the introduction under Results
 - 38 • We reduced section 3.2 considerably
 - 39 • We have moved section 3.6 and Fig. 7 to the supplement of the paper. Findings from these
40 observations are needed for the discussion. As these data were not published elsewhere
41 we cannot refer to this data in the discussion section if we delete it completely. Adding this
section to the supplement is, to our opinion, a good solution. As alternative, we can keep

1 Fig. 7 in the manuscript (but than in the M&M section 2.2) and leave section 3.6 in de
2 supplement without Fig. 7.

3
4 4. I suggest that you include a Table with your analysis of the load of TN, TP and SS - as it is
5 difficult only to follow the results in the text as you show also seasonal results between load
6 calculated from high frequency observations and your normal sampling - give e.g. annual and
7 monthly results in the Table.

8 • We first include a Table with monthly loads but this ended with a lot of numbers. Therefore,
9 we replaced the cumulative load diagrams (Fig. 5) with a figure that gives the monthly
10 loads of the three parameters and added the seasonal and annual load in a small table that
11 can be incorporated to the Figure.

12
13 5. You will need to rewrite the conclusion as it is more like a discussion in its present form - use
14 bullet points for main conclusions and an outlook in the end.

15 We have rewritten the conclusion with bullet points for main conclusions. We struggled a bit with
16 the structure. Now, we have separated this section in bullet points conclusions regarding P, NO₃ and
17 general conclusion. We ended the part with the P conclusions with two policy oriented outlooks.

18
19 6. Please avoid using a sentence such as P. 33, L. 2 would serenely be unfavorable for the transfer
20 model - you should instead acknowldge that the model is weak in explaining the processes that
21 might control N during this period such as biogeochemical processes in surface waters.
22

23 We have rewritten this sentence in:

24 *'Extensive rainfall during the second half of August resulted in a rising of the groundwater level close*
25 *to the tube drain level (Fig. S6) and thus to leaching of NO₃, stored in the soil profile, to the surface*
26 *water. As our TFN model is weak in explaining the processes that might control NO₃ concentration*
27 *such as biogeochemical processes in surface waters we did not include the summer period in our*
28 *model'.*

29
30 Minor comments:

31
32 1. P. 18, L. 15 - during evening or night hours.

33 Agreed.

34 2. P. 18, L. 26 - improve sentence as it is not clear if this is maximum allowed concentrations in
35 discharge waters.

36 We clarified that this refers to measured concentrations. The values are average concentration
37 instead of maximum concentration. This was accidentally wrong in the manuscript.

38 3. P. 19, L. 3 - please insert name/producer of filter used.

39 Done.

40
41 4. P. 23, L. 28 - precipitation amounted to 455 mm,

1 Agreed.

2

3 5. P. 28, Please use comma to separate 1000s through the manuscript and please remember to
4 give units - eg. line 16 - 18,200 kg for TP and 372,500 kg for NO3-N.

5 Done.

6

1 High-frequency monitoring reveals nutrient sources and transport processes
2 in an agriculture-dominated lowland water system

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10 Abstract

11 Many agriculture-dominated lowland water systems worldwide suffer from eutrophication caused
12 by high nutrient loads. Insight in the hydrochemical functioning of embanked polder catchments is
13 highly relevant for improving the water quality in such areas or for reducing export loads to
14 downstream water bodies. This paper introduces new insights in nutrient sources and transport
15 processes in a polder in the Netherlands situated below sea level using high-frequency monitoring
16 technology at the outlet, where the water is pumped into a higher situated lake, combined with a
17 low-frequency water quality monitoring program at six locations within the drainage area. Seasonal
18 trends and short scale temporal dynamics in concentrations indicated that the NO₃ concentration at
19 the pumping station originated from N-loss from agricultural lands. The NO₃ loads appear as losses
20 via tube drains after intensive rainfall events during the winter months due to preferential flow
21 through the cracked clay soil. Transfer function-noise modelling of hourly NO₃ concentrations
22 reveals that a large part of the dynamics in NO₃ concentrations during the winter months can be
23 related to rainfall. The total phosphorus (TP) concentration and turbidity almost doubled during
24 operation of the pumping station which points to resuspension of particulate P from channel bed
25 sediments induced by changes in water flow due to pumping. Rainfall events that caused peaks in
26 NO₃ concentrations did not result in TP concentration peaks. The rainfall induced and NO₃ enriched
27 quick interflow, may also be enriched in TP but retention of TP due to sedimentation of particulate P
28 then results in the absence of rainfall induced TP concentration peaks. Increased TP concentrations
29 associated with run-off events is only observed during a rainfall event at the end of a freeze-thaw
30 cycle. All these observations suggest that the P retention potential of polder water systems is

1 primarily due to the artificial pumping regime that buffers high flows. As the TP concentration is
2 affected by operation of the pumping station, timing of sampling relative to the operating hours of
3 the pumping station should be accounted for when calculating P export loads, determining trends in
4 water quality or when judging water quality status of polder water systems.

5 Keywords

6 Nitrate, Phosphorus, Nutrient retention, High-frequency monitoring, Time series analysis, Lowland
7 water system, Polder, Nutrient dynamics.

8 1 Introduction

9 Many surface water bodies suffer from eutrophication caused by high nutrient loads. Eutrophication
10 of surface waters can lead to turbid waters with decreased oxygen levels (hypoxia), toxin production
11 by algae and bacteria, and fish kills (Bouwman et al., 2013a). Policy makers of national governments,
12 the European Union and other authorities aim to improve water quality in surface water bodies that
13 receive nutrient load from agriculture or other sources like sewage effluent (EC, 2000). A sound
14 assessment of pressures and impacts on the aquatic ecosystem and a reliable assessment of water
15 status in catchments is, therefore, a topic of major importance. If the assessment of pressures is
16 flawed, the action plans will be ill founded and there is a risk that EU member states will not carry
17 out their work where it is most needed and in a cost effective way (EC, 2015). This holds strongly for
18 the Netherlands where nutrient surpluses and leaching are higher than elsewhere in Europe (van
19 Grinsven et al., 2012) and the world (Bouwman et al., 2013b), due to a highly concentrated and
20 productive agricultural sector.

21 For the evaluation of action programs and pilot studies, water authorities invest heavily in the
22 monitoring of NO₃ and P concentrations in surface water. Regional surface water quality networks in
23 EU member states are commonly sampled 12 times a year (Fraters et al., 2005). However, the
24 interpretation of grab sample data in terms of loads and fluxes is often problematic from such
25 monitoring networks (Rozemeijer et al., 2010). Grab sample frequencies are generally not sufficient
26 to capture the dynamical behavior of surface water quality and hydrological functioning of the
27 catchment (Kirchner et al., 2004;Johnes, 2007). It is increasingly recognized that incidental losses
28 and peak flows play an important role in the nutrient loads of surface water systems in the
29 Netherlands (Van der Salm et al., 2012;Regelink et al., 2013) and elsewhere (Withers et al., 2003).
30 Such incidental losses are considered to be related to peak flows after heavy rain storms and due to
31 overland flow or quick interflow via drains and cracked clay soils and related leaching of manure and
32 erosion of soil particles (Kaufmann et al., 2014). Some authors observed a lowering of NO₃

1 concentrations shortly after peak flow (e.g. Poor and McDonnell, 2007; Shrestha et al., 2013) caused
2 by dilution with NO₃-poor precipitation water. Others detected concentration peaks in response
3 events (e.g. Rozemeijer and Broers, 2007; Tiemeyer et al., 2008). Therefore, the NO₃ response to
4 rainfall events depends on the hydrochemical properties of the catchment (Rozemeijer et al., 2010).
5 In addition, the capacity of surface water bodies to retain nutrients is spatially and temporally
6 variable (e.g. Withers and Jarvie, 2008; Cirimo and McDonnell, 1997).

7 As a consequence of the dynamic behavior of nutrient transfer from land to surface water and in-
8 stream processes that impact nutrient retention combined with increasing demands for sound
9 assessments of the water system, there is an increasing interest in continuous or semi-continuous
10 monitoring of water quality at catchment outlets during the last decade (e.g. Bowes et al.,
11 2015; Wade et al., 2012; Jordan et al., 2007; Bieroza et al., 2014; Palmer-Felgate et al.,
12 2008; Rozemeijer et al., 2010; Kirchner et al., 2004; Cassidy and Jordan, 2011; Skeffington et al., 2015).
13 These studies showed catchment dependent non-stationary behavior of the concentration-discharge
14 relationships. High-frequency monitoring has proven to be a powerful tool to improve estimations of
15 annual export loads (e.g. Rozemeijer et al., 2010; Cassidy and Jordan, 2011), nutrients sources
16 (Bowes et al., 2015) and the hydrochemical functioning of a catchment (e.g. Wade et al.,
17 2012; Bieroza et al., 2014; Halliday et al., 2012). High-frequency nutrient monitoring has revealed the
18 presence of diurnal nutrient cycles in rivers and streams caused by biological processes or by P and N
19 inputs from sewage treatment works (e.g. Bowes et al., 2015; Halliday et al., 2012; Neal et al., 2012).
20 Large changes in concentrations or fluxes of materials over relatively short time periods are
21 increasingly recognized as important pathways of nutrient delivery to surface water bodies (Kaushal
22 et al., 2014). In the Netherlands, there is a still debate about the risk of incidental losses associated
23 with manure application (Akkermans and Hermans, 2014). The Netherlands adopted the European
24 Nitrate Directive in 1991 (EC, 1991), which regulates the use of nitrogen in agriculture through
25 national action plans. Among other measures, the regulation includes the period of manure
26 application. To reduce the risk of nutrient leaching to groundwater and surface water, manure
27 application on arable land is allowed from 1 February to 1 August and on grassland from 15 February
28 to 31 August (LNV, 2009). The potential risk for incidental nutrient losses after manure application in
29 February and March (before the start of the growing season) is not known. High-frequency
30 monitoring is a powerful tool to detect such incidental losses.

31 In many low-lying areas worldwide, water levels are managed by inlet of diverted river water in dry
32 periods and discharge via pumping stations in wet periods. Such an embanked land with a human
33 controlled water regime is called a 'polder'. In the Netherlands, these regulated polder catchments

1 cover 60% of the land surface (Van de Ven, 2003). The dense network of subsurface drains, ditches,
2 weirs, channels, pumping stations and the dynamic mixing of water from different sources (seepage,
3 precipitation and water inlet) results in a relatively complex hydrology. Many studies on nutrient
4 dynamics in natural catchments showed a relation between nutrient concentrations and discharge,
5 and this significantly improved the insight in the nutrient sources and pathways in the catchment.
6 The water flow in polders is, however, not a function of free discharge but is controlled by pumping
7 stations. The maximum discharge is controlled by the capacity of the pumping stations. Due to the
8 presence of a dense surface water system, the water storage capacity and the residence time of the
9 surface water in a polder is also higher when compared to natural, free drainage catchments which
10 may impact biogeochemical or hydrological in-stream processes controlling nutrient retention.
11 Insight in the hydrochemical functioning of polder catchments is highly relevant for improving the
12 water quality in the Netherlands.

13 To our knowledge, high-frequency monitoring of surface water quality has not been applied for
14 polder catchments up to now. Discharge-concentration relationships and short scale variation in
15 water quality in polder catchments are still unclear while nutrient sources and pathways are poorly
16 understood (Rozemeijer et al., 2014). High-frequency measurements reveal the short-term
17 variability in solute concentrations which may give valuable insight into the contribution of different
18 sources or different flow routes to the surface water pollution in polders.

19 The general aim of this study is to increase our understanding of the hydrochemical function of an
20 agriculture-dominated water system in a clay polder by analysis of high-frequency monitoring of
21 nutrient concentrations at the polder outlet combined with low-frequency surface water quality
22 data and groundwater quality data from different locations within the polder. The specific objectives
23 of this study are: (1) to increase insight in dynamics of nutrient concentrations and nutrient sources
24 in polder areas (2) to characterize the importance of incidental losses caused by intensive rainfall
25 events whether or not in combination with recent manure application and (3) to assess potential
26 effects of the operational management of the pumping station on the water quality.

27 2 Material and Methods

28 2.1 Study area

29 A continuous monitoring station was established in the Lage Vaart main channel nearby the
30 pumping station Blocq van Kuffeler (A in Fig. 1). This is one of the three pumping stations that
31 control the water level in Lage Afdeling pumped drainage area located within the Flevoland polder,
32 the most recent and at the same time biggest land reclamation project in the Netherland (Groen,

1 1997). The Flevoland polder consists of two pumped drainage areas, which are each drained by a
2 main channel. The Lage Afdeling drainage area drains into the Lage Vaart main channel (Fig. 1). The
3 size of the Lage Afdeling drainage area is 576 km², with altitude ranging between 3 and 5 m below
4 mean sea level. The Lage Afdeling drainage area is mainly rural. The land cover is dominated by
5 agriculture (76%), followed by woodlands and moors (18%) and urban or semi-urban areas (6%).

6 The geohydrology of the Flevoland polder area is generally described by a confining layer of
7 Holocene marine sediments, with a thickness of less than 0.5 m in the northeast to over 7 m
8 southwest, overlying a sandy aquifer deposited in the Pleistocene age. The soils consist for 50% of
9 clay soils, for 39% of silty clay loam and for 11% of sandy soils (Van den Eertwegh, 2002). A typical
10 characteristic of the soils in Flevoland is that the clay layer contains permanent and interconnected
11 cracks due to physical and chemical ripening of the soil after reclamation. The shrinkage cracks
12 disappeared in the plough layer by tillage activities, but are permanently present in the subsoil down
13 to about 1.0-1.5 m below the soil surface (Van den Eertwegh, 2002; Groen, 1997). From a depth of
14 1.2 to 1.5 m below the soil surface, clay deposits, if present, are permanently water saturated and
15 thus not ripened, resulting in a low-permeable soil layer. Due to altitudes below mean sea level and
16 below the water level of the surrounding lakes, there is upward groundwater seepage at most
17 locations within the Lage Afdeling drainage area.

18 The Lage Vaart main channel is connected via a series of secondary channels to a dense network of
19 field ditches and tube drains. Tube drains are generally installed at 0.95 m depth. The horizontal
20 spacing varies between less than 12 to 48 m, mainly dependent on the soil hydraulic conductivity
21 and groundwater seepage rate. The field ditches receive outflow from the tube drain, direct
22 drainage from subsurface flow, regional groundwater seepage and any surface run-off from the
23 connected field area. They drain freely into the secondary channels. The water level in the Lage
24 Afdeling is regulated by 97 weirs and three pumping stations that pump the excess water to the
25 higher situated Markermeer and Ketelmeer. The total pumping capacity is 11-12 mm d⁻¹. The Lage
26 Vaart main channel has a controlled constant water level of 6.2 m below mean sea level. The
27 pumping station Blocq van Kuffeler has four electrically powered pumps. Two pumps with a capacity
28 of 750 m³ min⁻¹ each drain the Lage Afdeling. Operation of the pumping stations with one pump
29 causes a flow velocity in the main channel of approximately 0.125 m sec⁻¹ and with both pumps the
30 flow velocity is approximately 0.25 m sec⁻¹. Up to 2008, the pumping station Blocq van Kuffeler was
31 powered with diesel engines. These diesel engines were replaced with electric engines during the
32 renovation of the pumping station in the autumn of 2008 and this conversion was finished in the
33 beginning of 2009. Since this renovation, the operational management of the pumping station is

1 automatically controlled by a series of water level pressure sensors in the area. The pumps run
2 predominantly during evening ~~enor~~ night hours because of cheaper power supply during these hours.
3 The discharge generated by the pumping stations is measured continuously. The Blocq van Kuffeler
4 pumping station drains the south-western part of the Lage Afdeling drainage area. The flow direction
5 of the water in the channels that are drained by pumping station Blocq van Kuffeler, is illustrated by
6 arrows in Fig. 1. Pumping station B is an emergency pumping station and only operates during
7 extremely wet conditions. Although there is no physical boundary between the area drained by
8 Blocq van Kuffeler and pumping station C, location 5 can be considered as the most upstream
9 location in the Lage Vaart that is drained by the Blocq van Kuffeler pumping station under normal
10 meteorological conditions. There is a sewage treatment plant in the area that discharges its effluent
11 to the Lage Vaart (Fig. 1). The average effluent discharge is $0.35 \text{ m}^3 \text{ min}^{-1}$. The TP and NO₃
12 concentration in the effluent water is measured weakly. ~~The average~~ TP concentration ~~in the~~
13 ~~effluent water~~ is ~~maximal~~ 0.5 mg L^{-1} and the ~~maximal-average~~ NO₃ concentration is 1.5 mg N L^{-1} . The
14 TP load to the Lage Vaart in the period October 2014 – October 2015 equals approximately $5,400 \text{ kg}$
15 and the NO₃ load $16,400 \text{ kg N}$. There are no other sources of sewage discharge to the surface water
16 within the Lage Afdeling drainage area.

17 2.2 Low-frequency monitoring

18 Grab samples were collected every two or four weeks from January 2014 to October 2015 from the
19 polder outlet and 5 other monitoring locations within the part of the Lage Afdeling drainage area
20 that is drained by the Blocq van Kuffeler pumping station (Fig. 1). Four locations are representative
21 for different types of land use (Table 1). Electrical conductivity, oxygen concentration, transparency,
22 temperature and pH of the samples were measured directly in the field. Sub-samples for
23 determination of dissolved substances were filtered through a $0.45 \text{ }\mu\text{m}$ poresize filter (Eijkelpamp).
24 The samples were transported and stored at 4°C . TP, dissolved reactive P (DRP), NO₃, NH₄ and Cl
25 were determined using standard colorimetric methods (APHA-AWWA-WPCF, 1989). Organic-N was
26 extracted by Kjeldahl extraction and measured by colorimetric method and sulphate was measured
27 using IC (Ion Chromatography).

28 The water quality in the Lage Afdeling drainage area showed spatial differences in water quality
29 related to land use (Fig. S1). High NO₃ concentrations were observed in water from the agricultural
30 during winter (location 3 and 4 in Fig. 1). The highest TP concentrations were observed in water from
31 the nature area 'Oostvaardersplassen (location 6 in Fig. 1). The DRP concentration of all sampling
32 locations showed a seasonal variation with higher concentrations during the summer months.

1 2.3 High-frequency measurements

2 Between October 2014 and October 2015 we measured the total-P (TP), total reactive P (TRP) and
3 NO₃ concentration, turbidity, conductivity and water temperature semi-continuously at the polder
4 outlet just before the pumping station. TRP include all P forms that are measured with the
5 molybdenum blue method (Murphy and Riley, 1962) in unfiltered samples, those include acid labile
6 phosphorus containing compounds (inorganic and organic) (Worsfold et al., 2005). The flow regime
7 at the monitoring location is governed almost exclusively by the pumping station. The conductivity
8 was measured continuously with a CTD-diver (Van Essen Instruments, Delft, the Netherlands).

9 The NO₃ concentration was measured using a double wavelength spectrophotometric sensor (DWS),
10 (Nitratax plus sc, Hach Lange GmbH, Düsseldorf, Germany). The DWS measures UV absorbance of
11 dissolved NO₃ at a wavelength of 218 nm at a measuring receiver (EM – element for measuring) and
12 at 228 nm at a reference receiver (ER – element for reference). The recorded measurements at two
13 different wavelengths are designed to compensate interference of organic and/or suspended matter
14 by interpreting the difference between the absorbance values at EM and ER (Huebsch et al., 2015).
15 The Nitratax sensor covers a NO_x-N detection range of 0.1 to 50.0 mg L⁻¹. The NO₃ concentrations
16 were recorded every 5 minutes. There was a small drift in the signal of the Nitratax sensor (max 0.35
17 mg N L⁻¹ per month). We, therefore, corrected the high-frequency NO₃ data using the NO₃
18 concentrations from the biweekly grab samples by calculating a linear drift for the separate
19 maintenance intervals of the sensor.

20 For the total phosphorus (TP) concentration measurements, we installed a Sigmatax sampler and a
21 Phosphax Sigma auto-analyzer (both Hach Lange GmbH, Düsseldorf, Germany). The total-P
22 concentrations were recorded every 20 minutes. The Sigmatax was installed for the automated
23 water sample collection and the pretreatment (ultrasonic homogenization). Next, the sample was
24 delivered to the Phosphax Sigma auto-analyzer. This sample was digested using the sulphuric acid-
25 persulphate method (APHA-AWWA-WPCF, 1989). After mixing and quickly heating and cooling down
26 the sample, the reagents were automatically added and the sample was measured at 880 nm using a
27 LED photometer. The Phosphax Sigma was automatically cleaned and calibrated daily. There was a
28 close agreement between the high-frequency TP data and the TP concentrations of the
29 accompanying two weekly grab samples analyzed by standard laboratory assays ($R^2 = 0.982$) and,
30 therefore, no need to correct the high-frequency TP data (Fig. S21).

31 The turbidity (FTU) was measured using a OBS (optical back scatter) sensor (SOLITAX t-line sc, Hach
32 Lange GmbH Düsseldorf, Germany) that receives the reflected light from the sediment-laden flow.

1 Instead of directly obtaining the suspended sediment concentration, a turbidity sensor measures the
2 turbidity of flow caused by suspended sediment (Gao, 2008). The FTU data was stored with a time
3 interval of 5 minutes. There was a close agreement between the high-frequency turbidity data (FTU)
4 and the suspended sediment (SS) concentrations (mg L^{-1}) of the grab samples ($R^2 = 0.965$) (Fig. S12).
5 The measured turbidity could thus be taken as a proxy for the SS concentration.

6 2.4 Background information

7 Precipitation data on an hourly basis for the Lage Afdeling were abstracted from HydroNet
8 (<http://portal.hydro.net.nl/>). This is an online database with precipitation data based on calibrated
9 radar images. The precipitation of the radar pixels were averaged over the Lage Afdeling drainage
10 area. Temperature data were retrieved from the Royal Dutch Meteorological Institute (KNMI, De Bilt,
11 the Netherlands) weather station Lelystad, located in the center of the Lage Afdeling. The Flevoland
12 polder has a moderate maritime climate with an average annual temperature of 9.9°C , an average
13 annual precipitation of 850 mm and an average of 8 days per year with a maximum temperature
14 below 0°C . Groundwater levels were monitored continuously with pressure sensors in five phreatic
15 groundwater wells located within the agricultural area of the Lage Afdeling (Fig. 1).

16
17 The groundwater quality data set from Griffioen et al. (2013) was used as background information.
18 This database was assembled from the national database of the TNO Geological Survey of the
19 Netherlands and contains complete groundwater analyses down to a depth of about 30 m with
20 sampling dates later than 1945. The groundwater in the Lage Afdeling is characterized as anoxic
21 fresh to saline (Cl between 7 and 4500 mg L^{-1}) and P-rich (TP between 0.01 and 3.6 mg P L^{-1}) with low
22 NO_3 concentrations (between 0 and $7 \text{ mg NO}_3 \text{ L}^{-1}$) (Fig. S23).

23 2.5 Transfer function-noise modelling

24 To increase insight in the driving forces of measured dynamics of nutrient concentrations,
25 preliminary research was done on the application of time series analysis, and more specifically
26 transfer function-noise (TFN) modelling, to estimate the impact of rainfall on NO_3 concentrations.
27 TFN models are very popular for describing dynamic causal relationships between time series and
28 have been widely applied in the field of groundwater modelling (e.g. Berendrecht et al.,
29 2003; Knotters and van Walsum, 1997). Although a small number of studies has used TFN models to
30 relate streamflow data to nutrient concentrations (Schoch et al., 2009; Worrall et al., 2003) or relate
31 precipitation data to high-frequency observation of dissolved organic carbon (Jones et al., 2014), to
32 our knowledge TFN models have not been applied yet on high-frequency monitoring data of

1 nutrients such as available in this study. Therefore, as a first step, we tried to relate the time series
2 of hourly NO₃ concentration measurements to rainfall using the following linear TFN model:

$$3 \quad \log(NO_3) = \theta(B)p_t + \mu + n_t \quad (1)$$

4 and

$$5 \quad n_t = \phi n_{t-1} + \varepsilon_t, \quad (2)$$

6 with p_t the precipitation at time t , $\theta(B) = \theta_0 + \theta_1 B + \dots + \theta_r B^r$ the transfer function (B is
7 backward shift operator, $B^i p_t = p_{t-i}$), μ is the reference or baseline level, n_t a stochastic first-order
8 autoregressive process, ϕ the autoregressive coefficient ($0 < \phi < 1$), and ε_t a zero-mean normally
9 distributed process (Box and Jenkins, 1970). As ε_t is assumed to be normally distributed, the time
10 series of NO₃ data was log-transformed to better satisfy this assumption. For reasons of flexibility
11 and model parsimony, we used a predefined transfer function as described by von Asmuth et al.
12 (2002), which has the form of a Gamma distribution function and has been successfully applied for
13 describing groundwater dynamics:

$$\theta_t = A^* t^{n-1} e^{-at}, \quad A^* = A \frac{a^n}{\Gamma(n)}$$

14 where the parameters A^* , a , n and the stochastic model parameter α are estimated using a log-
15 likelihood function, and $\Gamma(n)$ is the gamma function.

16 2.6 Export loads calculations and trend analysis

17 True NO₃ and TP export loads from the drainage area into the Markermeer were based on our high-
18 frequency concentration measurements and discharge data of the pumping station. In addition NO₃
19 and TP loads were estimated from linear interpolation of the low-frequency grab sample data
20 combined with the discharge data. Although advanced methods have been developed to improve
21 load estimates from low-frequency concentration data, none of the methods clearly outperformed
22 the methods that were based on simple linear or stepwise interpolation (Rozemeijer et al., 2010). To
23 quantify the event-driven TP export load generated by changes in the water flow due to pumping, a
24 hydrograph separation method was used to separate the high-frequency TP concentration data
25 series into short-term TP concentration peaks and baseline TP concentration. In this study we used
26 the same method as applied by Rozemeijer and Broers (2007). This method, originally developed by
27 (Hewlett and Hibbert, 1963), separates the baseline concentration and the peak concentration by a
28 separation line with a constant slope (Fig. S45). This line starts whenever the slope of the
29 concentration series exceeds a specified constant separation slope. The separation line ends when it

1 intersects the falling limb of the concentration series. For this study, a constant separation slope of
2 $0.02 \text{ mg P L}^{-1} \text{ d}^{-1}$ was used. With this relatively low slope value, concentration peaks were also
3 separated from the baseline concentration during situations of upward trends in TP concentrations.

4 Long term TP and NO_3 concentration measurements were available for the polder outlet. We used
5 two frequently applied methods for trend analysis of concentration-time series: (1) seasonal Mann-
6 Kendall tests (Hirsch and Slack, 1984) (2) Theil-Sen robust line (Hirsch et al., 1982) and (3) locally
7 weighted scatterplot smoothing (LOWESS) trend lines (Cleveland, 1979). These methods are
8 relatively insensitive to extreme values and missing data in the time series. The seasonal Mann-
9 Kendall trend test is a robust, non-parametric test on the significance of an upward or downward
10 trend. The Theil-Sen method is a robust non-parametric trend slope estimator. The LOWESS trend
11 lines were used to examine possible changes in trend slopes within the concentration time-series
12 period. We refer to Rozemeijer et al. (2014) for details on the statistical methods.

13 3 Results

14 ~~The results of the high frequency monitoring at the pumping station Blocq van Kuffeler and low-~~
15 ~~frequency monitoring within the Lage Afdeling drainage area will be presented in the next sections.~~
16 ~~First, we shortly describe the water discharge from the polder. Next, the general seasonal trends and~~
17 ~~short time scale dynamics in the high frequency nutrient concentrations will be presented. Finally,~~
18 ~~we present a general description of water quality in the Lage Afdeling based on low frequency~~
19 ~~monitoring.~~

20 3.1 Water discharge

21 The Blocq van Kuffeler pumping station responds rapidly to rainfall events in the drainage area by
22 automatically switching on one or two pumps (Fig. 2A). The interval in which the pumping station is
23 in operation decreased during the autumn months. During the winter months the pumping station
24 runs almost at a daily basis and continuously for several days during very wet periods. There was a
25 strong decline of the daily pumping hours from mid-April to the end of July. A wet period from mid-
26 August to mid-September resulted in an increase of pumping hours. The pumping station pumped
27 almost $67 \times 10^6 \text{ m}^3$ water from the polder into the Markermeer during the period from October 2014
28 until March 2015 and $33 \times 10^6 \text{ m}^3$ during the period from April 2015 until October 2015 (Fig. 3). This
29 corresponds to approximately 350 mm distributed across the entire drainage area for the winter half
30 year (Oct-March) and 170 mm for the summer half year (April-Sept.). The sum of the precipitation
31 ~~was amounted to~~ 455 mm and 517 mm for the winter half year and summer half year, respectively.

1 3.2 Seasonal trends in high-frequency nutrient data

2 ~~The high-frequency NO₃-concentration measured at the Blocq van Kuffeler pumping station ranged~~
3 ~~from 0.01 to 10.4 mg N L⁻¹ and the total phosphorus (TP) concentration ranged from 0.07 to 1.16 mg~~
4 ~~P L⁻¹ (Fig. 2). The NO₃ and TP concentrations from the biweekly grab samples and the accompanying~~
5 ~~one day antecedent precipitation and flow data are shown in Fig. 2 as well. The high-frequency NO₃~~
6 ~~data concentration ranged from 0.01 to 10.4 mg N L⁻¹ and showed~~ a seasonal pattern and a
7 response to rainfall ~~with high concentrations in winter and an increase during wet periods (Fig. 2B).~~
8 The NO₃ concentrations ~~were~~ low ~~at~~ from the start of the monitoring ~~in October 2014 and stayed~~
9 ~~low until~~ until the rainfall event on 15-17 half November. ~~Precipitation events before mid-November~~
10 ~~only had a minor influence on the NO₃ concentration. From mid-November to the third week of~~
11 ~~January, the NO₃ concentration gradually increased from a level of 1 mg N L⁻¹ to a maximum~~
12 ~~concentration of 9 mg N L⁻¹ from mid-November to the third week of January. Major increases of the~~
13 ~~NO₃ concentration occurred during pumping from 18 to 21 November, 16 to 23 December and 13 to~~
14 ~~18 January which showed that the NO₃ concentration responded to rainfall during this period. The~~
15 ~~concentration slightly decreased during dryer periods after these individual wet periods. During the~~
16 ~~dry period in the first three weeks of February, the NO₃ concentration decreased to a level of 1 mg N~~
17 ~~L⁻¹. Next, the concentration reached a maximum of 10.4 mg N L⁻¹ peaked at 24-25 February upon~~
18 ~~rainfall at 24-25 February and gradually decreased towards the end of March where it showed an~~
19 ~~increase again to high concentrations during the first 10 days of April the wet period in late March~~
20 ~~and early April. During April the concentration declined to a level around 0.5 mg N L⁻¹ or below and~~
21 ~~stayed at this low level until mid-August. The NO₃ concentration rapidly increased to approximately~~
22 ~~3 mg N L⁻¹ after a wet period in during mid-August. The concentration peaked with 4.6 mg N L⁻¹ on 2~~
23 ~~September and gradually decreased afterward towards the end of the monitoring period.~~

24 The high-frequency total-P (TP) ~~data concentration ranged from 0.07 to 0.57 1.16 mg P L⁻¹ showed a~~
25 ~~seasonal variation, a response to rainfall (Fig. 2C) and a response to pumping as well. The TP~~
26 concentration was ~~relatively, with concentrations that ranged from 0.25 to 0.4 mg P L⁻¹, high during~~
27 the first three weeks of the monitoring period ~~(0.25 to 0.4 mg P L⁻¹). In October and November, the~~
28 TP concentration decreased during wet periods ~~to a concentration of approximately 0.15-0.2 mg P L⁻¹~~
29 ~~and increased again during the dryer periods to levels around 0.3 to 0.4 mg P L⁻¹. During the wet~~
30 ~~first two weeks of December, the TP concentration decreased to a level around 0.1 mg P L⁻¹. This~~
31 ~~baseline level and remained low at this level until halfway February. During the relatively dry period~~
32 ~~in February and March there was a gradual increase of the TP concentration increased to a level~~
33 ~~around 0.2 mg P L⁻¹ and it remained at this level until mid-June. During the period from mid-June~~

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1 to mid-August the TP concentration gradually increased and peaked ~~with a concentration of 1.2 mg~~
2 ~~P L⁻¹ during a wet period~~ in mid-August. ~~After this wet period in mid-August the TP concentration~~
3 ~~decreased to a level of approximately 0.1-0.2 mg P L⁻¹. Towards the end of the monitoring period the~~
4 ~~TP concentration peaked once more at a level of approximately 0.6 mg P L⁻¹. With higher~~
5 ~~concentrations during the summer season and a decrease during wet periods, the TP concentration~~
6 ~~showed a seasonal variation and a response to rainfall that was opposite to the NO₃ concentration.~~

7 The high-frequency total-reactive P (TRP) data and the dissolved reactive P (DRP) data from the low-
8 frequency monitoring program showed rather high concentrations from the start of the monitoring
9 to early December 2014 and then declined ~~to concentration below 0.1 mg P L⁻¹ to a low level (Fig 2C).~~

10 The TRP and DRP concentration remained ~~at this low level~~ until the second half of May. During the
11 period from mid-May to mid-August the TRP and DRP concentrations followed the trend of the
12 increasing TP concentrations.

13 ~~The baseline level of the suspended sediment (SS) concentration was low during the period~~
14 ~~October to January (Fig 2D). In January the SS concentration increase and it stayed at a relative high~~
15 ~~level to April. During the end of April and May the SS concentration decreased again.~~

16 3.3 Short scale dynamics in high-frequency nutrient data

17 Significant increases of the NO₃ concentration up to 8 mg N L⁻¹ in short time scales appeared during
18 pumping within five days after major rainfall events on 15-18 November, 10-12 December, 19-20
19 December, 7-9 January, 12-14 January, 21-22 February, 29 March-2 April, 14-18 August and 26-31
20 August (Fig. 43 and Table 2). The precipitation during these events ~~peaked was~~ around 20 mm or
21 above. ~~After these NO₃ concentration peaks, the concentration declined during pumping.~~ The
22 increase in NO₃ concentration did not appear after the precipitation events on 20-23 October, 3-4
23 November 17-23 June and 27-29 July. ~~As it will be discussed in section 4, this is likely due to the~~
24 ~~absence of tube drain discharge upon these precipitation events. For the events from mid-November~~
25 ~~to early April, it applies that the response of the NO₃-concentration to rainfall was delayed and~~
26 ~~occurred about five days after the rainfall event. After this NO₃-concentration peak, the~~
27 ~~concentration declined during pumping. The period of five days between rainfall event and peak in~~
28 ~~the NO₃-concentration at the pumping station is representative for the average residence time of~~
29 ~~water in the Lage Afdeling drainage area during wet conditions. Catchment mean residence times~~
30 ~~are much shorter during wet periods compared dry periods (Van der Velde et al., 2012). The five~~
31 ~~days travel time of the water in the field ditches, sub-channels and main channel during wet~~

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1 conditions is in line with model calculated mean annual residence times of water in the Lage Vaart
2 main channel of 6.6 days (Van den Eertwegh, 2002).

3 There is a structural response of the TP concentration and the turbidity on operation of the pumping
4 station. The TP concentration and turbidity always peaked directly after the start of the pumping-
5 engines and decreased again during the period of pumping and afterwards (Fig. 2 and Fig. 34).

6 Pumping events with one pump resulted in an average increase of the TP concentration of 0.06 mg L⁻¹
7 and turbidity of 4.4 FTU while events with two pumps resulted in an average increase of 0.13 mg L⁻¹
8 and turbidity of 22 FTU (Table 3). The TP concentration was on average a factor of 1.30 and 1.83
9 higher during pumping with one pump and two pumps, respectively, compared to the concentration
10 before pumping. The increase of the TP concentration and turbidity during operation of the pumping

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11 station and the larger increase during pumping with two pumps compared to one pump (Table 2)

12 indicates that the increase of the TP concentration is related to resuspension of P from bed

13 sediments due to increased flow velocities. The TRP concentrations also showed an increase in

14 concentration during pumping. As the colorimetric measurement of TRP takes place in an acidic

15 solution it is plausible to attribute the increase of the TRP concentration during pumping to the

16 dissolution of particulate Fe or Ca bound inorganic P. The data shows the largest increase of TP

17 concentrations (0.16-0.60 mg P L⁻¹) during pumping with two pumps after longer periods without

18 pumping (21 Oct., 2 Nov., 8 Dec., 20 Feb., 23 June, 25 July and 15 Aug.) and decreasing TP peaks

19 were observed with subsequent pumping events (Fig. 43). This indicates that during no pumping

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20 conditions, an erodible layer builds up by sedimentation of particulate P. When the water flow

21 velocities in the main channel increase upon pumping, the P becomes suspended and transported

22 downstream. Short-term declines of the TP concentrations to values below the pre-pumping

23 concentration were observed during pumping or shortly after pumping induced by rainfall periods in

24 October, June and August (Fig. 34).

25 A significant short-term change in NO₃ and TP concentrations and the conductivity during a period

26 without pumping that was not linked to pumping appeared during rainfall on 25 and 26 January (Fig.

27 2 and Fig. 34). This period marked the end of a freeze-thaw cycle that started on 20 January. During

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28 this period the top soil became frozen. The precipitation during the night of 24 January consisted of

29 snow and this resulted in a snow cover of a few centimeters. Upon rainfall on the frozen soil on 25

30 January, the decrease in the NO₃ concentration decreased (from 6.1 to 1.5 mg N L⁻¹) and increase in

31 the TP concentration increased (from 0.09 to 0.21 mg P L⁻¹) as observed on 26 January cannot be

32 explained by operation of the pumping station or by antecedent precipitation (5.5 mm on 24 January

33 and 2.1 mm on 25-26 January). Together with the changes in NO₃ and TP concentrations, an

1 ~~increase of~~ the turbidity increased (from 8 to 57 FTU), ~~a decrease in~~ the TRP concentration decrease
2 (from 0.06 to 0.02 mg P L⁻¹) and ~~decrease of~~ the conductivity decreased (from 235 to 122 mS cm⁻¹)
3 (Fig. S43) ~~were observed~~.

4 A cold period with daily average temperatures below 0 °C started at 20 January and ended on 24
5 January (Fig. 3). As a consequence the top soil was frozen, the precipitation during the night of 24
6 January consisted of snow and this resulted in a snow cover of a few centimeters. Soil freeze-thaw
7 processes significantly increase the potential erosion during run-off events that follow thaw in hill
8 slope areas (Ferrick and Gatto, 2005), but also in relatively flat areas (Gentry et al., 2007). Where
9 under normal conditions rainfall infiltrates into the soil, the thaw and precipitation on 25 January
10 likely resulted in run-off. This temporally diluted the NO₃ concentration and conductivity and
11 increased the TP concentration and turbidity. This strongly indicates that the increase of the TP
12 concentration was caused by erosion of soil surface particles. The TRP did not increase during this
13 event, suggesting the TP largely existed of non-labile organic P.

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14 3.4 Decomposition of high-frequency nitrate data

15 As shown in section 3.2, NO₃ concentrations were low from the start of the monitoring period until
16 the rainfall event on 15 November and during April the NO₃ concentrations decreased again.
17 Precipitation events before mid-November and after April only had a minor influence on the NO₃
18 concentration. For the period between 15 November and 30 April a transfer function-noise modelling
19 of hourly NO₃ concentrations reveals that the model can relate quite a large part of the dynamics to
20 rainfall: the coefficient of determination R² = 0.7. The measured time series together with the model
21 simulation and the residual series are shown in Fig. 54.

22 Overall, the transfer model describes slow dynamics well; short-term dynamics cannot be related to
23 rainfall with the transfer model and are described by the stochastic model. The estimated
24 autoregressive coefficient ($\phi = 0.98$) is quite low given the high sampling interval of 1 hour,
25 indicating that most of the temporal structure in the time series has been captured by the transfer
26 model.

27 The results in Fig. 4 show that during no-rain periods the decline in concentration is modelled well.
28 The various periods of rainfall show different results: in December the increase in concentration is
29 modelled well, in January the concentration is overestimated, while in February and March the
30 concentration is underestimated. The overestimation in January can be explained by dilution while
31 recent manure application is a plausible explanation for the underestimation of modelled
32 concentrations in February and March (see section 4). The largest negative residuals appeared

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1 during the thaw event on 26 January (see section 3.3) while the largest positive residuals appeared
2 on 24-25 February.

3 The estimated model parameters and their standard deviation are given in Table S1. The estimated
4 impulse response function for transferring an impulse of 1 mm rainfall into log-NO₃ concentration is
5 given in Fig. S56. The smooth character of the function is due to predefined structure of the function,
6 which is the Gamma distribution function. The time to peak is 5.4 days with a response of 0.033
7 log(mg NO₃-N mg L⁻¹), while 95% of the total response happens within 43 days. The time to peak as
8 revealed by the TFN model matches well with the delay of approximately five days between rainfall
9 events and peak concentrations (Fig. 24).

10 The reference or baseline level follows from the model estimation and has a value of $\mu = -1.13$, or
11 back-transformed from logarithm: $e^{-1.13} = 0.32$ mg N L⁻¹ which means that after a long no-rain
12 period, the NO₃ concentration will decline to 0.32 mg N L⁻¹. The current time series does not include
13 seasonal patterns; during spring and summer season the NO₃ concentration cannot be related to
14 rainfall only. The groundwater levels drop below the tube drain levels (i.e. precipitation may not lead
15 to discharge) and denitrification or in-stream nutrient uptake processes reduce the NO₃
16 concentration, so other driving forces and non-linearity have to be included in the TFN model for
17 modelling the summer season.

18 3.5 Nutrient loads and fluxes at polder outlet

19 ~~Cumulative loads at the polder outlet based on either linear interpolation within the low frequency~~
20 ~~dataset or the high frequency dataset are given in Fig. 5. For TP the cumulative 'baseline' load~~
21 ~~calculated from the high frequency dataset after separation of the pumping event driven short term~~
22 ~~TP peaks are given in Fig. 5 as well.~~ The annual loads based on the high-frequency dataset equaled
23 19,500 kg for TP ~~and~~ 388,500 kg for NO₃-N ~~and~~ 1,788,000 kg for SS which corresponds to 0.98 kg ha⁻¹
24 ~~for TP, 19.4 kg ha⁻¹ for NO₃-N and 89.4 kg ha⁻¹ for SS (Fig. 3).~~ The TP load during the winter months
25 (October – March) was almost equal to the load during the summer months (April – September)
26 while for NO₃ almost 80% of the annual load occurred during the winter months, with January and
27 February as most important months. The annual loads calculated from the ~~low frequency grab~~
28 sample data equaled 18,200 kg for TP and 372,500 kg for NO₃-N. The annual baseline TP load after
29 separation of the TP concentration peaks was 15,400 kg. The difference between the total load and
30 the baseline load equaled 4100 kg, i.e., 21 % of the annual TP load can be attributed to resuspension
31 of TP due to changes in water flow induced by the pumping station.

1 During the period from 1 Oct 2014 to 1 April 2015 the cumulative TP load calculated from the ~~low-~~
2 ~~frequency~~ ~~grab sample~~ data matched the baseline TP load and underestimated the high frequency
3 load with 17%. ~~The months December and January showed the largest difference between the grab~~
4 ~~sample load and the high-frequency load.~~ The ~~winter~~ -low-frequency NO₃ load overestimated the
5 high-frequency load by 6.5%, ~~mainly due to a higher monthly low-frequency load in February.~~ From
6 ~~April-May~~ to mid-August 2015 there was almost no NO₃-~~export~~-load. ~~In August and September the~~
7 ~~grab sample load was lower than the high-frequency load.~~ ~~During the period from April 2015 to~~
8 ~~October 2015 the difference between the baseline load and grab sample load increase.~~ The annual
9 grab sample load underestimated the ~~best available data~~ ~~high-frequency~~ load with ~~64~~%.

10 Time series of TP and NO₃ concentrations in grab samples at the Blocq van Kuffeler pumping-station
11 over the period 2000-2015 are given in Fig. 6. The red lines in Fig. 6 show the LOWESS trend line and
12 the black lines show the Theil-Sen slope over the period 2000-2015. The NO₃ concentration showed
13 no significant upward or downward trend over the period 2000-2015. The time series of TP
14 concentration showed different trends over the period 2000-2015. After a period with minor
15 increase for 2000 to 2009, the LOWESS trend line reveals a decline in TP concentrations in the period
16 2009-2010 followed by an increase from 2011 to 2015. The Theil-Sen slope showed a decline of TP
17 concentration (-0.0053 mg P L⁻¹ per year) over the years 2000-2015. This downward trend was
18 significant according the seasonal Mann-Kendall trend tests.

19 The blue and green lines give the Theil-Sen slopes for the periods 2000-2008 and 2009-2015,
20 respectively, before and after renovation of the pumping station. Where the Theil-Sen slope showed
21 a decline of TP concentration over the years 2000-2015, it showed upward trends of 0.0023 mg P L⁻¹
22 per year and 0.011 mg P L⁻¹ per year over the separate periods 2000-2008 and 2009-2015,
23 respectively. The upward trend for the period 2009-2015 was significant according the seasonal
24 Mann-Kendall trend tests. The NO₃ concentrations showed no significant upward or downward
25 trend over the separate periods 2000-2008 and 2009-2015.

26 ~~3.6 Water quality within Lage Afdeling drainage area~~

27 ~~The low frequency dataset of almost two years with analyses from 6 locations within the Lage~~
28 ~~Afdeling drainage area showed spatial differences in water quality related to land use and~~
29 ~~subsurface characteristics. High chloride concentrations were observed at monitoring locations 1, 3~~
30 ~~and 5, where location 1 and 3 showed higher concentrations during summer than during winter (Fig.~~
31 ~~7). Chloride is an indicator for the contribution of deep groundwater to the surface water. Chloride~~
32 ~~concentrations above 500 mg L⁻¹ were commonly observed in the deeper groundwater in the area~~

1 upstream of location 3 and 5 (Fig. S2). Location 3 shows an inverse relation between the NO_3^- and Cl
2 concentrations ($R^2 = -0.67$) which illustrates the soil and shallow groundwater as source of NO_3^- in the
3 surface water. The Lage Vaart channel acts as a drainage channel for groundwater under the
4 confining Holocene layer, which is often brackish/saline (Van den Eertwegh, 2002). This explains the
5 relatively high Cl concentrations of location 1 during summer.

6 Low NO_3^- concentrations were observed in discharge water from the nature area
7 Oostvaardersplassen (location 6) throughout the year whereas high NO_3^- concentrations were
8 observed in water from the agricultural areas Lepelaartocht and Gruttotocht (location 3 and 4) in the
9 winter (8.3 and 13 mg N L^{-1} in February 2014 and 2015, respectively). The NO_3^- concentration in the
10 urban area water (location 2) did not exceed 2 mg N L^{-1} . The NO_3^- concentrations of the Lage Vaart
11 channel water at the pumping station (location 1) during the winter months were lower compared
12 to the NO_3^- concentrations at the outlet of the agricultural areas. As denitrification is limited during
13 winter time, this indicates dilution of agriculture dominated water with water from nature areas or
14 urban areas. This is confirmed by the SO_4^{2-} data that demonstrate some dilution of the agriculture-
15 dominated water as well. The locations with high SO_4^{2-} concentrations exhibit an inverse pattern with
16 the Cl concentration ($R^2 = -0.45$ for location 3). This shows the occurrence of pyrite oxidation in the
17 shallow subsurface (Griffioen et al., 2013) in the Lage Afdeling drainage area except for location 6
18 that drains the Oostvaarderplassen which has no tube drains and high groundwater levels
19 throughout the year. The N-Kjeldahl concentrations varied between 0.77 and 5.8 mg N L^{-1} but
20 showed little variation over the year for the individual agriculture dominated and urban dominated
21 sampling locations. The N-Kjeldahl concentration in the water from the Gruttotocht (location 3) was
22 almost twice as high as from the Lepelaartocht (location 4).

23 The TP concentration of the low frequency monitoring program varied between 0.05 and 0.72 mg P
24 L^{-1} (Fig. 7). From all sampling locations within the Lage Afdeling, the water from the
25 Oostvaardersplassen (location 6) had the highest TP concentrations. The TP concentration of this
26 water ranged between 0.37 and 0.72 mg P L^{-1} from January to July 2014. The concentration dropped
27 to a level around 0.3 mg P L^{-1} or lower in August 2014 and stayed at this level until April 2015. From
28 April 2015 to mid-September 2015 the TP concentration ranged between 0.35 and 0.74 mg P L^{-1} . The
29 TP concentration at the Oostvaarderplassen and Blocq van Kuffeler were higher during the first
30 months of 2014 compared with the same period in 2015. The long term data series for Blocq van
31 Kuffeler showed high TP concentrations during the first months of 2014 as well compared with
32 concentrations in other recent years (Fig. 6.). We do not have a clear explanation for this
33 observation. The DRP concentrations were low during the first half year of 2014 and 2015. There

1 was an increase of the DRP concentration in July 2014 and July 2015. During the first half year of
2 2014 and 2015 the TP concentration was dominated (> 90 %) by particulate P while in the second
3 half year about 50% of the TP concentration consisted of DRP.

4 The seasonal variation of the DRP concentrations of the Lage Vaart channel water at the pumping
5 station (location 1) followed the trend of the Oostvaardersplassen. Although less pronounced, this
6 seasonal variation applied as well for the agriculture dominated water (location 3 and 4) and the
7 urban water (location 2). The TP concentrations were higher during the summer months than during
8 winter months. The groundwater within the Lage Afdeling drainage area has relatively high dissolved
9 P concentrations (Fig. S2)

10 4 Discussion

11 4.1 Identification of nutrients sources and dynamics in nutrient concentrations

12 ~~The first objective of our study was gaining insight in the dynamics of nutrient concentrations and~~
13 ~~nutrient sources of a typical agriculture dominated lowland water system. We examine the added~~
14 ~~value of TFN modelling of high frequency NO₃ data for identification of NO₃ sources and dynamics~~
15 ~~and, in addition, combining high frequency monitoring data at the polder outlet with low-~~
16 ~~frequency surface water quality data and groundwater data from the drainage area.~~

17 4.1.1 Nitrate

18 ~~Given the low NO₃ concentrations in groundwater (Fig. S2) and the high NO₃ concentrations in the~~
19 ~~surface water at the outlet of the agriculture dominated areas during winter months (Fig. 7), it is~~
20 ~~clear that almost all NO₃ in the surface water at the polder outlet has an agricultural source. The~~
21 ~~high frequency monitoring data at the Blocq van Kuffeler pumping stations additionally provides~~
22 ~~insights in the processes and dynamics of NO₃ delivery to the surface water.~~

23 The high-frequency NO₃ data showed a seasonal trend with ~~a gradually increase from mid-November~~
24 ~~to mid-January higher concentrations during winter compared summer. Low-frequency surface water~~
25 ~~data showed. Given the low NO₃ concentrations in groundwater (Fig. S2) and the high NO₃~~
26 ~~concentrations in the surface water at the outlet of the agriculture-dominated areas within the~~
27 ~~drainage area during winter months (Fig. 7, S1), whereas the groundwater has low NO₃~~
28 ~~concentrations (Fig. S3). From this, it is clear that almost all NO₃ in the surface water at the polder~~
29 ~~outlet has an agricultural source.~~ An increase of NO₃ concentrations from summer to winter is
30 observed in a large majority of agriculture-dominated headwater in The Netherlands (Rozemeijer et
31 al., 2014) and natural catchments elsewhere (Wade et al., 2012). Catchments where NO₃

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1 concentrations are controlled by a combination of effluent loads from sewage treatment works and
2 dilution by rainfall commonly show a decline in NO₃ from summer to winter (Bowes et al.,
3 2015;Wade et al., 2012). The NO₃ pattern is therefore thought to be due to a combination of
4 interflow or shallow draining groundwater with high fertilizer or manure inputs and NO₃ enrichment
5 during autumn and winter. Increased crop uptake of NO₃ during the growing season combined with
6 the effect of in-stream processes result in declined NO₃ concentrations during the summer months.

7 The annual NO₃ load from the WWTP to the Lage Vaart is approximately 4 % of the NO₃ export load
8 at the polder outlet. The low NO₃ concentrations during the summer months and the rapid increase
9 after a very wet period during August additionally indicate that the influence of sewage effluent on
10 the NO₃ concentrations is limited. The discharge from the channel that drains the nature area
11 Oostvaarderplassen (Fig. 1, location 6) enters the Lage Vaart between the WWTP and the pumping
12 station and is 2 to 3 times higher than the discharge from the WWTP. This implies that there is
13 limited flow of the WWTP effluent towards the pumping station during no pumping conditions.

14 Beside the seasonal variation, we structurally observed an increase of NO₃ concentrations after
15 intensive rainfall events during the winter months, except for the rainfall event during the thaw on
16 24-25 January. A reduction in NO₃ concentrations coinciding with periods of intensive rainfall is
17 commonly reported in high-frequency monitoring studies in natural catchments and attributed to
18 dilution of the surface water by surface run-off (Bowes et al., 2015;Rozemeijer et al., 2010). Our
19 structurally observed increase implies that run-off, which dilutes the NO₃ concentration of the
20 surface water does not commonly occur in the polder. Dilution of the NO₃ concentration upon
21 rainfall was only observed during the thaw on 25 January. Soil freeze-thaw processes significantly
22 increase the potential erosion during run-off events that follow thaw in hill slope areas (Ferrick and
23 Gatto, 2005) but also in relatively flat areas (Gentry et al., 2007). Where during normal conditions
24 rainfall infiltrates into the soil, the thaw and precipitation on 25 January likely resulted in run-off.
25 This temporally diluted the NO₃ concentration and conductivity and increased the TP concentration
26 and turbidity.

27 During normal conditions with soil temperatures above 0 °C-it, therefore, indicates that, rainfall
28 initiates a sudden increase of quick interflow via subsurface tube drains, cracks or other macropores
29 to the Lage Vaart channel water which results in leaching for NO₃ stored in the soil profile to the
30 surface water.

31 This is confirmed by the TFN model which showed that quite a large part of the NO₃ dynamics during
32 the winter months can be related to rainfall. The results in Fig. 5 show that during no-rain periods

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1 the decline in concentration is modelled well. The various periods of rainfall show different results:
2 in December the increase in concentration is modelled well, in January the concentration is
3 overestimated, while in February and March the concentration is underestimated. The
4 overestimation in January can be explained by dilution in combination with a decrease of in the NO₃
5 stock stored in the soil profile due to leaching with rain during previous months. The largest positive
6 residuals appeared on 24-26 February. Recent manure application is a plausible explanation for the
7 underestimation of measured concentrations in February and March (see section 3.3). The largest
8 negative residuals appeared during the thaw on 25 and 26 January. The residuals of the TFN model
9 help to get a better understanding of the dynamic NO₃ behavior of the polder catchment.

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10 Drainage water

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11 The tube drain water in the Flevoland polder contains relatively high NO₃ concentrations. Meinardi
12 and Van den Eertwegh (1997) ran a monitoring program on tube drain water composition at 14
13 farms in the Flevoland polder during 1992-1995 and reported concentrations between 5 - 25 mg N L⁻¹
14 ¹. Another monitoring program on nutrient concentration of tube drain water at 6 farms in Flevoland
15 from 2004 to 2008 gave farm-average NO₃ concentrations of 14 - 18 mg N L⁻¹ (van Boekel et al.,
16 2012). These ~~concentrations can~~ concentrations can only explain the observed NO₃ concentration at
17 the pumping station during wet conditions when the tube drain water is athe dominant source
18 contributor of the Lage Vaart channel water. Groundwater levels within the polder are commonly
19 low and tube drainage is rare during the summer and early autumn (Van den Eertwegh, 2002; Groen,
20 1997). In autumn, when evapotranspiration decreases, the groundwater levels rise upon rainfall
21 events to around or above the level of the tube drains, which are present at a depth of 0.95 m below
22 the soil surface, and this initiates drain discharge. This is illustrated by the measured groundwater
23 levels within the Lage Afdeling drainage area (Fig. S67) that shows a direct response of the
24 groundwater level on rainfall combined with a seasonal trend that shows rising groundwater levels
25 during the months October and November and quite stable levels from December to March. Rainfall
26 events between the start of the monitoring and mid-November and between April and mid-August
27 did not result in tube drain discharge. The low NO₃ concentration of the surface water during these
28 periods, are thus, explained by the absence of tube drain discharge. Extensive rainfall during the
29 second half of August resulted in a rising of the groundwater level close to the tube drain level (Fig.
30 S67) and thus to leaching of NO₃, stored in the soil profile, to the surface water. ~~As This is also the~~
31 ~~reason that we started the TFN model on 15 November,~~ our TFN model is weak in explaining the
32 processes that might control NO₃ concentration such as biogeochemical processes in surface waters

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1 This is also the reason that we started the TFN model on 15 November did not include the summer
2 period in our model.

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3 Preferential transport As rainfall is not a driving force for the NO₃ concentration before mid-
4 November and after April, starting the TFN model on 1 October and continuing after 1 May would
5 serenely be unfavorable for the transfer model.

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6 The high-frequency data showed a quick response of the NO₃ concentration at the pumping station
7 to rainfall once the groundwater level is at the tube drain level. This can be explained by the
8 presence of cracked clay soils that results in a rapid response of drainage to rainfall events in winter
9 (Groen, 1997; Van den Eertwegh, 2002). Preferential transport of water and nutrients through cracks
10 and macropores is known to play an important role in heavy clay soils (e.g. Van der Salm et al., 2012),
11 which explains the quick response of NO₃ concentration of the surface water to rainfall events. Due
12 to regular plowing rainwater easily infiltrates into the top soil layer where exchange of NO₃ from
13 manure, fertilizers and plant debris occurs. The top soil or plough layer is commonly well aerated,
14 and therefore, quite optimal for conversion of organic nitrogen and ammonium to NO₃. After
15 leaching of this water from the plough layer to the cracked soil layer it quickly contributes to tube
16 drain discharge. Due to short residence time of this water in the soil, the influence of denitrification
17 on the NO₃ concentration is limited. This implies that the NO₃ concentration at the polder outlet and
18 the related export load from the polder are strongly controlled by quick interflow including tube
19 drain discharge during the winter months.

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20 The period of five days between rainfall event and peak in the NO₃ concentration at the pumping
21 station is representative for the average residence time of water in the Lage Afdeling drainage area
22 during wet conditions. Catchment mean residence times are much shorter during wet periods
23 compared dry periods (Van der Velde et al., 2012). The five days travel time of the water in the field
24 ditches, sub-channels and main channel during wet conditions is in line with model calculated mean
25 annual residence times of water in the Lage Vaart main channel of 6.6 days (Van den Eertwegh,
26 2002).

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27 **4.1.2 Phosphorus**

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28 In contrast to the NO₃ concentration, the TP concentration at the pumping station decreased after
29 the wet periods in the autumn of 2014 and the late summer of 2015 (Fig. 2C). The interflow
30 discharge via subsurface tube drains, cracks or other macropores that resulted in an increase of NO₃
31 concentrations diluted the TP concentrations. Likely this can be attributed to the relative decrease of
32 the groundwater contribution to the channel water during periods of increased interflow discharge.

1 This indicates that the sources of TP in the channel water at the polder outlet can largely be
2 attributed to exfiltration of P-rich groundwater that occurs throughout the year, presumably
3 ~~combined in combination~~ with effluent loads from the WWTP and biogeochemical remobilization of
4 P from channel sediments during the summer and autumn. The low DRP:TP ratio of the surface
5 water within the Lage Afdeling as observed during the first half year of 2014 and 2015 (Fig. ~~S1~~) can
6 be explained by transition of dissolved P to particulate P at the groundwater-surface water interface.
7 This commonly occurs after exfiltration of anaerobic groundwater into surface water due to
8 oxidation processes (e.g. van der Grift et al., 2014; Baken et al., 2015).

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9 The annual TP load from the WWTP to the Lage Vaart is approximately 27 % of the TP export load at
10 the polder outlet. As discussed previously for NO₃, the effect of the WWTP on the NO₃ concentration
11 at the pumping station seems to be small. For TP, however, the WWTP load cannot be neglected.
12 The discharge water from the Oostvaardersplassen has relatively high TP concentrations (Fig. ~~S71~~)
13 and may contribute to the increase in TP concentration at the pumping station during no pumping
14 periods. The source of the TP in the Oostvaardersplassen is groundwater and feces of wildlife. The
15 Oostvaardersplassen is an important wintering area for birds that import nutrients from elsewhere.

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16
17 ~~Additional to this groundwater input signal, the~~ high DRP:TP ratios of the low-frequency monitoring
18 program during the second half year of 2014 and the summer of 2015 indicates that mineralization
19 of organic P from algae or plant debris, or release of DRP from bed sediments can be considered as
20 ~~an additional second~~ P source during summer and autumn when the TP concentration reached a
21 maximum level between 0.8 and 1.2 mg P L⁻¹. Mineralization of organic P mainly occurs after the
22 growing season and the release of DRP from bed sediments is reported during summer and autumn
23 due to temperature and redox dependent biogeochemical remobilization processes for lakes (e.g.
24 Lavoie and Auclair, 2012; Boers and van Hese, 1988), wetlands, fens and floodplain soils (e.g. Zak et
25 al., 2006; Loeb et al., 2008) but also for streams and rivers (e.g. Duan et al., 2012; Jarvie et al., 2008).
26 Low O₂ concentrations in the water column are reported as an indicator for remobilization of P from
27 bed sediments (Geurts et al., 2013). ~~The decline of the O₂ concentrations in t~~he surface water at
28 low-frequency monitoring locations ~~showed a decline of the O₂ concentrations in~~ during the summer
29 and autumn months (Fig. ~~S71~~), thus, indicates that biogeochemical remobilization may occur in the
30 channels of the Lage Afdeling.

31 ~~Surface run-off as a source of P in the surface water was only observed at the end of the freeze-thaw~~
32 ~~cycle on 25-26 January. The TRP did not increase during this event, suggesting the TP largely existed~~

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1 of non-labile organic P. Run-off is generally not an important transport process controlling P dynamic
2 in the polder catchment.

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3 *Resuspension of particulate P*

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4 The increase of the TP concentration and turbidity during operation of the pumping station and the
5 larger increase during pumping with two pumps compared to one pump (Table 3) indicates indicate
6 that the increase of the TP concentration is related to resuspension of P from bed sediments due to
7 increased flow velocities. ~~As a result of resuspension of particulate P from bed sediments due to~~
8 ~~increased flow velocities, we structurally observed an increase of TP concentrations during pumping.~~
9 This indicates that d During no-pumping conditions, an erodible layer builds up by sedimentation of
10 particulate P. When the water flow velocities in the main channel increase upon pumping, the P
11 becomes suspended and transported downstream. The largest increase of the TP concentration
12 during pumping with two pumps after longer periods without pumping and the decreasing TP peaks
13 with subsequent pumping events supports this mechanism.

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14 Resuspension of particulate P retained by sediments during high discharge events is an important
15 transport mechanism in natural catchments (e.g. Evans et al., 2004; Mulholland et al., 1985; Nyenje et
16 al., 2014; Haygarth et al., 2005; Palmer-Felgate et al., 2008). Our data shows that this mechanism is
17 also relevant for P transport in polders where flow velocities vary more abruptly and are maximized
18 by the capacity of the pumping station. The changes in TP concentration during pumping are,
19 however, significantly lower than reported during peak water discharge amongst storms in natural
20 catchments. For an agriculture-dominated lowland catchment in the Netherlands, Rozemeijer et al.
21 (2010) reported a mean increase in TP concentration during discharge from 0.15 to 0.95 mg P L⁻¹
22 coming from 47 rainfall events over a year. Particulate P (PP) increases up to a factor of 100 were
23 reported by Stutter et al. (2008) in response to storm events. Evans et al. (2004) measured PP
24 concentrations up to 3.93 mg P L⁻¹ in a lowland stream during high discharge conditions while the
25 mean concentration equaled 0.1 mg P L⁻¹. Haygarth et al. (2005) reported 10 to 20 times higher
26 mean TP concentrations during storm flow conditions compared to base flow conditions. With data
27 from 76 storms Correll et al. (1999) showed that concentrations of PP increased up to three orders
28 of magnitude during storms.

29 These changes are all considerably larger than the average factor of 1.30 and 1.83 that we observed
30 at the pumping station during pumping with one and two pumps, respectively. The P export from
31 natural catchments during pulses at high flow in less than 10% of the time may amount to about 80%
32 of the annual export (Kaushal et al., 2014). With 143 pumping events during the period from

1 ~~October 2014 to September 2015, discharge-related changes that lead to resuspension of P appear~~
2 ~~more frequent in this polder catchment compared to natural catchments. As only~~~~For our polder~~
3 ~~catchment we calculated that only~~ 21% of the annual TP export load ~~can be related to resuspension~~
4 ~~this cannot be considered as the dominant P transport mechanism of TP due to changes in water~~
5 ~~flow induced by the pumping station. With 143 pumping events during the period from October~~
6 ~~2014 to October 2015, discharge-related changes that lead to resuspension of P appear more~~
7 ~~frequent in polders compared to natural catchments. The artificial pumping regime that buffers high~~
8 ~~flows in polder area thus results in a high potential of polder areas to retain TP concentrations that~~
9 ~~increase during dry periods in the summer and autumn by sedimentation of PP. Consequently, this,~~
10 ~~likely may as a result of~~ in a higher potential of polder areas for ~~DRP release from the bed sediments~~
11 ~~during summer months by biogeochemical remobilization which attributes to TP export loads during~~
12 ~~the summer period additionally contributes the TP export loads.~~ Therefore, it can be concluded that
13 ~~total P export loads from transport mechanisms in~~ polder catchment can be characterized as less
14 incidental ~~and and less peak flow controlled peak flow controlled and more controlled by~~
15 biogeochemical remobilization from bed sediments than those from natural catchments.

17 4.2 Incidental nutrient losses to surface water after manure application

18 ~~The second objective of our study was to determine the relevance of incidental nutrient losses~~
19 ~~caused by intensive rainfall events in combination with recent manure application.~~

20 The NO₃ concentration peaked at the polder outlet on 24 February, four days after an intensive
21 rainfall event that marked the end of a relative dry period that started early February. The increase
22 of the NO₃ concentration is almost two times higher compared to the other peaks in NO₃
23 concentration after a rainfall event (Table 2). This suggests that the NO₃ peak of 10.4 mg N L⁻¹ was
24 caused by an incidental loss after manure application that started on 1 February. The TFN model
25 revealed high residual NO₃ concentrations up to almost 8 mg N L⁻¹ during this NO₃ peak that cannot
26 be explained by rainfall (Fig. 45). The NO₃ concentration peaks on 27 February and 3 March also
27 showed large positive residuals of 4.2 and 3.4 mg N L⁻¹, respectively. The wet period in January
28 resulted, however, in predicted NO₃ concentrations that were higher than the measured
29 concentrations. The negative residuals in January can be explained by leaching of the NO₃ stored in
30 the soil profile during the winter season in combination with the appearance of some degree of
31 dilution of the remaining NO₃ by precipitation water during this period. Dilution of the NO₃
32 concentration upon rainfall events commonly observed in catchments (e.g. Rozemeijer et al.,

1 2010;Wade et al., 2012). A plausible explanation for the large positive residuals in February and
2 March is recent manure application that started on 1 February and temporary soil storage of applied
3 N during the first dry weeks of February.

4 The TP concentration peaked on 21 February during the beginning of the rainfall event,
5 simultaneously with a turbidity peak after the start-up of the pumps following upon a relatively dry
6 period of more than one week without pumping (Fig. 34). It is, therefore, not likely that this peak
7 was caused by an incidental loss after manure application but caused by hydrodynamic resuspension
8 of the Lage Vaart bed sediment. The absence of a TP peak after the rainfall event on 21-22 February
9 can be attributed to the soil characteristics of the area. We already discussed that the water quality
10 at the polder outlet is strongly controlled by quick interflow via tube drains or cracks and that
11 surface run-off only influenced the water quality when it rained during the end of a freeze-thaw
12 cycle. Although it is known that tube drain discharge after rainfall events in combination with recent
13 manure application on cracked clay soils may contain significant TP concentrations (Van der Salm et
14 al., 2012), these peaks did not appear at the polder outlet. Several other studies ask attention for
15 elevated TP concentrations in drain and trench flow within a few weeks after application of
16 fertilizers or liquid farm manure (Hodgkinson et al., 2002;Simard et al., 2000;Djodjic et al., 2000). It is
17 unknown if these peaks appear after rainfall events in the tube drain discharge or in the receiving
18 field ditches in the Lage Afdeling drainage area. Therefore, it is unclear if the absence of TP peaks
19 simultaneously with the NO₃ peaks at the polder outlet can be attributed to sedimentation of PP
20 from agricultural sources in the field ditches or sub-channels where it may become a source for DRP
21 release from bed sediments during the summer and autumn months or that there is almost no
22 particulate or dissolved P leaching from the top-soil to the surface water due to the sorption
23 capacity of the top-soil. From other [lowland](#) areas it is known that the dissolved P loads to surface
24 water from tube drains and shallow groundwater discharge are low due to precipitation with Fe
25 hydroxides with a high affinity to retain P, at the oxic/anoxic interface around the tube drains and
26 ditch sediment (van der Grift et al., 2014;Baken et al., 2015).

27 4.3 Water quality affected by the operational management of the pumping station

28 ~~The third objective of our study was to assess the potential effects of the operational management~~
29 ~~of the pumping station on the water quality.~~ Since the renovation of the pumping station in the
30 autumn of 2008, it runs typically overnight during normal meteorological conditions, as reason of
31 cheaper power supply. The low-frequency sampling is always performed during daytime. The
32 distribution of pumping hours and sampling moments over the day during the period October 2014
33 – September 2015 and boxplots of measured TP concentrations over the day during the months

1 | January and February 2015 are shown in Fig. S78. These two months were selected because boxplots
2 | for longer time series are dominated by the seasonal trends in the TP concentration. The median,
3 | quartile and maximum TP concentrations were higher during night hours than during daytime. As a
4 | result, the monitoring program systematically misses the TP peak that occurs during pumping and
5 | consequently does not measure diurnal cycles in water quality caused by the pumping station. The
6 | reported time series from the low-frequency sampling program is, thus, not fully representative for
7 | the TP concentration at the polder outlet. As a consequence, export fluxes from the polder as
8 | calculated from low-frequency sample data underestimate the true export P-loads (Fig. 53). The NO₃
9 | concentration showed no structural response on pumping, further illustrating the importance of
10 | resuspension of P by pumping.

11 | The preferred timing of sampling during regular working-hours is also critical for trend detection in
12 | the resulted dataset time series (Fig. 6). Trend analysis before and after replacement of the diesel
13 | engines compared with trend analysis over the years 2000-2015 indicates that the trend of slightly
14 | decreasing concentrations over the years 2000-2015 may be caused by the sudden decrease of
15 | concentrations after renovation of the pumping station which is an artifact of a change in pumping
16 | regimes.

17 | The number of diesel powered pumping stations in the Netherlands has rapidly declined during the
18 | last decades. There were around 200 diesel or hybrid (diesel + electric) powered pumping stations in
19 | operation in 1990. Currently, there are only 40 remaining and these pumping stations have mainly a
20 | function for emergency situations (Gemalen, 2015). During the same period, electric powered
21 | pumping stations have been equipped with automatic switching systems. Nowadays, a large
22 | majority of pumping stations operates predominantly during night hours. As the pumping station is
23 | the outlet of a (artificial) water system it is often a monitoring location for surface water quality as
24 | well. The renovation of pumping stations may thus have had a substantial impact on reported trends
25 | in water quality on a regional or even a national scale.

26 | 5 Conclusions

27 | High-frequency monitoring at the outlet of an agriculture-dominated lowland water system
28 | combined with low-frequency monitoring at several other locations in the polder appears to be an
29 | effective tool to reveal difficult to notice responses in surface water quality. P and NO₃ react
30 | differently. Conclusions regarding P are the following:

- The P retention potential of the polder water systems is enhanced compared to natural catchments due to the artificial pumping regime that prevents high discharge flow and therefore limits resuspension of particulate P.
- Groundwater seepage, biogeochemical remobilization and wastewater treatment plant effluent are sources of TP in the surface water. The relative importance of these sources, however, cannot be determined.
- Rainfall events do not results in TP concentration peaks. Transport of particulate P that originates from groundwater and (agricultural) drains discharge is strongly retained but particulate P can be remobilized due to biogeochemical processes in the sediment layer at other moments. This makes it difficult to link agricultural practice to P concentrations in the surface water and this should be accounted for when judging measures to reduce P loads from agriculture.
- The artificial pumping regime and high retention capacity of polder catchments, however, enables the potential for measures to reclaim P from the water systems after being leached from the soil but before being transported to downstream surface water bodies.

Conclusions with respect to N:

- The NO₃ load to surface water originates from subsurface drains in the agricultural area, likely in combination with quick interflow via clay cracks, that start discharging upon intensive rainfall events and result in a quick response of the NO₃ concentration at the polder outlet.
- Intensive rainfall events within a few weeks after manure application in February results in incidental losses of NO₃.

In general it can furthermore be concluded that:

- Surface run-off is generally not an important transport mechanism controlling NO₃, P and SS dynamics in the polder catchment, expect at the end of a freeze-thaw cycle.
- The timing of sampling relative to the operating hours of a pumping station affects the concentration of TP and SS and this should be accounted for when calculating P export loads, determining trends in water quality or when evaluating water quality against ecological thresholds and standards.

High-frequency monitoring at the outlet of an agriculture-dominated lowland water system in combination with low frequency monitoring within the area significantly improves insight in nutrient sources and transport processes.

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1 Discharge water from subsurface drains, likely in combination with quick interflow via clay cracks,
2 has a dominant contribution to NO_3^- loads to surface water, mainly originating from N losses from
3 agricultural lands during the winter. Transfer function noise modelling of hourly NO_3^- concentrations
4 reveals that quite a large part of the dynamics in NO_3^- concentrations during the winter months can
5 be related to rainfall once groundwater tables have risen close to the tube drain levels. The NO_3^-
6 loads appear as incidental losses upon intensive rainfall events and cause high NO_3^- concentrations at
7 the polder outlet within approximately five days after the rainfall event. Such dynamics are difficult
8 to detect with grab samples.

9 Total P cannot be linked to a dominant source. The TP concentration decreases in response to wet
10 periods, this implies that groundwater seepage is an important source of TP. High DRP/TP ratios in
11 grab samples from different location within the polder in the summer and autumn months further
12 suggest that biogeochemical remobilization from bed sediments or mineralization of organic P from
13 algae or plant debris additionally contributed to the TP concentration. The effluent load of a
14 wastewater treatment plant finally attributes the TP concentration. Agriculture did not seem to be a
15 direct source of the TP concentration at the polder outlet. At the moments when water at the polder
16 outlet was enriched in NO_3^- , originated from the agricultural land, it had low TP concentrations.

17 Short scale responses of the NO_3^- and TP concentration on rainfall events indicate that run-off is not
18 an important process that controls nutrient export from the polder. A decline of the NO_3^-
19 concentration of the channel water (caused by dilution with NO_3^- poor run-off water) in combination
20 with an increase in the TP concentration and turbidity (by surface erosion and associated particulate
21 P transport) was only observed at the polder outlet during a rainfall event at the end of a freeze-
22 thaw cycle. Under non-freezing conditions, rainfall infiltrates into the soil where it gets enriched in
23 NO_3^- and contributes to tube drain discharge due to preferential flow through the cracked clay soil.
24 This drain discharge may also be enriched in TP but this is then buffered in the water system due to
25 sedimentation of particulate P where it may become a source for DRP release from bed sediments
26 during the summer and autumn months.

27 High frequency monitoring shows that the water discharge from the polder generated by the
28 pumping station initiates short scale hydrodynamic resuspension of particulate P from the channel
29 bed sediment and thus an increase of the TP concentration in the surface water during pumping.
30 This process is responsible for 21% of the annual TP export load from the polder catchment. Changes
31 in the TP concentration upon pumping are considerably smaller compared to discharge driven
32 concentration changes in response to rainfall events in natural catchments. These findings suggest
33 that the P retention capacity of polder water systems is high because flow velocities are maximized

1 by the power of the pumping station. This result in a large P retention compared with natural
2 catchments where incidental losses during peak flow conditions control the export load.
3 A change in pumping regime caused by a transformation of the pumping station from powering with
4 diesel engines to electric engines leads to a trend suggesting decreasing TP concentrations in the
5 surface water that now should be considered artificial. Our data suggest increasing TP
6 concentrations when analysing the individual time series before and after the transformation. The
7 timing of sampling relative to the operating hours of the pumping station affects the concentration
8 and this should be accounted for when calculating P export loads, determining trends in water
9 quality or when judging water quality against ecological thresholds and standards. High frequency
10 monitoring appears to be an effective tool to reveal this kind of difficult to notice artificial responses
11 in surface water quality.

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18 References

- 19 APHA-AWWA-WPCF: Standard Methods for the Examination of Water and Waste Water, edited by:
20 Clesceri, G., Trussell, American Public Health Association, Washington, D.C, 1989.
- 21 Baken, S., Verbeek, M., Verheyen, D., Diels, J., and Smolders, E.: Phosphorus losses from agricultural
22 land to natural waters are reduced by immobilization in iron-rich sediments of drainage ditches,
23 Water Research, 71, 160-170, 2015.
- 24 Berendrecht, W. L., Heemink, A. W., van Geer, F. C., and Gehrels, J. C.: Decoupling of modeling and
25 measuring interval in groundwater time series analysis based on response characteristics, Journal of
26 Hydrology, 278, 1-16, [http://dx.doi.org/10.1016/S0022-1694\(03\)00075-1](http://dx.doi.org/10.1016/S0022-1694(03)00075-1), 2003.
- 27 Bierzoza, M. Z., Heathwaite, A. L., Mullinger, N. J., and Keenan, P. O.: Understanding nutrient
28 biogeochemistry in agricultural catchments: The challenge of appropriate monitoring frequencies,
29 Environmental Sciences: Processes and Impacts, 16, 1676-1691, 10.1039/c4em00100a, 2014.
- 30 Boers, P. C. M., and van Hese, O.: Phosphorus release from the peaty sediments of the Loosdrecht
31 Lakes (The Netherlands), Water Research, 22, 355-363, 1988.
- 32 Bouwman, A. F., Bierkens, M. F. P., Griffioen, J., Hefting, M. M., Middelburg, J. J., Middelkoop, H.,
33 and Slomp, C. P.: Nutrient dynamics, transfer and retention along the aquatic continuum from land
34 to ocean: towards integration of ecological and biogeochemical models, Biogeosciences, 10, 1-22,
35 10.5194/bg-10-1-2013, 2013a.
- 36 Bouwman, L., Goldewijk, K. K., Van Der Hoek, K. W., Beusen, A. H. W., Van Vuuren, D. P., Willems, J.,
37 Rufino, M. C., and Stehfest, E.: Exploring global changes in nitrogen and phosphorus cycles in

1 agriculture induced by livestock production over the 1900–2050 period, *Proceedings of the National*
2 *Academy of Sciences*, 110, 20882-20887, 10.1073/pnas.1012878108, 2013b.

3 Bowes, M. J., Jarvie, H. P., Halliday, S. J., Skeffington, R. A., Wade, A. J., Loewenthal, M., Gozzard, E.,
4 Newman, J. R., and Palmer-Felgate, E. J.: Characterising phosphorus and nitrate inputs to a rural river
5 using high-frequency concentration–flow relationships, *Science of The Total Environment*, 511, 608-
6 620, <http://dx.doi.org/10.1016/j.scitotenv.2014.12.086>, 2015.

7 Box, G. E. P., and Jenkins, G. M.: *Time Series Analysis: Forecasting and Control*, Holden-Day, San
8 Francisco, 575 pp., 1970.

9 Cassidy, R., and Jordan, P.: Limitations of instantaneous water quality sampling in surface-water
10 catchments: Comparison with near-continuous phosphorus time-series data, *Journal of Hydrology*,
11 405, 182-193, <http://dx.doi.org/10.1016/j.jhydrol.2011.05.020>, 2011.

12 Cirno, C. P., and McDonnell, J. J.: Linking the hydrologic and biogeochemical controls of nitrogen
13 transport in near-stream zones of temperate-forested catchments: a review, *Journal of Hydrology*,
14 199, 88-120, [http://dx.doi.org/10.1016/S0022-1694\(96\)03286-6](http://dx.doi.org/10.1016/S0022-1694(96)03286-6), 1997.

15 Cleveland, W. S.: Robust locally weighted regression and smoothing scatterplots, *Journal of the*
16 *American Statistical Association*, 74, 829-836, 1979.

17 Correll, D. L., Jordan, T. E., and Weller, D. E.: Transport of nitrogen and phosphorus from Rhode River
18 watersheds during storm events, *Water Resources Research*, 35, 2513-2521, 1999.

19 Djodjic, F., Ulén, B., and Bergström, L.: Temporal and spatial variations of phosphorus losses and
20 drainage in a structured clay soil, *Water Research*, 34, 1687-1695, [http://dx.doi.org/10.1016/S0043-](http://dx.doi.org/10.1016/S0043-1354(99)00312-7)
21 [1354\(99\)00312-7](http://dx.doi.org/10.1016/S0043-1354(99)00312-7), 2000.

22 Duan, S., Kaushal, S. S., Groffman, P. M., Band, L. E., and Belt, K. T.: Phosphorus export across an
23 urban to rural gradient in the Chesapeake Bay watershed, *Journal of Geophysical Research:*
24 *Biogeosciences*, 117, G01025, 10.1029/2011JG001782, 2012.

25 Evans, D. J., Johnes, P. J., and Lawrence, D. S.: Physico-chemical controls on phosphorus cycling in
26 two lowland streams. Part 2–The sediment phase, *Science of The Total Environment*, 329, 165-182,
27 10.1016/j.scitotenv.2004.02.023, 2004.

28 Ferrick, M. G., and Gatto, L. W.: Quantifying the effect of a freeze-thaw cycle on soil erosion:
29 Laboratory experiments, *Earth Surface Processes and Landforms*, 30, 1305-1326, 10.1002/esp.1209,
30 2005.

31 Fraters, B., Kovar, K., Willems, W., Stockmarr, J., and Grant, R.: Monitoring effectiveness of the EU
32 Nitrates Directive Action Programmes. Results of the international MonNO3 workshop in the
33 Netherlands, 11-12 June 2003, RIVM, Bilthoven, The Netherlands, 2005.

34 Gao, P.: Understanding watershed suspended sediment transport, *Progress in Physical Geography*,
35 32, 243-263, 2008.

36 Gemalen: Available from: <http://www.gemalen.nl/> (Accessed 12 May 2015), 2015.

37 Geurts, J. J. M., Hetjens, H., and Lamers, L. P. M.: Remobilization of nutrients after un-deepening of
38 lakes (in dutch), Radbouduniversiteit Nijmegen, Nijmegen, 2013.

39 Griffioen, J., Vermooten, S., and Janssen, G.: Geochemical and palaeohydrological controls on the
40 composition of shallow groundwater in the Netherlands, *Applied Geochemistry*, 39, 129-149,
41 <http://dx.doi.org/10.1016/j.apgeochem.2013.10.005>, 2013.

42 Groen, K. P.: Pesticide leaching in polders, Field and model studies on cracked clays and loamy sand,
43 Van land tot zee, Ministerie van Verkeer en Waterstaat, Directoraat-Generaal Rijkswaterstaat,
44 Lelystad, 1997.

45 Halliday, S. J., Wade, A. J., Skeffington, R. A., Neal, C., Reynolds, B., Rowland, P., Neal, M., and Norris,
46 D.: An analysis of long-term trends, seasonality and short-term dynamics in water quality data from
47 Plynlimon, Wales, *Science of The Total Environment*, 434, 186-200,
48 <http://dx.doi.org/10.1016/j.scitotenv.2011.10.052>, 2012.

49 Haygarth, P. M., Wood, F. L., Heathwaite, A. L., and Butler, P. J.: Phosphorus dynamics observed
50 through increasing scales in a nested headwater-to-river channel study, *Science of The Total*
51 *Environment*, 344, 83-106, <http://dx.doi.org/10.1016/j.scitotenv.2005.02.007>, 2005.

Formatted: Dutch (Netherlands)

1 Hewlett, J. D., and Hibbert, A. R.: Moisture and energy conditions within a sloping soil mass during
2 drainage, *Journal of Geophysical Research*, 68, 1081-1087, 1963.

3 Hirsch, R. M., Slack, J. R., and Smith, R. A.: Techniques of trend analysis for monthly water quality
4 data, *Water Resources Research*, 18, 107-121, 10.1029/WR018i001p00107, 1982.

5 Hirsch, R. M., and Slack, J. R.: A Nonparametric Trend Test for Seasonal Data With Serial Dependence,
6 *Water Resources Research*, 20, 727-732, 10.1029/WR020i006p00727, 1984.

7 Hodgkinson, R. A., Chambers, B. J., Withers, P. J. A., and Cross, R.: Phosphorus losses to surface
8 waters following organic manure applications to a drained clay soil, *Agricultural Water Management*,
9 57, 155-173, [http://dx.doi.org/10.1016/S0378-3774\(02\)00057-4](http://dx.doi.org/10.1016/S0378-3774(02)00057-4), 2002.

10 Huebsch, M., Grimmeisen, F., Zemann, M., Fenton, O., Richards, K. G., Jordan, P., Sawarieh, A., Blum,
11 P., and Goldscheider, N.: Technical Note: Field experiences using UV/VIS sensors for high-resolution
12 monitoring of nitrate in groundwater, *Hydrol. Earth Syst. Sci.*, 19, 1589-1598, 10.5194/hess-19-1589-
13 2015, 2015.

14 Jarvie, H. P., Mortimer, R. J. G., Palmer-Felgate, E. J., Quinton, K. S., Harman, S. A., and Carbo, P.:
15 Measurement of soluble reactive phosphorus concentration profiles and fluxes in river-bed
16 sediments using DET gel probes, *Journal of Hydrology*, 350, 261-273, 2008.

17 Johnes, P. J.: Uncertainties in annual riverine phosphorus load estimation: Impact of load estimation
18 methodology, sampling frequency, baseflow index and catchment population density, *Journal of*
19 *Hydrology*, 332, 241-258, <http://dx.doi.org/10.1016/j.jhydrol.2006.07.006>, 2007.

20 Jones, T. D., Chappell, N. A., and Tych, W.: First Dynamic Model of Dissolved Organic Carbon Derived
21 Directly from High-Frequency Observations through Contiguous Storms, *Environmental Science &*
22 *Technology*, 48, 13289-13297, 10.1021/es503506m, 2014.

23 Jordan, P., Arnscheidt, A., McGrogan, H., and McCormick, S.: Characterising phosphorus transfers in
24 rural catchments using a continuous bank-side analyser, *Hydrology and Earth System Sciences*, 11,
25 372-381, 2007.

26 Kaufmann, V., Pinheiro, A., and Castro, N. M. d. R.: Simulating transport of nitrogen and phosphorus
27 in a Cambisol after natural and simulated intense rainfall, *Journal of Contaminant Hydrology*, 160,
28 53-64, <http://dx.doi.org/10.1016/j.jconhyd.2014.02.005>, 2014.

29 Kaushal, S. S., Mayer, P. M., Vidon, P. G., Smith, R. M., Pennino, M. J., Newcomer, T. A., Duan, S.,
30 Welty, C., and Belt, K. T.: Land use and climate variability amplify carbon, nutrient, and contaminant
31 pulses: A review with management implications, *Journal of the American Water Resources*
32 *Association*, 50, 585-614, 10.1111/jawr.12204, 2014.

33 Kirchner, J. W., Feng, X., Neal, C., and Robson, A. J.: The fine structure of water-quality dynamics: the
34 (high-frequency) wave of the future, *Hydrological Processes*, 18, 1353-1359, 10.1002/hyp.5537,
35 2004.

36 Knotters, M., and van Walsum, P. E. V.: Estimating fluctuation quantities from time series of water-
37 table depths using models with a stochastic component, *Journal of Hydrology*, 197, 25-46,
38 [http://dx.doi.org/10.1016/S0022-1694\(96\)03278-7](http://dx.doi.org/10.1016/S0022-1694(96)03278-7), 1997.

39 Lavoie, M., and Auclair, J.-C.: Phosphorus Mobilization at the Sediment–Water Interface in Softwater
40 Shield Lakes: the Role of Organic Carbon and Metal Oxyhydroxides, *Aquatic Geochemistry*, 18, 327-
41 341, 10.1007/s10498-012-9166-3, 2012.

42 LNV: Fourth Dutch Action Programme (2010-2013) concerning the Nitrates Directive; 91/676/EEC. ,
43 The Hague, 2009.

44 Loeb, R., Lamers, L. P. M., and Roelofs, J. G. M.: Prediction of phosphorus mobilisation in inundated
45 floodplain soils, *Environmental Pollution*, 156, 325-331, 10.1016/j.envpol.2008.02.006, 2008.

46 Meinardi, C. R., and Van den Eertwegh, G. A. P. H.: Investigations on tile drains in clayey regions of
47 the Netherlands, Part II: Interpretation of data, RIVM, Bilthoven, The Netherlands, 1997.

48 Mulholland, P. J., Newbold, J. D., Elwood, J. W., Ferren, L. A., and Jackson, R. W.: Phosphorus
49 Spiralling in a Woodland Stream: Seasonal Variations, *Ecology*, 66, 1012-1023, 1985.

50 Murphy, J., and Riley, J. P.: A modified single solution method for the determination of phosphate in
51 natural waters, *Analytica Chimica Acta*, 27, 31-36, 1962.

1 Neal, C., Reynolds, B., Rowland, P., Norris, D., Kirchner, J. W., Neal, M., Sleep, D., Lawlor, A., Woods,
2 C., Thacker, S., Guyatt, H., Vincent, C., Hockenhull, K., Wickham, H., Harman, S., and Armstrong, L.:
3 High-frequency water quality time series in precipitation and streamflow: From fragmentary signals
4 to scientific challenge, *Science of The Total Environment*, 434, 3-12,
5 <http://dx.doi.org/10.1016/j.scitotenv.2011.10.072>, 2012.

6 Nyenje, P. M., Meijer, L. M. G., Foppen, J. W., Kulabako, R., and Uhlenbrook, S.: Phosphorus
7 transport and retention in a channel draining an urban, tropical catchment with informal
8 settlements, *Hydrol. Earth Syst. Sci.*, 18, 1009-1025, 10.5194/hess-18-1009-2014, 2014.

9 Palmer-Felgate, E. J., Jarvie, H. P., Williams, R. J., Mortimer, R. J. G., Loewenthal, M., and Neal, C.:
10 Phosphorus dynamics and productivity in a sewage-impacted lowland chalk stream, *Journal of*
11 *Hydrology*, 351, 87-97, <http://dx.doi.org/10.1016/j.jhydrol.2007.11.036>, 2008.

12 Poor, C. J., and McDonnell, J. J.: The effects of land use on stream nitrate dynamics, *Journal of*
13 *Hydrology*, 332, 54-68, <http://dx.doi.org/10.1016/j.jhydrol.2006.06.022>, 2007.

14 Regelink, I. C., Koopmans, G. F., van der Salm, C., Weng, L., and van Riemsdijk, W. H.:
15 Characterization of Colloidal Phosphorus Species in Drainage Waters from a Clay Soil Using
16 Asymmetric Flow Field-Flow Fractionation, *J. Environ. Qual.*, 42, 464-473, 10.2134/jeq2012.0322,
17 2013.

18 Rozemeijer, J. C., and Broers, H. P.: The groundwater contribution to surface water contamination in
19 a region with intensive agricultural land use (Noord-Brabant, The Netherlands), *Environmental*
20 *Pollution*, 148, 695-706, 10.1016/j.envpol.2007.01.028, 2007.

21 Rozemeijer, J. C., Van der Velde, Y., Van Geer, F. C., De Rooij, G. H., Torfs, P. J. J. F., and Broers, H. P.:
22 Improving load estimates for NO₃ and P in surface waters by characterizing the concentration
23 response to rainfall events, *Environmental Science and Technology*, 44, 6305-6312, 2010.

24 Rozemeijer, J. C., Klein, J., Broers, H. P., Van Tol-Leenders, T. P., and Van Der Grift, B.: Water quality
25 status and trends in agriculture-dominated headwaters; a national monitoring network for assessing
26 the effectiveness of national and European manure legislation in The Netherlands, *Environmental*
27 *Monitoring and Assessment*, 186, 8981-8995, 10.1007/s10661-014-4059-0, 2014.

28 Schoch, A. L., Schilling, K. E., and Chan, K.-S.: Time-series modeling of reservoir effects on river
29 nitrate concentrations, *Advances in Water Resources*, 32, 1197-1205,
30 <http://dx.doi.org/10.1016/j.advwatres.2009.04.002>, 2009.

31 Shrestha, R. R., Osenbrück, K., and Rode, M.: Assessment of catchment response and calibration of a
32 hydrological model using high-frequency discharge nitrate concentration data, *Hydrology Research*,
33 44, 995-1012, 10.2166/nh.2013.087, 2013.

34 Simard, R. R., Beauchemin, S., and Haygarth, P. M.: Potential for preferential pathways of
35 phosphorus transport, *Journal of Environmental Quality*, 29, 97-105, 2000.

36 Skeffington, R. A., Halliday, S. J., Wade, A. J., Bowes, M. J., and Loewenthal, M.: Using high-frequency
37 water quality data to assess sampling strategies for the EU Water Framework Directive, *Hydrol. Earth*
38 *Syst. Sci.*, 19, 2491-2504, 10.5194/hess-19-2491-2015, 2015.

39 Stutter, M. I., Langan, S. J., and Cooper, R. J.: Spatial contributions of diffuse inputs and within-
40 channel processes to the form of stream water phosphorus over storm events, *Journal of Hydrology*,
41 350, 203-214, 10.1016/j.jhydrol.2007.10.045, 2008.

42 Tiemeyer, B., Lennartz, B., and Kahle, P.: Analysing nitrate losses from an artificially drained lowland
43 catchment (North-Eastern Germany) with a mixing model, *Agriculture, Ecosystems & Environment*,
44 123, 125-136, <http://dx.doi.org/10.1016/j.agee.2007.05.006>, 2008.

45 van Boekel, E. M. P. M., Roelsma, J., Massop, H. T. L., Hendriks, R. F. A., Goedhart, P. W., and Jansen,
46 P. C.: Nitraatconcentraties in het drainwater in zeekleigebieden : oriënterend onderzoek naar de
47 oorzaken van de verhoogde nitraatconcentraties, *Alterra-rapport;2360*, Alterra Wageningen UR,
48 Wageningen, 2012.

49 Van de Ven, G. P.: Man-made lowlands, history of water management and land reclamation in the
50 Netherlands, *Uitgeverij Matrijs*, Utrecht, 2003.

Formatted: Dutch (Netherlands)

1 Van den Eertwegh, G. A. P. H.: Water and Nutrient budgets at field and regional scale, travel times of
2 drainage water and nutrient loads to surface water, PhD thesis Wageningen University, 2002.
3 van der Grift, B., Rozemeijer, J. C., Griffioen, J., and van der Velde, Y.: Iron oxidation kinetics and
4 phosphate immobilization along the flow-path from groundwater into surface water, *Hydrol. Earth
5 Syst. Sci.*, 18, 4687-4702, 10.5194/hess-18-4687-2014, 2014.
6 Van der Salm, C., van den Toorn, A., Chardon, W. J., and Koopmans, G. F.: Water and nutrient
7 transport on a heavy clay soil in a fluvial plain in the Netherlands, *Journal of Environmental Quality*,
8 41, 229-241, 10.2134/jeq2011.0292, 2012.
9 van Grinsven, H. J. M., ten Berge, H. F. M., Dalgaard, T., Fraters, B., Durand, P., Hart, A., Hofman, G.,
10 Jacobsen, B. H., Lalor, S. T. J., Lesschen, J. P., Osterburg, B., Richards, K. G., Techen, A. K., Vertès, F.,
11 Webb, J., and Willems, W. J.: Management, regulation and environmental impacts of nitrogen
12 fertilization in northwestern Europe under the Nitrates Directive; a benchmark study,
13 *Biogeosciences*, 9, 5143-5160, 10.5194/bg-9-5143-2012, 2012.
14 von Asmuth, J. R., Bierkens, M. F. P., and Maas, K.: Transfer function-noise modeling in continuous
15 time using predefined impulse response functions, *Water Resources Research*, 38, 23-21-23-12,
16 10.1029/2001WR001136, 2002.
17 Wade, A. J., Palmer-Felgate, E. J., Halliday, S. J., Skeffington, R. A., Loewenthal, M., Jarvie, H. P.,
18 Bowes, M. J., Greenway, G. M., Haswell, S. J., Bell, I. M., Joly, E., Fallatah, A., Neal, C., Williams, R. J.,
19 Gozzard, E., and Newman, J. R.: Hydrochemical processes in lowland rivers: Insights from in situ,
20 high-resolution monitoring, *Hydrology and Earth System Sciences*, 16, 4323-4342, 10.5194/hess-16-
21 4323-2012, 2012.
22 Withers, P. J. A., Ulén, B., Stamm, C., and Bechmann, M.: Incidental phosphorus losses – are they
23 significant and can they be predicted?, *Journal of Plant Nutrition and Soil Science*, 166, 459-468,
24 10.1002/jpln.200321165, 2003.
25 Withers, P. J. A., and Jarvie, H. P.: Delivery and cycling of phosphorus in rivers: A review, *Science of
26 The Total Environment*, 400, 379-395, 2008.
27 Worrall, F., Swank, W. T., and Burt, T. P.: Changes in stream nitrate concentrations due to land
28 management practices, ecological succession, and climate: Developing a systems approach to
29 integrated catchment response, *Water Resources Research*, 39, HWC11-HWC114, 2003.
30 Worsfold, P. J., Gimbert, L. J., Mankasingh, U., Omaka, O. N., Hanrahan, G., Gardolinski, P. C. F. C.,
31 Haygarth, P. M., Turner, B. L., Keith-Roach, M. J., and McKelvie, I. D.: Sampling, sample treatment
32 and quality assurance issues for the determination of phosphorus species in natural waters and soils,
33 *Talanta*, 66, 273-293, 2005.
34 Zak, D., Kleeberg, A., and Hupfer, M.: Sulphate-mediated phosphorus mobilization in riverine
35 sediments at increasing sulphate concentration, *River Spree, NE Germany, Biogeochemistry*, 80, 109-
36 119, 2006.

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1 Tables

2 Table 1. Locations of the low-frequency monitoring program in Lage Afdeling pumped drainage area
3 that is drained by the Blocq van Kuffeler pumping station.

location	Description
1	Lage Vaart main-channel at pumping station "Blocq van Kuffeler"; outlet of the Lage Afdeling drainage area
2	Outlet of sub-channel that drains the urban area of the city "Almere"
3	Outlet of sub-channel that drains the agricultural "Gruttotocht"
4	Outlet of sub-channel that drains the agricultural "Lepelaartocht"
5	Far end of Lage Vaart main channel that is drained by the pumping station "Blocq van Kuffeler"
6	Outlet of channel that drains the nature area "Oostvaardersplassen"

4

5 Table 2. Rainfall events and response of NO₃ concentration (in mg N L⁻¹).

Rainfall event	date	mm	NO ₃ concentration before event	Maximum NO ₃ concentration after event
1	20-23 Oct	31	0.7	0.8
2	3-4 Nov	16	0.8	0.9
3	15-18 Nov	23	0.8	4.6
4	10-12 Dec	29	1.0	5.3
5	19-20 Dec	24	2.4	5.9
6	7-9 Jan	14	3.0	5.8
7	12-14 Jan	24	4.1	9.0
8	20-21 Feb	26	0.8	10.4
9	29 Mar-2-Apr	43	0.8	6.1
10	17-23 June	40	0.2	0.5
11	27-29 July	47	0.5	0.7
12	14-18 Aug	87	0.6	3.4
13	26-31 Aug	59	2.4	4.7

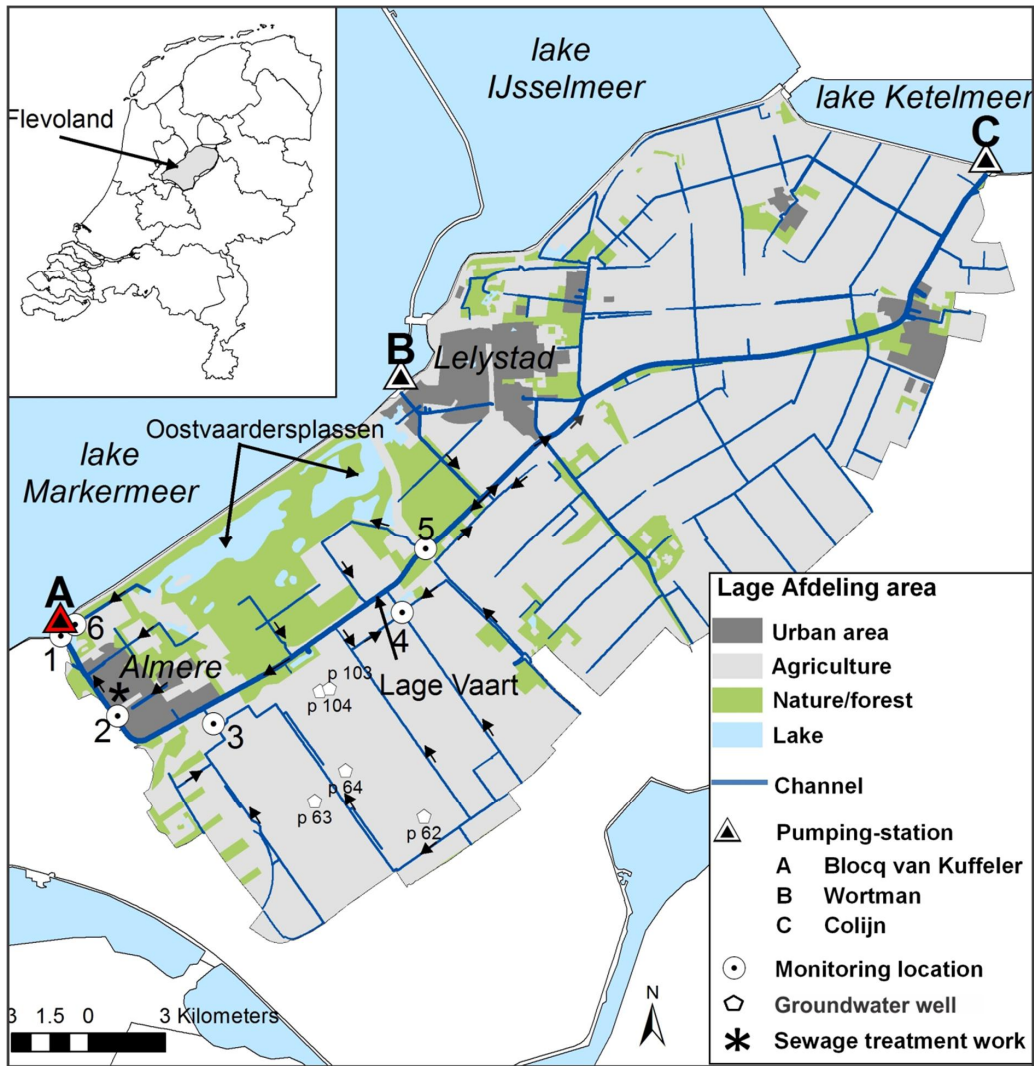
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7 Table 3. Summary of TP and turbidity peaks, calculated as difference between the maximum value
8 during the peak minus the value before the peak, induced by the pumping station

	Δ TP (mg L ⁻¹) 1 pump	Δ turbidity (FTU) 1 pump	Δ TP (mg L ⁻¹) 2 pumps	Δ turbidity (FTU) 2 pumps
n peaks	72	79	59	60
average	0.06	4.4	0.13	22.1
median	0.04	4.4	0.10	21.1
P25	0.01	1.8	0.07	14.0
P75	0.08	8.3	0.14	29.2
max	0.58	26.2	0.61	52.0
min	-0.01	-1.5	0.03	5.9

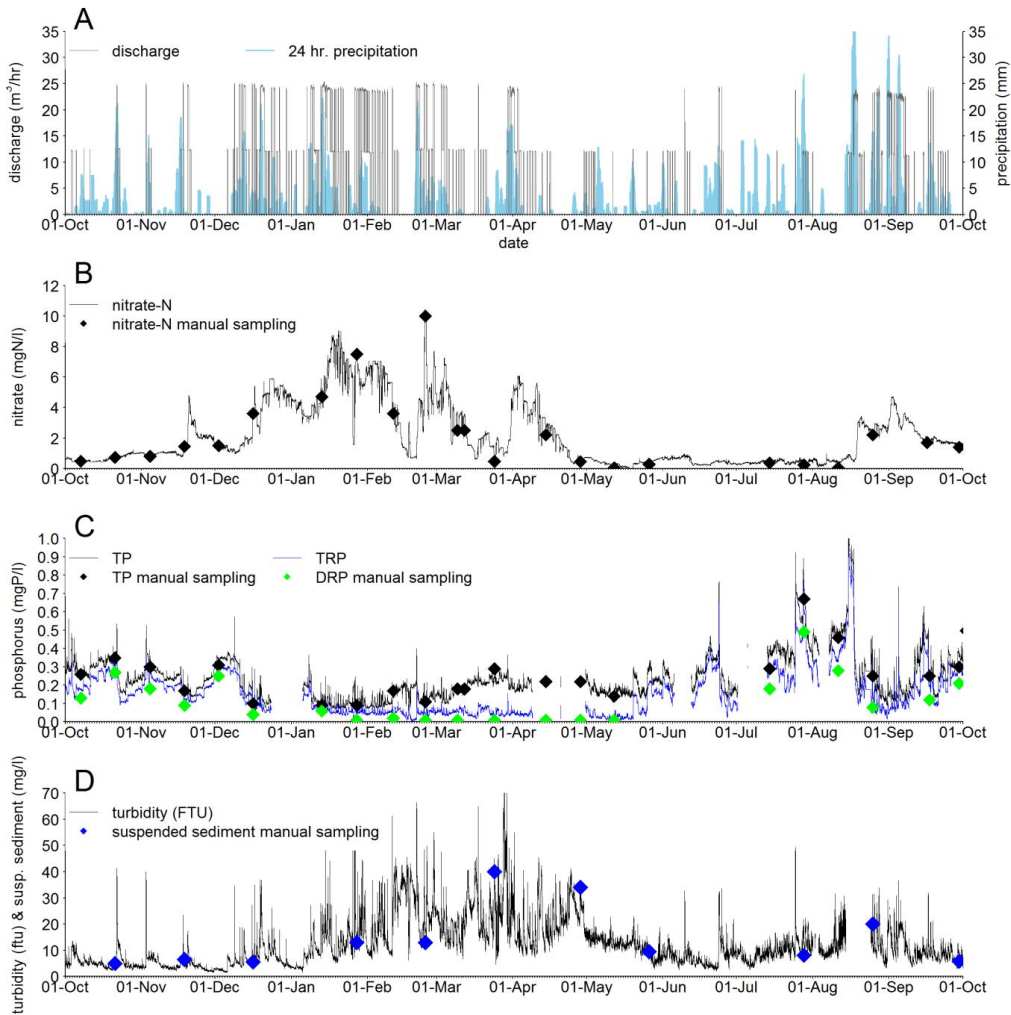
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1 Figures

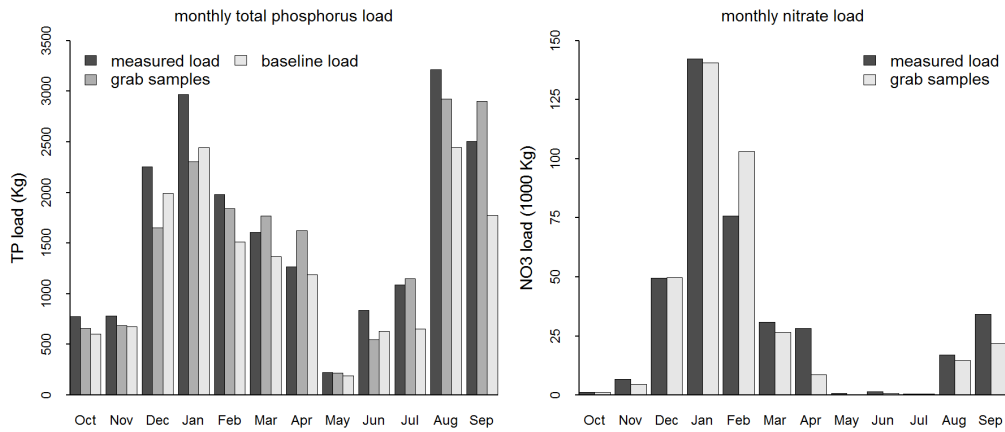


2
3 Figure 1. Map of the Lage Afdeling pumped drainage area, the continuous monitoring station at
4 location A, the low-frequency surface water monitoring locations and the groundwater level
5 monitoring wells. The flow direction of the water in the channels that are drained by pumping
6 station Blocq van Kuffeler is illustrated by arrows.

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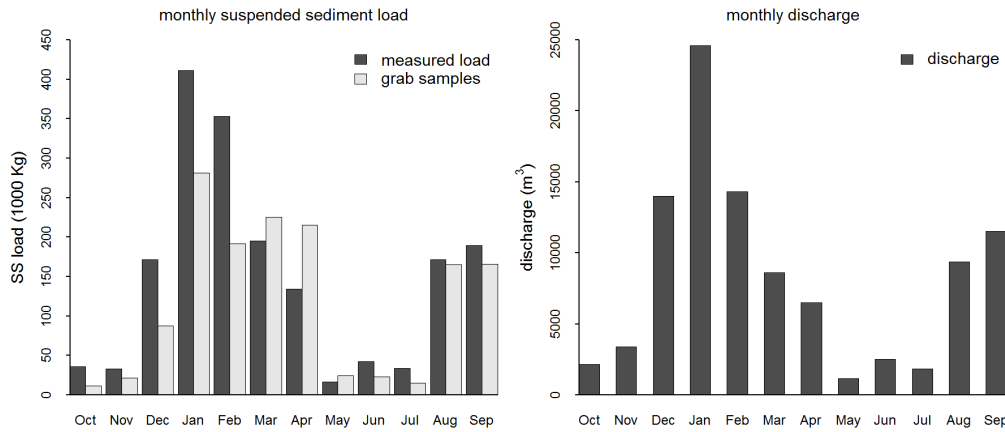


1
 2 Figure 2. High-frequency monitoring data for the Lage Vaart channel at the pumping station Blocq
 3 van Kuffeler ~~together with the 1 day antecedent precipitation and discharge as generated by the~~
 4 ~~pumping station: (A) discharge as generated by the pumping station and 1 day antecedent~~
 5 ~~precipitation; (top)B) nitrate-N 5 minutes data, with NO₃-N manual sampled biweekly data; (middle)C)~~
 6 ~~total phosphorus and total reactive phosphorus 20 minutes data, with TP and DRP manual sampled~~
 7 ~~biweekly data; (bottom)D) turbidity 5 minutes data, with suspended sediment manual sampled~~
 8 monthly data.



1

Seasonal and annual load	winter (Oct-Mar)	summer (Apr-Sep)	year	Seasonal and annual load	winter (Oct-Mar)	summer (Apr-Sep)	year
TP (kg)	10350	9150	19500	NO ₃ (1000 kg)	306.3	82.3	388.6
TP grab samples (kg)	8900	9350	18300	NO ₃ grab samples (1000 kg)	326.2	46.4	372.6
TP baseline (kg)	8600	6900	15500				

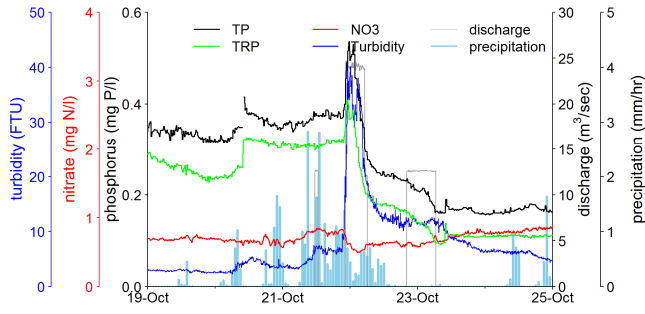


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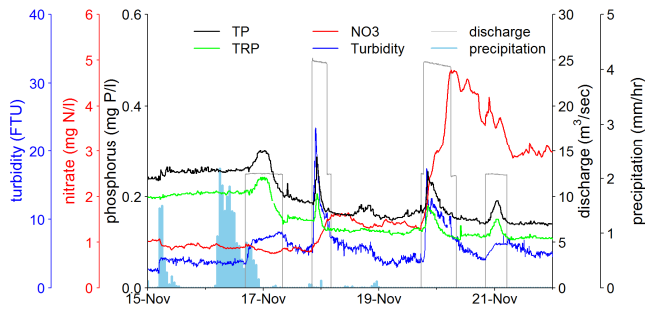
Seasonal and annual load	winter (Oct-Mar)	summer (Apr-Sep)	year	Seasonal and annual load	winter (Oct-Mar)	summer (Apr-Sep)	year
SS (1000 kg)	1200	588	1788	Discharge (m3)	67000	33000	100000
SS grab samples (1000 kg)	819	609	1428				

3 Figure 3: TP, NO₃ and SS export loads and discharge at the pumping station Blocq van Kuffeler. The
 4 measured load is calculated from the high-frequency data, the grab sample load is calculated from
 5 interpolation of the low-frequency data, the TP baseline load was calculated with the high-frequency
 6 TP data after separation of the short-scale concentration peaks generated by the pumping station.

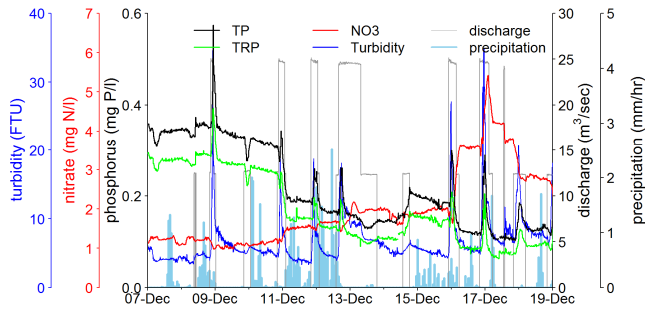
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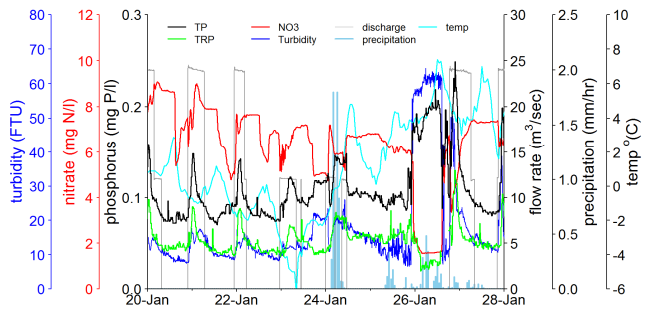
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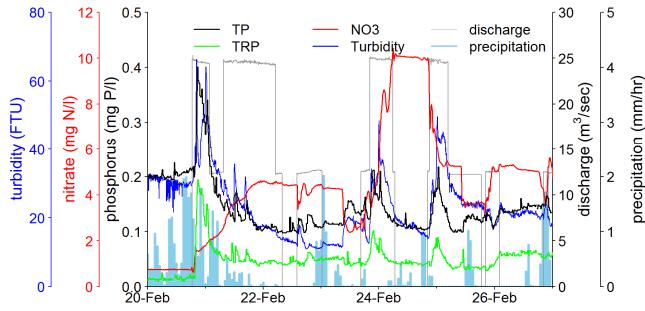
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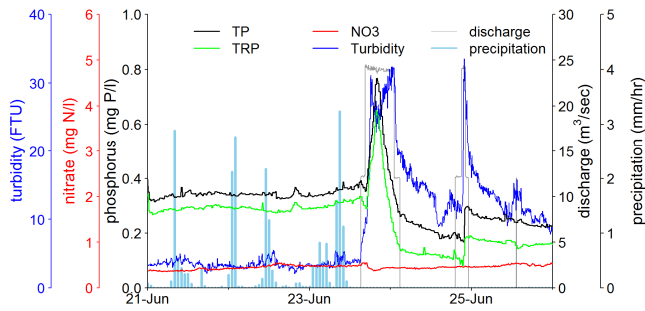
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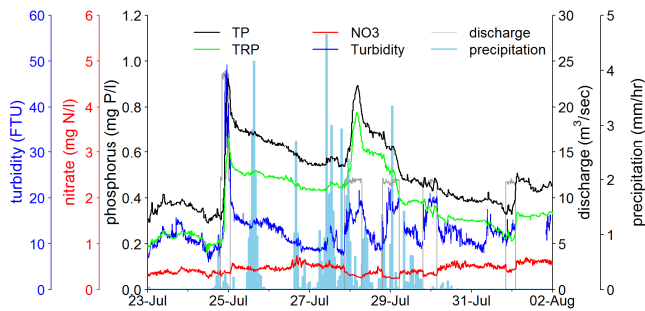
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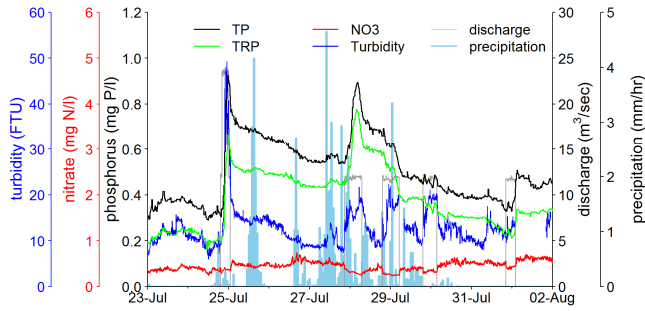
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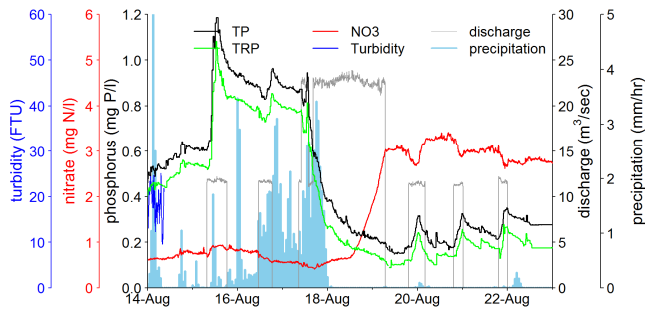
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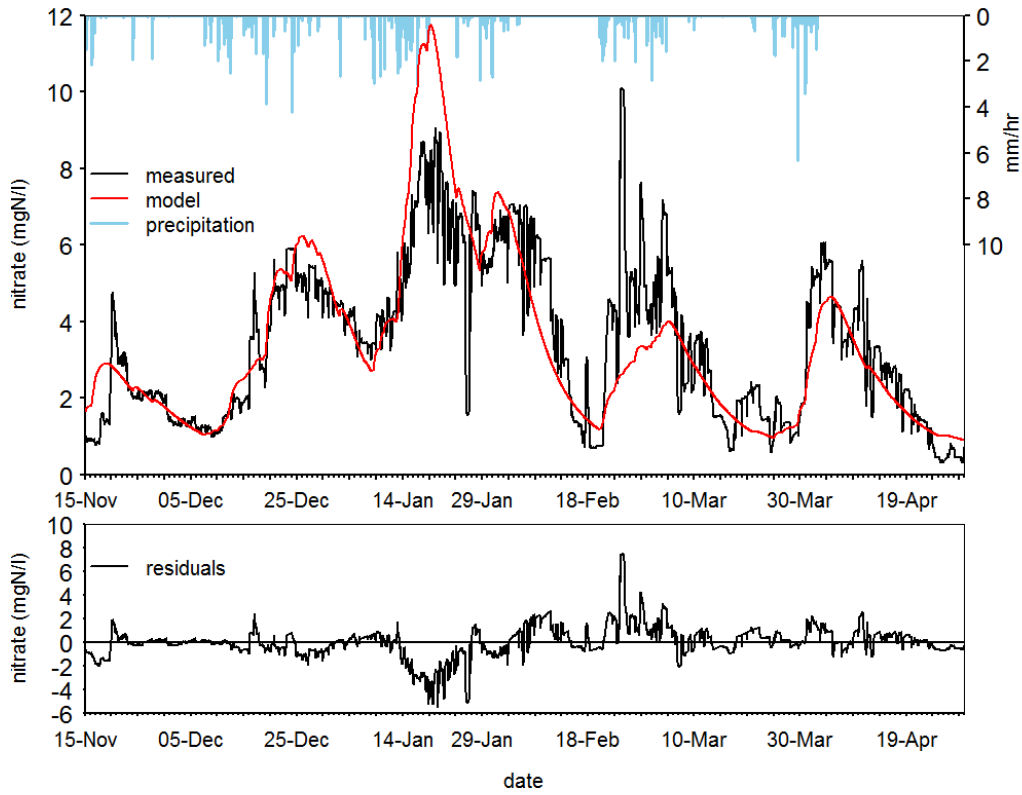
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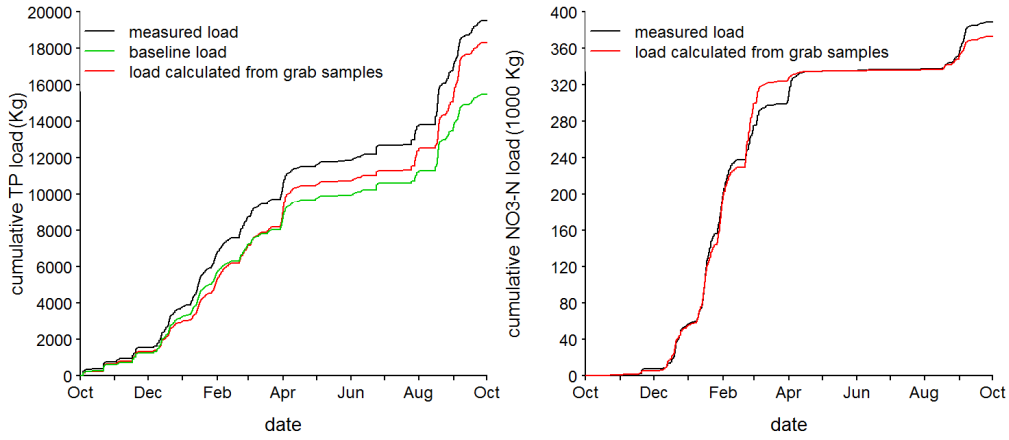
3 Figure 43. Examples of surface water NO_3 , TP and turbidity dynamics at the pumping station Blocq
4 van Kuffeler during meteorological events between October 2014 and August 2015 together with
5 the pumping regime and precipitation (in mm hr^{-1}). The January event demonstrates the effect of
6 freeze-thaw on the nutrient concentrations while the other events show the nutrient dynamics upon
7 rainfall events.



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 2 | Figure 54. Measured and simulated NO₃ concentrations and rainfall data (top); and residual NO₃
 3 | series (bottom).

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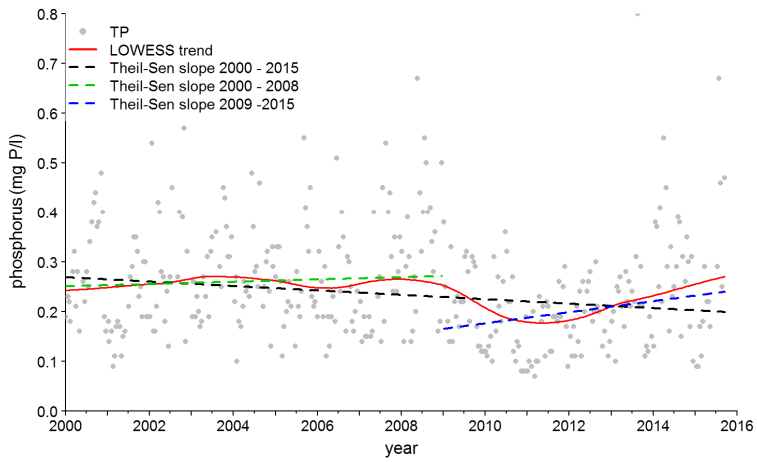
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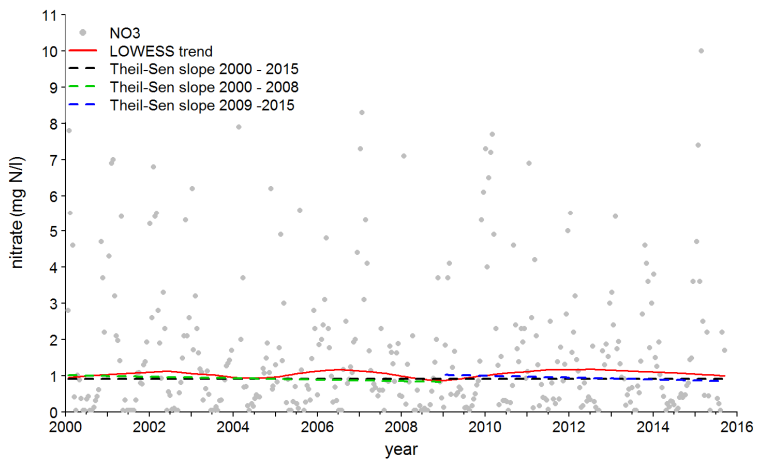
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Figure 5: Measured and calculated TP and NO₃ loads at the pumping station Blocq van Kuffeler, the baseline load was calculated with the high frequency TP data after separation of the short scale concentration peaks generated by the pumping station.

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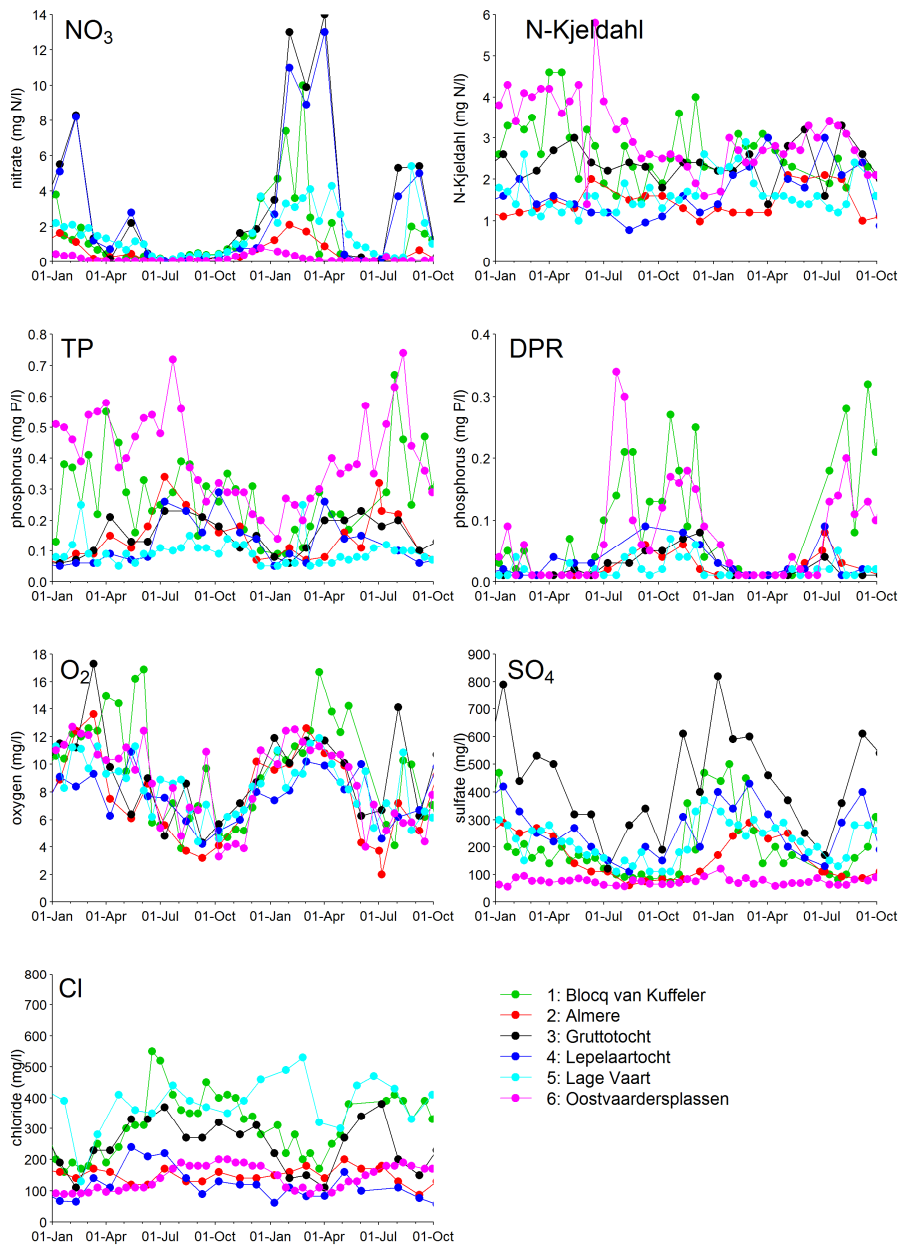


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2

3 Figure 6: Trends in TP and NO₃ concentrations over the period 2000-2015 at location 1 (Blocq van
 4 Kuffeler).



1
 2 **Figure 7. Low-frequency time series of NO₃⁻, N-kjeldahl, TP, DPR, O₂, SO₄ and Cl⁻ concentration at**
 3 **surface water sampling location in the Lage Afdeling drainage area during the period January 2014 to**
 4 **October 2015. Fig. 1 shows the positions of the monitoring locations.**