1 Long-term monitoring of nitrate transport to drainage from

2 three agricultural clayey till fields

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7 Abstract

8 The application of nitrogen (N) fertilisers to crops grown on tile-drained fields is required to 9 sustain most modern crop production, but it poses a risk to the aquatic environment since tile 10 drains facilitate rapid transport pathways with no significant reduction in nitrate. To maintain 11 the water quality of the aquatic environment and the provision of food from highly efficient 12 agriculture in line with the EU's Water Framework Directive and Nitrates Directive, fieldscale knowledge is essential for introducing water management actions on-field or off-field 13 14 and producing an optimal differentiated N-regulation in future. This study strives to provide such knowledge by evaluating on 11 years of nitrate-N concentration measurements in 15 16 drainage from three subsurface-drained clayey till fields (1.3-2.3 ha) representing 17 approximately 71 % of the surface sediments in Denmark dominated by clay. The fields differ 18 in their inherent hydrogeological field settings (e.g. soil-type, geology, climate, drainage and 19 groundwater table) and the agricultural management of the fields (e.g. crop type, type of N 20 fertilisers and agricultural practices). The evaluation revealed three types of clavey till fields 21 characterised by: (i) low net precipitation, high concentration of nitrate-N, and short-term low 22 intensity drainage at air temperatures often below 5 °C; (ii) medium net precipitation, medium 23 concentration of nitrate-N, and short-term medium intensity drainage at air temperatures often above 5 °C; and (iii) high net precipitation, low concentration of nitrate-N and long-term high 24 intensity drainage at air temperatures above 5 °C. For each type, on-field water management 25 26 actions, such as the selection of crop types and introduction of catch crops, appeared relevant, 27 whereas off-field actions only seemed relevant for the latter two field types given the 28 temperature-dependent reduction potential of nitrate off-field. This initial well-documented 29 field-scale knowledge from fields that are representative of large areas in Denmark is a first 30 step towards establishing a differentiated N-regulation for clayey till areas. Additionally, it

- 1 provide a unique starting point by identifying important parameters for future mapping of
- 2 catchment-scale variations in nitrate concentrations and fluxes.
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1 **1 Introduction**

2 Future regulations covering aquatic environments under the Nitrates Directive (EEC, 1991), the EU Water Framework Directive (EC, 2000), the EU Groundwater Directive (EC, 2006) 3 4 and additional national laws and regulations (Danish-EPA, 2012) call for long-term 5 monitoring data to describe the complex interaction between soil, geology, geochemistry and 6 hydrology at a local level as well on larger scales. Since Denmark presented its first national 7 hydrological action plan in 1985, several political agreements have been adopted the aim of 8 protecting the aquatic environment and nature in general. Recently, the Danish Commission 9 on Nature and Agriculture issued a report (The Commission on Nature and Agriculture, 10 2013) that recommended that nitrogen (N) regulations should be adapted locally in future and, 11 if possible, at field scale. As an integrated part of such a policy, there is a need to develop 12 tools to identify fields that are vulnerable or non-vulnerable to nitrate-N leaching.

13 N is an essential plant nutrient, which is why N-application to agricultural land is essential for sustaining food and fibre production for a rapidly growing population. The agricultural 14 15 sector, however, has been identified as the largest nonpoint source contributor of nitrate to surface and groundwater bodies (Bakhsh et al., 2004; Beaudoin et al., 2005; Billy et al., 16 17 2011). The effects of agriculture may extend beyond the boundaries of fields or farms since 18 the drainage systems will inevitably affect the flow pathways of water away from agricultural 19 land and into the receiving water bodies (Robinson and Rycroft, 1999). However, the 20 nonpoint loss of N from agricultural fields is controlled by an array of factors such as soil 21 properties (physical, chemical and biological), climatic factors (precipitation and temperature 22 patterns), farming practices (cropping system, fertilisation and tillage) and hydrology (Billy et 23 al., 2011; Dinnes et al., 2002). Thus, high doses of N fertilisers may increase the N-transport to drainage, especially when there is no further crop response (Delin and Stenberg, 2014). 24 25 Crop response by catch crops in intensive agriculture in northern France has reduced the mean 26 concentration by 50 % on an annual scale (Beaudoin et al., 2005) and a winter cover of 27 mustard has reduced the nitrate-N concentrations in temperate soils (Premrov et al., 2014).

The installation of tile drains is a common agricultural water management practice for improving moisture and aeration conditions (Tiemeyer *et al.*, 2010; Van Der Ploeg *et al.*, 1999) in areas with shallow groundwater and seasonally perched groundwater tables. The presence of tile drains enhances crop yields on soils, which are highly productive and poorly natural drained, and helps reduce year-on-year variability in yields (Nangia *et al.*, 2010) by promoting earlier sowing and improving traffic ability. The drains remove water from the

land more quickly than under natural conditions and increase infiltration and deliver shallow 1 groundwater more quickly to surface water, preventing its recharge to groundwater. This 2 artificial drainage modifies N-dynamics by facilitating the rapid transport of nitrate-N and 3 greatly reducing or even suppressing the water residence time within natural retention zones. 4 5 However, tile drain systems not only remove excess water from the root zone, but also facilitate N-transport, primarily as soluble nitrate-N, from the bottom of the root zone to the 6 7 edge of the field (Billy et al., 2011; Mulla and Strock, 2008) and to surface water (Cordeiro et 8 al., 2014; Eidem et al., 1999; Gilliam et al., 1979; Lapen et al., 2008). They also contribute to 9 hypoxia (Billy et al., 2011). Drainage increases N-losses from agricultural areas as compared 10 to former undrained fields. In addition, no biogeochemical processes such as denitrification 11 are known to occur in buried pipes, which are considered inert pathways (Billy et al., 2013; 12 Gilliam et al., 1999) whereas denitrification processes may be common at temperatures above approximately 5 °C in the root zone with renewable source of bioavailable organic matter and 13 14 restricted access of oxygen due to high water saturation or high microbial activity 15 (Christensen et al., 1990; Ernstsen et al., 1998). Drainage decreases the possibilities of nitrate 16 being denitrified or adsorbed by plants on-field (Gilliam et al., 1999) and off-field, the temperature-drainage signatures are crucial for denitrification in natural and constructed 17 18 wetlands. Therefore the loss of nitrate-N leaching from the root zone is not solely an 19 environmental concern, but also an economic loss to farmers (Schjonning et al., 2013).

The greatest intensity of drainage in Europe is concentrated in the northern areas around the Baltic and North Seas. This is largely due to climatic conditions and the presence of glacially derived clayey tills that can cause prolonged water-logging (Robinson and Rycroft, Since around 1850, 50 % of agricultural land in Denmark has been systematically tile drained, with horizontal spacing of 8-20 metres (Olesen, 2009) and a total length of around 750,000 km (Breuning-Madsen, 2010).

26 A number of factors may affect the loss of nitrate-N from fields via drainage. The local 27 climate (temperature and precipitation), soil type, crop type, length of growing season, 28 options of winter cover crops and catch crops, tillage and soil management are also important 29 factors in the concentration and leaching of nitrate-N from the root zone. Some of these 30 factors are field specific and cannot be changed, *e.g.* the nitrate uptake of field crops is highly variable in the course of a year and between crops as the N uptake and growth stage are very 31 32 complex (Gastal and Lemaire, 2002), whereas other factors may be adjusted to minimise the 33 N-loss for the benefit of the environment and farmers' incomes.

To the authors' knowledge, there has been limited documentation of nitrate-N 1 2 concentrations and leaching via tile drain systems from field-size areas that have been monitored over a long period. In Denmark, data exist on nitrate-N concentrations and 3 leaching from a few short-term local-scale studies (Bennetzen, 1978; Hansen and Pedersen, 4 5 1975; Pedersen, 1983; Simmelsgaard, 1998; Simmelsgaard and Djurhuus, 1998). Common among these studies is that there is no knowledge of the specific field contribution of nitrate-6 7 N to drainage because the extent of the field's tile drain system is unknown or because there 8 are different fields with different crops contributing to the drainage. To go from field scale to 9 catchment scale, smaller-scale monitoring is important to provide an understanding of the 10 ways in which field-scale processes influence catchment-scale discharge and water quality 11 (Rozemeijer et al., 2010; van der Velde et al., 2010).

12 To be able to optimise the use of N in agriculture and minimise nitrate leaching to the 13 aquatic environment, it is imperative to be able to identify a field's natural ability to reduce 14 nitrate. The aim of this study was twofold: (i) to provide detailed field-scale insight with regard to the impact of inherent conditions (air temperature, precipitation, and 15 hydrogeological setting) and management (type of crops, crop development, amount and 16 source of N, and time for application of N fertilisers) on nitrate transport to drainage obtained 17 18 from long-term monitoring in three agricultural clayey till fields exposed to different climatic 19 conditions; and (ii) to elaborate on the impact of such detailed insight on future N regulation and N management "on-field" and "off-field" 20

21 2 Material and Methods

22 2.1 Field descriptions

Three fields (Faardrup, Silstrup and Estrup) were selected to cover different types of clayey geological settings and climates, primarily expressed by the amount of annual precipitation (Fig. 1). The study sites are representative for about 71 % of the clayey surface sediments in Denmark, with Faardrup representing 30 %, Silstrup also 30 % and Estrup 11 % (Barlebo *et al.*, 2007). A summary of the main characteristics of each field is provided in Tables 1 and 2 (Lindhardt *et al.*, 2001; Rosenbom *et al.*, 2015).

The three fields (1.3 - 2.3 ha) are located on clay till plains with a small slope (0-3 %). The clayey till plain at Faardrup is homogenous, the clayey till plain at Silstrup comprises dislocated Oligocene clay, and at Estrup there is a complex structure with deposits of different ages and compositions. At Faardrup and Silstrup, located on sediments from the Late Weichselian age (about 15000 years BP), moderately well-drained argiudoll was
 mapped with hapludoll (Silstrup) and both hapludoll and vermudoll (Faardrup). Estrup is
 located on older sediments deposited under the Saalian glaciation (about 100,000 years BP)
 with argiudoll and glossudalf (Table 2).

5 The content of organic matter in the surface layer disturbed by cultivation (Ap-horizon) was lowest (1.4-1.5 %C) in Faardrup, medium in Silstrup (1.6-2.0 %C), and highest (1.6-3.2 6 7 %C) in Estrup. In all fields, the C-contents decreased markedly at depths just below the Ap-8 horizon to contents of 0.06 to 0.8 % C. The clay content in the upper 0.2 m (Ap horizon) was 9 10-27 % and varied to six metres below the surface at between 1-65 % due to the 10 heterogeneity of the clayey till. Postglacial leaching processes have formed an upper calcium-11 free zone about one metre thick in the youngest sediments of Faardrup and Silstrup and down 12 to 1 to 4 metres at Estrup, due to the much longer ongoing weathering processes. In the 13 calcareous zone below, the CaCO₃ content varied between 21 % and 82 % in all three fields.

Subsurface tile drains were installed in the fields between the 1940s and 1960s. The drains were established at a depth of approximately 1.1 m with a horizontal spacing of 10-20 m. Before monitoring began, the fields' tile drain system was isolated from the surrounding tile drain systems to ensure that drainage only came from the fields. Any modifications were made outside the monitoring fields (Lindhardt *et al.*, 2001).

19 **2.2 Farming practices**

20 The farming practices for the fields were in line with conventional practices in the different 21 regions and with the application of nitrogen as recommended as good management practice in 22 Denmark. In 2001-2011, crops of spring barley and winter wheat were the most common 23 types in rotation with spring or winter rape, fodder or sugar beets and maize in all the fields. No catch crop has been grown in any of the fields. Red fescue for grass seed production was 24 25 grown in Faardrup and Silstrup, as well as peas in Silstrup and Estrup. Each year, the amount of N fertilisers applied was adjusted to suit the selected crop type and the previous year's 26 climatic conditions. As an average for 2001-2011, the annual N-application was 136 kg N ha 27 1 yr⁻¹ at Faardrup, 139 kg N ha⁻¹yr⁻¹ at Silstrup, and 142 kg N ha⁻¹yr⁻¹ at Estrup (Table 3). 28 There were considerable differences in the average annual N-application, ranging from 0 kg N 29 ha⁻¹yr⁻¹ for the pea crop to 223 kg N ha⁻¹yr⁻¹ for spring barley undersown with red fescue. At 30 31 Faardrup only inorganic N fertilisers were used, whereas at Silstrup and Estrup animal slurry was applied from time to time (Table 3). 32

1 2.3 Monitoring setup

2 The specific growth stages for the different crops were characterized using the BBCH scale 3 (Meier, 2001), e.g., the BBCH growth stages for cereals: germination (00-09), leaf 4 development (10-19), tillering (20-29), steam elongation (30-49), inflorescence emergence 5 (51-59), flowering (61-69), development of fruit (71-77), ripening (83-89), and senescence 6 (92-99) (Meier, 2001). Precipitation was measured on site and air temperature was recorded at 7 nearby meteorological stations. The water table was registered in piezometers constructed of 8 6.3 cm diameter polyvinyl chloride with 0.5 m screens placed at depths of 3.0-3.5 m, 4.5-5.0 9 m, and 6.1-6.6 m at the edge of the Faardrup, Estrup, and Silstrup fields. Daily monitoring of 10 the water table at Silstrup and Estrup was performed using a D-Diver (Van Essen Instruments, 11 Delft, The Netherlands) and monthly monitoring of the water table at Faardrup was performed with a hand-held water level meter, type 010 (HT Hydrotechnik, Obergünzburg, Germany). 12 13 ISCO samplers (Teledyne ISCO, Lincoln, NE, USA) were used to collect samples of drainage 14 water. Drainage water was sampled time proportionally until July 2004 and then flow 15 proportionally, with sub-samples collected for every 3000 L of drainage during the winter season (September-May) and for every 1500 L during the summer season (June-August). 16 17 Each week, all the collected subsamples were pooled and a sample analysed in the laboratory.

18 **2.4 Nitrate-N analysis**

Samples of drainage were refrigerated at all times until analysis. The water samples were 0.45 µm filtrated (Millex HV syringe filter, Millipore, Ireland) and nitrate-N (NO₃-N) was measured using a Metrohm Anion system equipped with a Metrosep A Supp 15 – 250 IC column and a suppressor module (MSM) and with conductivity detection (Mehrohm, Herisau, Switzerland). The eluent was a mixture of 1 mM NaHCO₃ and 3.2 mM Na₂CO₃. The system was connected to an 838 Advanced IC Sample processer (Mehrohm, Herisau, Switzerland).

25 **2.5 Calculation of N-flux in drainage**

The total mass of nitrate-N transported out of the field by drainage was calculated by multiplying the concentration of nitrate-N for the pooled water sampled by the drainage volume between the time of sampling and the previous time of sampling. The nitrate-N losses were all calculated as kg N ha⁻¹, taking into account the different sizes of the three fields.

1 3 Results

2 **3.1 Climate**

3 All three fields are located in a temperate climate with average air temperatures between 8.8 4 °C and 9.0 °C for 2001-2011. During monitoring, the minimum (yearly) average temperatures were 5.6-5.8 °C and the maximum (yearly) average temperatures were 11.4-12.2 °C, with 5 summer temperatures up to 30.4-32.1 °C and winter temperatures down to -14.1 °C and -19.5 6 7 °C (Fig. 2 and Table 4). The geographical location of the fields led to differences in annual 8 precipitation. Average annual precipitation between 2001 and 2012 was 685 mm at Faardrup 9 and 943 mm at Silstrup, with the highest being Estrup at 1089 mm (Table 1). As the fields' 10 average annual evaporation only varied slightly (between 459 mm and 476 mm), the annual 11 leaching to drainage and groundwater was 226 mm at Faardrup, 471 mm at Silstrup, and 617 12 mm at Estrup (Table 1).

In 2001-2012, 49 %, 16 %, and 3.3 % of drainage at Faardrup was on days on which air temperatures were above 5 °C, 10 °C and 15 °C respectively (Fig. 2 and Table 5). At Silstrup 56 %, 12 %, and 0.3 % and at Estrup 58 %, 22 %, and 5.4 % of drainage occurred in the respective temperature categories (Fig. 2 and Table 5).

17 3.2 Hydrogeological setting

18 At Faardrup, monthly registration of the groundwater table at the edge of the field showed the 19 overall pattern over the year, even though its full amplitude was not monitored as the filter 20 had not been installed sufficiently deeply to register deepest water tables (Fig. 3). The water 21 table approached the soil surface by the end of autumn and during winter, and dropped in spring, summer and early autumn to about 3.5 m depth or even deeper. During 2001-2012, the 22 average daily drainage was 0.26 mm d⁻¹ and max daily drainage was 14.9 mm d⁻¹ during an 23 average 88 days of drainage per year (Table 5). Typically, drainage was below 3 mm d⁻¹ and 24 rarely above 10 mm d⁻¹ (Fig. 3). For 2001-2012, the average annual drainage was 43 % of 25 26 total discharge to drainage plus groundwater (Table 1).

At Silstrup, the groundwater table was above drainage depth from late autumn to late winter and dropped during spring and summer to a depth of 3.82 m at its lowest (October 2009) (Fig. 3). During 2001-2011, the average daily drainage was 0.50 mm d⁻¹ and max daily drainage was 27.8 mm d⁻¹ during an average 86 days of drainage (Table 5). Often, the drainage events were above 5 mm d⁻¹, a few were up to 10 mm d⁻¹, with a maximum drainage of 28 mm d⁻¹ in March 2010 (Fig. 3). For 2001-2012 the average annual drainage was 40 % of
total discharge to drainage plus groundwater (Table 1).

In Estrup, the groundwater table was highly dynamic and only located deeply for short periods. The lowest groundwater table (4.35 m) was in July 2008 (Fig. 3). On average for 2001-2012, the average daily drainage was 1.16 mm d⁻¹ and max daily drainage was 57.8 mm d⁻¹ during an annual average 243 days of drainage (Table 5). Daily drainage was often above 5-6 mm d⁻¹, in some cases up to 10 mm d⁻¹, and the highest amounts registered were 35 mm d⁻¹ in March 2005 and 58 mm d⁻¹ in February 2010 (Fig. 3). For 2001-2012 the average annual drainage was 68 % of total discharge to drainage plus groundwater (Table 1).

10 **3.3 Management - N fertilisers and crop types**

The time of application and amount of N fertilisers applied to the crops were in line with present legislation (concerning application of slurry) and the type of crop (concerning application of slurry and inorganic N fertiliser) (Ministry of Food, Agriculture and Fisheries, The Danish AgriFish Agency, http://agrifish.dk/).

15 At the Faardrup field only inorganic N fertiliser was used (Table 3). The time of application for the different crops is indicated in Fig. 4. For sugar beet, spring barley and 16 17 maize, N fertiliser was applied immediately before or after sowing. N fertiliser was applied 18 twice during the growing season for winter cover crops: for winter wheat the first occasion 19 was at about growth stage BBCH 12-26 and the second occasion was at about BBCH stage 20 23-32, while for winter rape it was at around sowing time and at growth stage 14-37. The red 21 fescue received small amounts of fertiliser just after the spring barley was harvested and at the 22 beginning of the growing season. The application of N fertilisers was not reflected in 23 simultaneous/subsequent increases in nitrate-N concentrations in the drainage. A total of 16 applications of N were performed. 24

The time of N-application at Silstrup was as described for Faardrup. A crop of pea in 2001 received no N fertilisation (Table 3 and Fig. 4). At Silstrup inorganic N fertiliser, injected slurry (pig/cattle) or a combination of inorganic N fertiliser and slurry were applied to the crops. Application took place at BBCH stage 20-30 for both winter wheat (2004 and 2007) and red fescue (2010 and 2011). In 2001-2012, the application of inorganic N fertiliser (in total 13 times) and slurry (in total seven times) was not reflected in immediate increased Nconcentrations in the drainage, except for fodder beet in 2008.

32 At Estrup, inorganic N fertiliser, injected slurry (cattle/sow/pig) or a combination of the 33 two was applied to all the crops except the pea crop (Table 3 and Fig. 4). Application of fertilisers was performed at BBCH 20-30 for winter wheat and at about BBCH 30 for winter rape. In total, inorganic N fertiliser and slurry were applied 12 times and seven times respectively. In seven of the 11 years (2003, 2005, 2006, 2007, 2008, 2009 and 2010) the application of N fertilisers was reflected in short-term increases (maximum concentration of 47 mg N L^{-1} in 2007) in drainage from the field.

6 **3.4 Nitrate concentrations**

In Faardrup, the nitrate-N concentrations in drainage were often well above the European 7 limit for drinking water supply (11.3 mg N L⁻¹) (Fig. 2). The drainage signature was 8 characterised by a large range of nitrate-N concentrations (3-34 mg N L⁻¹). Prolonged high 9 nitrate-N concentrations were measured below bare soil after harvesting winter wheat in 10 11 2000/2001, 2004/2005 and 2009, and after harvesting maize in 2006. Increasing nitrate-N 12 concentrations were measured up to a BBCH growth stage of about 30 for winter rape (2002/2003, 2006/2007) and winter wheat (2007/2008). At later BBCH stages and up to 13 14 harvest, nitrate-N concentrations decreased. Decreasing nitrate-N concentrations were also observed during the growing seasons of sugar beet (2001), spring barley (2002, 2006), maize 15 (2005) and red fescue (2010/2011). 16

In 2001-2012, the nitrate-N concentrations in drainage at Silstrup varied between 1-34 mg 17 N L⁻¹ and rarely exceeded the European limit for drinking water (Fig. 2). Prolonged elevated 18 19 nitrate-N concentrations were measured below bare soil after fodder beet (2000/2001, 20 2008/2009), spring barley (2001/2002), maize (2002/2003) and winter wheat (2004/2005, 2007/2008), whereas the nitrate-N concentrations in drainage after red fescue remained at a 21 constant low level (1-2 mg N L⁻¹) (Fig. 2). Remarkably high (maximum 22 mg N L⁻¹) nitrate-22 N concentrations, well above the EU limit for drinking water, were recorded only following 23 the winter wheat crop (2003/2004). 24

Except for some short-term peak concentrations, the N-concentration in drainage at Estrup was below the EU limit for drinking water (Fig. 2). This also applied to the periods with bare soil, where the nitrate-N concentrations remained almost constant, except following spring barley (2004/2005) when concentrations decreased, and after winter wheat (2011/2012) when they increased. Concentrations of nitrate-N decreased during the growing seasons of fodder beet (2003), winter wheat (2007) and winter rape (2009).

1 **3.5 Nitrate-N fluxes**

2 At Faardrup, short-term N-leaching events, often in winter (or the start of the year), with daily fluxes of nitrate-N below 1 kg N ha⁻¹d⁻¹ were common (Fig. 2). Some leaching events were in 3 the range of 0.5-1 kg N ha⁻¹d⁻¹ and very few events were in the range of 1-4 kg N ha⁻¹d⁻¹. The 4 5 nitrate-flux signature was highly related to the drainage events, as indicated by the step-like shape in 2002/2003, 2005 and 2011 (Fig. 5) and a good correlation ($R^2 = 0.83$) between 6 7 cumulated nitrate flux and drainage intensity in the time between water sampling (Fig. 6). In this field, annual nitrate fluxes varied between 3 and 24 kg N ha⁻¹yr⁻¹ and made up the 8 9 equivalent of 2-19 % of the annual applied N fertilisers (Fig. 7). The total nitrate flux for 2001-2012 in drainage was 142 kg N ha⁻¹ and in this period 1493 kg N ha⁻¹ was applied as N 10 11 fertilisers (Table 3).

12 Most nitrate-N leaching events in 2001-2012 at Silstrup took place during the autumn and winter, and often the daily fluxes were below 1 kg N ha⁻¹d⁻¹. Only a few were within the range 13 of 1-2 kg N ha⁻¹d⁻¹ and were rarely above 2 kg N ha⁻¹d⁻¹ (Fig. 2). Large and lasting drainage 14 (2002, 2004, 2006/2007, 2008 and 2009/2010) coincided with high N-fluxes out of the field 15 (Fig. 5), and increasing N-fluxes with increasing drainage cumulated for the time interval 16 between sampling ($R^2=0.60$) (Fig 6). The annual export of nitrate-N from the field with 17 18 drainage varied between 3-32 kg N ha⁻¹yr⁻¹, equivalent to 2-33 % of the annual applied N 19 fertilisers (inorganic N fertilisers and slurry) (Fig. 7). For the period 2001-2012 the total nitrate-N leaching from the field was 153 kg N ha⁻¹, as compared to the total application of 20 1524 kg N ha⁻¹ (Table 3). 21

22 Leaching from Estrup was present throughout the year, often with daily nitrate-N fluxes of below 1 kg N ha⁻¹d⁻¹, and only above 1 kg N ha⁻¹d⁻¹ during a few events (Fig. 2) and the 23 correlation between the N-fluxes and drainage, cumulated between sampling, was $R^2=0.55$ 24 (Fig. 6). The drainage signature at Estrup, with a long drainage period and renewable pool of 25 crop-generated available organic matter with low nitrate fluxes, made the cumulated N-26 leaching curve very smooth, with only a few small steps (Fig. 5). The annual nitrate-N 27 leaching was 13-32 kg N ha⁻¹yr⁻¹, the equivalent of 8-22 % of the total amount of N applied to 28 the field (Fig. 7). In 2001-2012 the total loss with drainage was 205 kg N ha⁻¹ and the total N-29 application was 1563 kg N ha⁻¹ (Table 3). 30

1 4 Discussion

2 4.1 Climate

3 For 2001-2012, the average air temperature at the three fields was approximately the same 4 (8.8–9.0 °C) throughout the years studied. The maximum temperatures (30-32 °C) were measured in June to August and the minimum (-14.1 - -19.5°C) in December to February. 5 6 Even though the average air temperatures varied only slightly between the three fields, air 7 temperature during drainage was markedly different. At Faardrup, an often late drainage 8 resulted in 51 % run-off at temperatures below 5 °C (below biological "zero") whereas at 9 Silstrup and Estrup most drainage (56 % and 58 %) was at temperatures above 5°C. At 10 Estrup, with drainage taking place during about two-thirds of the year, 5.4 % of drainage was 11 at temperatures above >15 °C. The field-specific temperature-drainage signature provides 12 valuable input to future water management, including on-field and off-field N regulation.

13 The almost identical temperature regimes made evaporation vary only slightly between 14 the three fields, and hence groundwater recharge was lowest in Faardrup where there was the 15 least precipitation. The low groundwater recharge at Faardrup generated a higher 16 concentration of nitrate-N in water leaving the root zone from the different crops and from 17 bare soil than at the two other fields in Silstrup and Estrup. Due to the low recharge and 18 despite Faardrup receiving the lowest total amount of N as well as in a form readily available 19 to plants, concentrations of nitrate-N in drainage from this field remained highest. At Silstrup 20 and Estrup much higher precipitation managed to keep the average concentration of nitrate-N 21 below the EU limit for drinking water.

22 4.2 Hydrological setting

23 The drainage signatures were significantly different at the three fields, with each 24 representative of its own type due to the local climatic and hydrological conditions. At Faardrup, where there is low precipitation and recharge and a long-lasting deeply located 25 groundwater table, drainage runoff was short-lived (on average 88 days year⁻¹), commenced 26 late (middle of winter), was at the lowest air temperature (51 % at <5 °C) and was of low 27 intensity (average 0.26 mm d⁻¹). Compared to the other two fields, Silstrup had a medium 28 precipitation and discharge. The groundwater table was located deep for a long period of time. 29 30 whereas drainage (on average 86 days year⁻¹) often started in autumn at higher temperatures (56 % at >5 °C), and was of a higher intensity (average 0.50 mm d⁻¹). At Estrup, where there 31 is the highest precipitation and recharge as well as a shallow and highly dynamic ground 32

water table, drainage occurred most of the year (on average 243 days year⁻¹), and often at high 1 2 intensity (1.16 mm d^{-1}). The different drainage signatures also led to differences in the total nitrate-N leaching for 2001-2012, with the least drainage leaching at Faardrup with 142 kg N 3 ha⁻¹, medium leaching at Silstrup with 153 kg N ha⁻¹, and most leaching at Estrup with 205 kg 4 N ha⁻¹. Drainage mainly occurred when a water table was close to the surface. Drainage was 5 6 only observed a few times at Silstrup (e.g. July 2001 and 2002, May 2003 and October 2007), 7 when the groundwater table was far below the drainage depth, due to preferential flow. This 8 flow may rapidly take chemicals from the surface to the tile drain system (Kladivko et al., 9 1999). The combination of site-specific climatic and hydrological conditions resulted in far 10 greater drainage at Estrup (average 68 % in 2001-2012) than at Faardrup (40 %) and Silstrup 11 (43 %), providing the greatest input to groundwater in the latter two fields.

12 Besides the transport of nitrate-N in drainage out of the fields, the prevailing hydrological 13 conditions also strongly influenced the potential for denitrification of nitrate-N released from 14 decaying material such as plant parts and bacteria and other soil fauna and flora. The shallow water table at Estrup for two thirds of the year, together with higher temperatures (58 % at >5 15 16 °C), made the potential for in-situ denitrification much greater here than at Faardrup and 17 Silstrup, where a deep water table and vadose conditions impaired the presence of the 18 oxygen-free conditions essential for the denitrification process to occur. Under these 19 conditions, in-situ denitrification would be limited to oxygen-free micro-environments.

20 4.3 N fertilisers

21 Only at Estrup, with its shallow and highly dynamic groundwater table, were the applications 22 of N fertilisers (inorganic N fertilisers as well as slurry) reflected in the concentration of 23 nitrate-N, but only as a short-term increase in the nitrate-N concentration after application. Even though the applications of N fertilisers at Faardrup and Silstrup were performed at 24 25 similar plant growth stages, no immediate effects on nitrate-N concentrations were observed. 26 This applied to both inorganic N fertilisers and injected slurry. This may be because most of 27 the N applications were performed just around the time of sowing or at the beginning of the 28 growing season when crops efficiently use up available nitrate-N. Gentry et al. (2014) have 29 found that changing from autumn to spring N-application for maize improves water quality in 30 tiled-drained catchments.

It seems that current regulations concerning the timing of N application – often in spring
 - have helped to reduce the immediate impact of nitrate on drainage quality at fields like

Faardrup and Silstrup. At Estrup, however, leaching of nitrate could be reduced further if the
time for N application is aligned with the weather conditions so that high intensity rainfall is
avoided just after application.

4 **4.4 Crop types**

5 The results from all three fields confirmed that the choice of crops and crop rotation had an 6 effect on the leaching of nitrate-N into drainage, primarily due to their different growth 7 periods. Thus it is clear that crops with a long growing season (sugar beet, red fescue, winter 8 rape and winter wheat) help to reduce the concentration of nitrate-N compared to crops with a 9 short growing season (maize, spring barley and pea) and thereby leave the land without vegetation for a longer period of time. Red fescue used the applied N fertilisers most 10 efficiently, and thereby lowered the concentration of nitrate-N to about 1 mg N L⁻¹. Since 11 crops with a short growing period leave soil bare (without winter cover crops or catch crops) 12 13 at high air (and soil) temperatures, this favours mineralisation and the release of nitrate-N that 14 can percolate to the tile drain system or groundwater in winter months (Premrov et al., 2014). 15 Pea in rotation with wheat at Estrup seemed not to increase the concentration of nitrate-N at Estrup (2001/2002), whereas at Silstrup the concentrations increased markedly (2003/2004), 16 17 which was similar to observations by Beaudoin et al. (2005).

18 **4.5** Nitrate-N concentrations

19 The nitrate-N concentrations in drainage exhibited annual and seasonal variability over the 11-year monitoring period. The final nitrate-N concentrations in drainage were governed by 20 21 multiple factors, of which climate conditions (precipitation and evaporation) and types of crop were the most significant. Thus, the low precipitation and percolation at Faardrup seemed to 22 23 be the governing factors behind the field often having the highest nitrate-N concentration in 24 the drainage even though it had the lowest application of N fertilisers. Nitrate-N 25 concentrations at Faardrup for most crop types exceeded the EU limit for drinking water. 26 Profitable crop production at reduced input of N fertilisers may be very difficult in these low 27 rainfall areas, but the right choice of crops combined with growing of catch crops may be a solution for optimal N-management of such fields. The net outcome of catch crops, however, 28 may lead to lower N-fluxes in drainage but higher nitrate concentrations due to the catch crop 29 30 water uptake and hence lower ground water infiltration.

The monitoring highlighted that crop rotation with crops that have long growing seasons, *e.g.* red fescue, winter rape and winter wheat, efficiently reduced the pool-leachable nitrate-N. Reduced nitrate-N loss in drainage with cover crops has also been recorded by Drury *et al.* (2014) when planting a winter wheat cover crop in a cool, humid agricultural soil. The concentrations of nitrate-N at Silstrup and Estrup were below the average concentration of 18 mg N L⁻¹ measured in drainage from 15 systematic tile-drained Danish agricultural clayey till areas (3-22 ha) in 1971-1981 (Hansen and Pedersen, 1975; Pedersen, 1983).

6 Unlike at Faardrup, the nitrate-N concentrations at Silstrup and Estrup were often below 7 the EU drinking water limit. Even though the lowest overall nitrate-N concentrations were 8 recorded at Estrup (highest precipitation), the lowest nitrate-N concentrations were measured 9 at Silstrup under a crop of red fescue that efficiently took up all nitrate-N available.

10 **4.6 Nitrate-N fluxes**

11 The results from 2001-2012 showed that the nitrate-N losses were the product of nitrate-N 12 concentrations in the flowing water, and that the amount of water (drainage) was essential for the total impact on the aquatic environment. Leaching of nitrate-N from Faardrup and Silstrup 13 14 with a deeply located groundwater table during the summer was concurrent with days of high drainage. At Estrup, where the groundwater table is closer to the surface, transport of nitrate-15 N to the aquatic environment was almost continuous. The annual nitrate-N losses to drainage 16 ranged from 3 to 32 kg N ha⁻¹yr⁻¹, with the lowest after winter rape at Faardrup and the 17 18 highest after winter wheat and rape at Silstrup and Estrup, respectively. However, despite 19 major differences in climate, hydrological pattern and amounts of N fertilisers applied, the average nitrate-N losses for 2001-2011 were between 13 and 19 kg N ha⁻¹, equivalent to 10-20 21 13 % of the amount applied, with the highest losses at Estrup. This was below the 25 % found in a long-term study by Lucey and Goolsby (1993). The losses in kg N ha⁻¹ were within the 22 range of 10 and 29 kg N ha⁻¹ reported for 15 Danish clayey till agricultural areas monitored 23 between 1971 and 1980 (Pedersen, 1983). The average nitrate-N loss for the 15 areas was 22 24 N ha⁻¹yr⁻¹ and therefore higher than the 15 kg N ha⁻¹yr⁻¹ on average for the three fields in the 25 present study. The annual nitrate-N leaching at the three fields fell within the typical range for 26 27 most European and North American research studies, even though nitrate-N leaching in some studies has reached up to 100 kg N ha⁻¹ (Randall *et al.*, 2008) and up to 105 kg N ha⁻¹ for 28 poorly drained loess soils (loam soils) in Indiana, USA (Kladivko et al., 1999). 29

30 Due to longer periods of high water saturation of the layers close to the surface, including 31 at times of high air (and soil) temperatures and renewable inputs of surface-derived 32 bioavailable organic carbon in the top two metres (Pabich *et al.*, 2001) *e.g.* from injected slurry, crop material and roots, it was expected that the *in situ* denitrification process would
 be more efficient for nitrate-N attenuation at Estrup than at Faardrup or Silstrup, where most
 of the year the groundwater table is down to around a depth of four metres.

4 **5** Summary and conclusions

5 To the authors' knowledge this study presents the first long-term monitoring (11 years) of 6 nitrate-N concentrations and fluxes in drainage from well-defined tile-drained fields in 7 Denmark. Data collected simultaneously on nitrate-N, drainage, climate (precipitation and air 8 temperature), ground water table, crops (types and growth stage) and N fertilisers (type and 9 time of application) were collected in 2001-2012 at three fields across Denmark. The annual 10 average air temperature and evaporation were around the same in all fields, but the average annual precipitation varied between 685 mm y^{-1} (Faardrup) at the lowest to 1089 mm y^{-1} at the 11 highest (Estrup). Major differences in drainage nitrate-N concentrations were identified. Most 12 13 often the highest concentrations were measured in drainage from the low precipitation field, 14 where concentrations were often above the European limit for drinking water, irrespective of 15 whether the field was with or without crop cover. At the two other fields the nitrateconcentrations in drainage often remained below the EU limit. However, different types of 16 17 crops strongly influenced N-concentrations, with the lowest nitrate-N concentrations during and after red fescue and the highest concentrations after a pea crop or crops with a short 18 19 growing season followed by bare soil. Nitrate-N fluxes out of the fields were primarily 20 controlled by the site-specific hydrological setting and only to a minor extent by the nitrate-N 21 content in the drainage. The nitrate-N flux signature of the two fields with deeply located groundwater tables at times and only short and medium-length drainage periods showed 22 23 major nitrate-N leaching events that were concurrent with intense drainage, whereas more 24 even nitrate-N fluxes were obtained from the field with longer drainage periods. The total 25 impact on the aquatic environment in 2001-2012 due to drainage losses varied between 142 and 205 kg N ha⁻¹yr⁻¹, equivalent to 10-13 % of the amount of N fertilisers applied. In fields 26 27 with a shallow vadose zone as well as lengthy periods of water saturation, a renewable source 28 of organic matter from crop residues may make the denitrification processes much more 29 efficient, and thereby contribute to lower nitrate-N concentrations and nitrate-N fluxes in 30 drainage. The time at which drainage out of the field occurs may also influence the potential efficiency of water management actions off-field, e.g. in natural or small constructed wetlands 31 32 where the effectiveness due to microbial processes and plant growth is highly temperature dependent. Here, as at Estrup, long-term leaching of nitrate-N, including at higher 33

temperatures, presented the best possibilities for off-field water management actions to reduce 1 2 the impact of nitrate, whereas the short-term leaching of nitrate at Faardrup at low temperatures presented a limited potential of on-field or off-field water management actions 3 4 due to the very short period of water saturation at low temperatures. The documented 5 differences in nitrate-N fluxes and concentrations at a local scale reveal some of the future challenges that need to be addressed when regulating for N fertilisers at field scale, such as in 6 7 the regulation already proposed. Future N-regulation cannot be based on a single factor, but 8 has to take a combination of field-specific factors into consideration, such as the inherent 9 physical appearance and agricultural management of the fields. The long-term monitoring has 10 revealed that the outcome of on-field or off-field water management actions could differ 11 between fields with different hydrogeological settings and climatic conditions. This initial 12 well-documented field-scale knowledge from three clayey till fields that are representative of 13 large areas in Denmark is a first step towards establishing a differentiated N-regulation for 14 such areas. A future challenge will be to obtain the necessary knowledge to cover all field 15 types represented within a catchment (van der Velde et al., 2010).

16 Acknowledgements

This study was funded by the Danish Pesticide Leaching Assessment Programme. The authors would like to thank the many people who have participated in the programme over the past 11 years, especially Finn Plauborg and Finn Christensen for providing climate data, Ulla Husballe Rasmussen and Mette Ejsing-Duun for meticulous analytical work, and Jens Barsballe, Poul Boesen, Hennrik Bruun, Lasse Gudmundsson, Kristine Riis Hansen, Søren Have Jepsen, Jens Molbo, Niels Peter Pedersen and Henning C. Thomsen who collected the various types of data.

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Table 1. Precipitation (P), evaporation (E), drainage (D), and groundwater discharge (G) for

2 the agrohydrological year (June-June) for 2001-2012 for the Faardrup, Silstrup and Estrup

Field	Р	E	D	G	D+G	D/(D+G)	D/P
			mm y ⁻¹			%	
Faardrup	685	459	95	131	226	43	13
Silstrup	943	472	188	283	471	40	20
Estrup	1089	476	421	196	617	68	39

3 fields (partly after Lindhardt *et al.*, 2001).

Field	Faardrup	Silstrup	Estrup	
Size of field (ha)	2.3	1.7	1.3	
Slope (%)	1-3	1-2	0-1	
Soil type (USDA)	Argiudoll	Argiudoll	Argiudoll	
	Hapludoll	Hapludoll	Glossudalf	
	Vermudoll			
Drainage status	Moderately	Moderately	Moderately	
	well drained	well drained	well drained	
Depth of tile drains (m)	1.1	1.1	1.1	
C in Ap (0-0.2 m) (%)	1.4-1.5	1.6-2.0	1.6-3.2	
C below Ap (0.2-6 m) (%)	0.06-0.23	0.06-2.1	0.1-50	
Clay in Ap (0-0.2 m) (%)	15-16	18-27	10-20	
Clay below Ap (0.2-6 m) (%)	16-37	18-58	1-65	
Depth to calcareous zone (m)	1.5	1.1	1-4	
CaCO ₃ (%) (0-6 m)	0-21	0-46	0-82	
Age of sediments	Late Weichselian	Late Weichselian	Saalian glaciation	
	glaciation	glaciation		
Landscape	Till plain	Till plain	Till plain	
Geology	Homogenous	Dislocated	Complex structure	
		structure		

Table 2. Key parameters for the three fields according to Lindhardt *et al.* (2001) and
 Rosenbom *et al.* (2015).

Table 3. Annual average of minimum, mean, and maximum air temperatures for 2001-2011

2 at Faardrup, Silstrup and Estrup.

Temperature	Faardrup	Silstrup	Estrup
Average minimum air temperature (°C)	5.6	6.0	5.8
Minimum air temperature (°C)	-19.5	-14.5	-15.7
Average maximum air temperature (°C)	12.0	11.4	12.2
Maximum air temperature (°C)	31.3	30.4	32.1
Average mean air temperature (°C)	8.8	8.7	9.0

Table 4. Crops and winter cover crops (in bold), crop rotation, types of N-source (F:
 inorganic fertilisers; C: cattle manure, S: sow manure and P: pig manure), and the amount of
 N fertilisers (kg N ha⁻¹yr⁻¹) applied in 2001-2012 at Faardrup, Silstrup and Estrup.

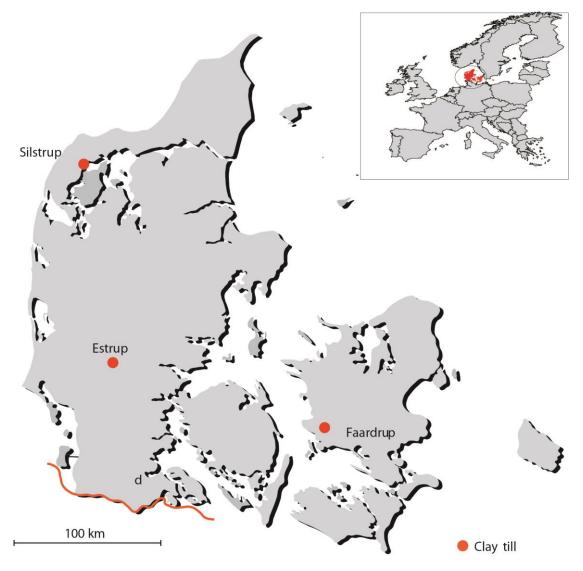
	Faardrup			Silstrup			Estrup		
Year	Crop	Source	Dose kg N ha ⁻¹ yr ⁻¹	Crop	Source	Dose kg N ha ⁻¹ yr ⁻¹	Crop	Source	Dose kg N ha ⁻¹ yr ⁻¹
2001	Sugar beet	F	110	Spring barley	F	118	Peas Winter wheat	-	0
2002	Spring barley Winter rape	F	125	Maize	F,C	163	Winter wheat	F	147
2003	Winter rape Winter wheat	F	145	Peas Winter wheat	-	0	Fodder beet	С	169
2004	Winter wheat	F	154	Winter wheat	F	170	Spring barley	F	105
2005	Maize	F	129	Spring barley Winter rape	F, P	167	Maize	F, S	164
2006	Spring barley Winter rape	F	130	Winter rape Winter wheat	F, P	96	Spring barley Winter wheat	F	112
2007	Winter rape Winter wheat	F	151	Winter wheat	F	162	Winter wheat Winter wheat	F	178
2008	Winter wheat	F	156	Fodder beet	F	244	Winter wheat	F	180
2009	Sugar beet	F	110	Spring barley Red fescue	F, P	223	Spring barley 223 Winter wheat		167
2010	Spring barley Red fescue	F	179	Red fescue	F	58	Winter rape Winter wheat	F	181
2011	Red fescue	F	104	Red fescue	F, P	123	Winter wheat	F, S	160
Total			1493			1524			1563
Yearly average			136			139			142

Table 5. Minimum (min.), maximum (max.) and average (avg.) daily drainage, average

annual number of days with drainage, average air temperatures during drainage, and

3 cu	mulated drainage,	all 2001-2012 at Faardrup	o, Silstrup and Estrup.
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	Drainage				Average air temperature (°C)				Drainage
Field	min.	max.	avg.	avg.	<5	≥5	>10	>15	2001-2011
	$\operatorname{mm} d^{-1}$ d		days yr ⁻¹	% (of cumul	mm			
Faardrup	0.1	14.9	0.26	88	51	49	16	3.3	961
Silstrup	0.1	27.8	0.50	86	44	56	12	0.3	2304
Estrup	0.1	57.8	1.16	243	42	58	22	5.4	4921



- **Figure 1.** Map of Denmark showing the three clayey till fields: Faardrup, Silstrup and Estrup
- 3 (Lindhardt *et al.*, 2001).

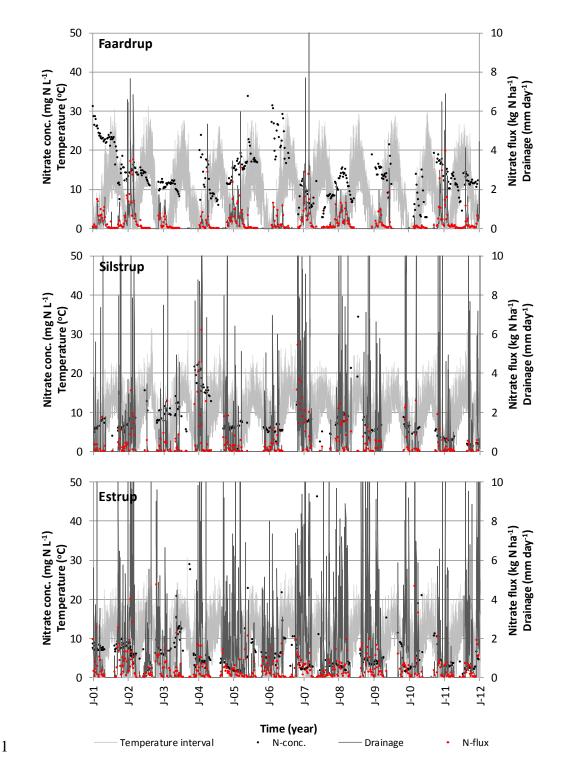


Figure 2. Concentration of nitrate-N, nitrate-N fluxes, drainage and the daily minimummaximum temperature span for temperatures above zero degrees Celsius for 2001-2012 at
Faardrup, Silstrup and Estrup.

- 5
- _
- 6

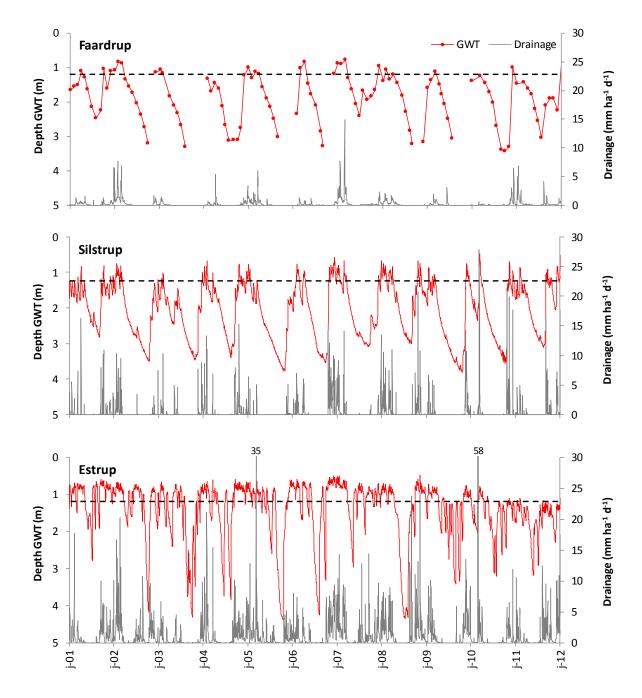


Figure 3. Drainage and depth to groundwater table (GWT) in 2001-2012 at Faardrup, Silstrup
and Estrup. The black dashed lines indicate tile drain depth.

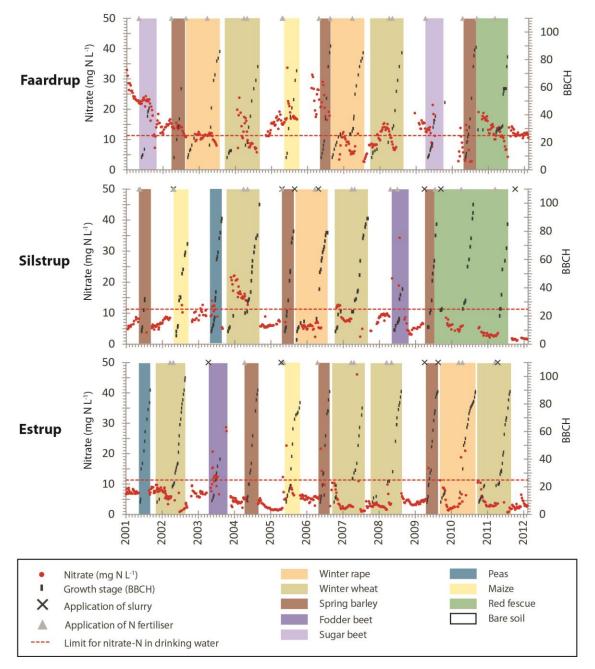


Figure 4. Crop type, specific growth stage of crops according to the BBCH scale, application
of inorganic N fertiliser and injection of slurry and concentration of nitrate-N (mg N L⁻¹) in
drainage at Faardrup, Silstrup and Estrup.

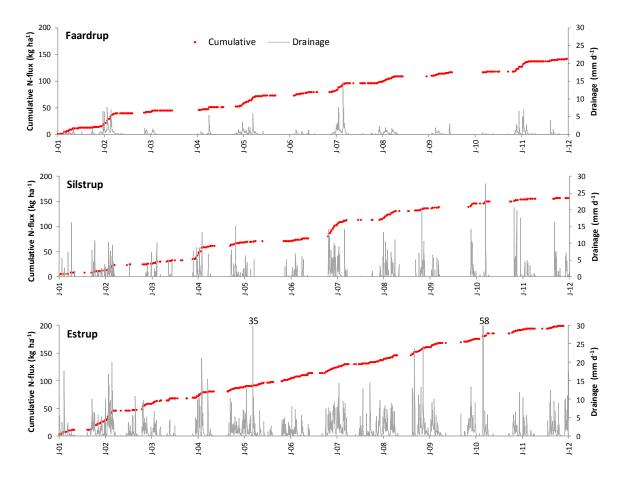


Figure 5. Cumulative nitrate-N flux and drainage for 2001-2012 at Faardrup, Silstrup and
Estrup.

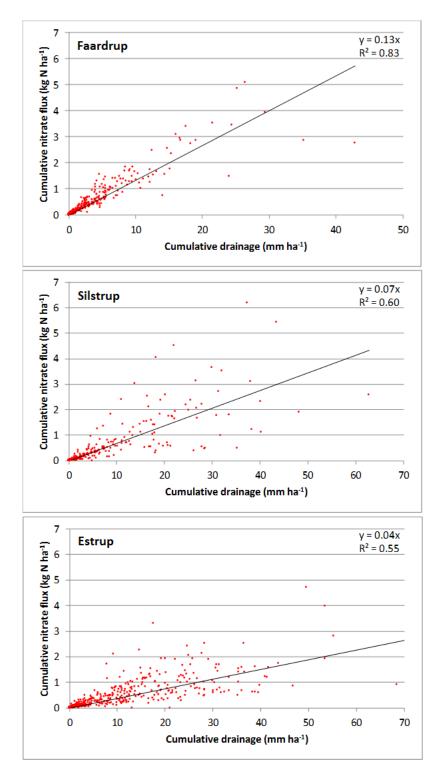
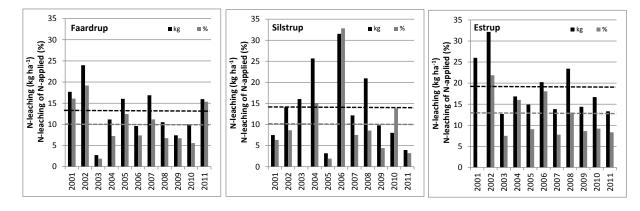




Figure 6. Nitrate-N flux per cumulated drainage for each sampling period at Faardrup,
Silstrup and Estrup.



1

2 Figure 7. Nitrate-N in drainage in mass (kg N ha⁻¹) and of applied N (%) (inorganic N

3 fertilisers plus slurry) for the years 2001 to 2011 at Faardrup, Silstrup and Estrup. The black

4 dashed line indicates the average annual N-leaching in 2001-2012 and the grey dashed line is

5 the average nitrate-N leaching of applied N in 2001-2012.