

## Supplementary material

### S1. Volatilization results

Simulated N volatilization for one field in Aurajoki catchment was  $13 \text{ kg ha}^{-1} \text{ a}^{-1}$ , which was 67% of the surface applied  $\text{NH}_4^+$  in manure (Figure S1a). At the Aurajoki catchment scale (9618 field plots), mean N volatilization was  $7.7 \text{ kg ha}^{-1} \text{ a}^{-1}$  (including also N volatilization from mineral N fertilizer), and surface applied N in manure was  $18 \text{ kg ha}^{-1} \text{ a}^{-1}$  (Figure S1b).

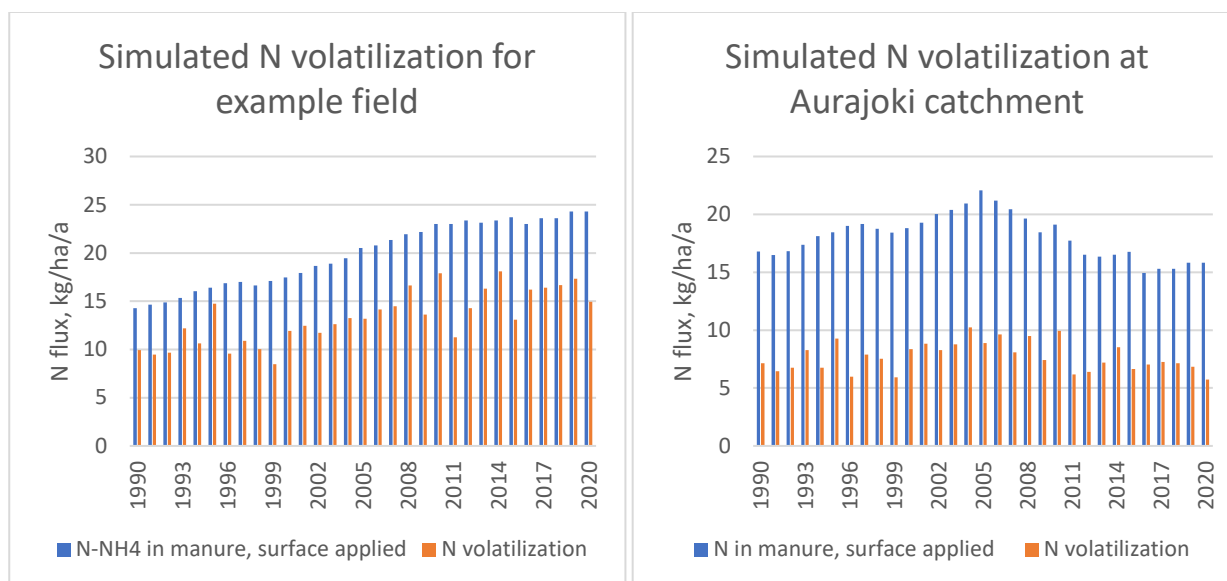


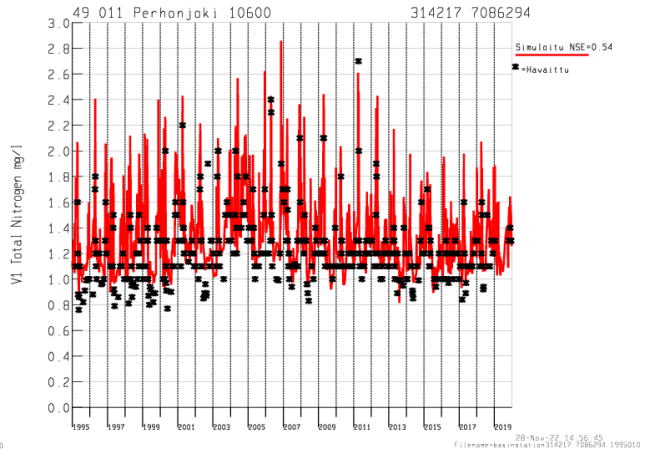
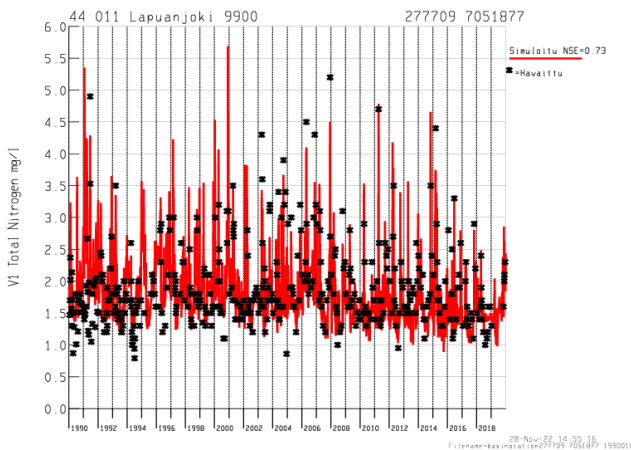
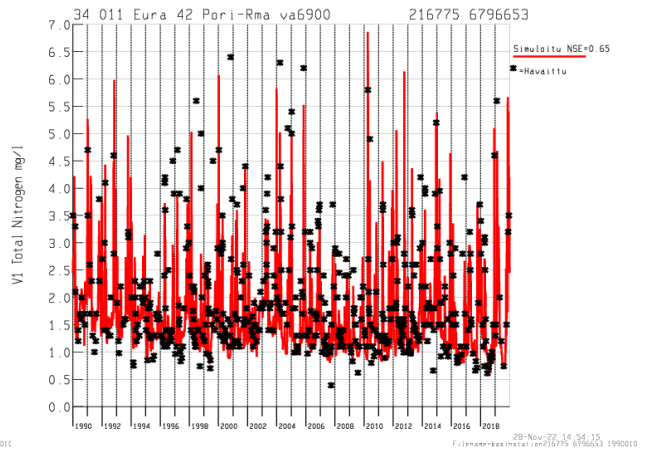
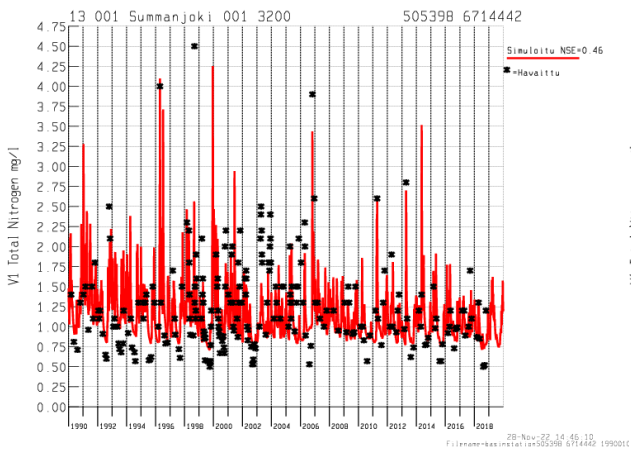
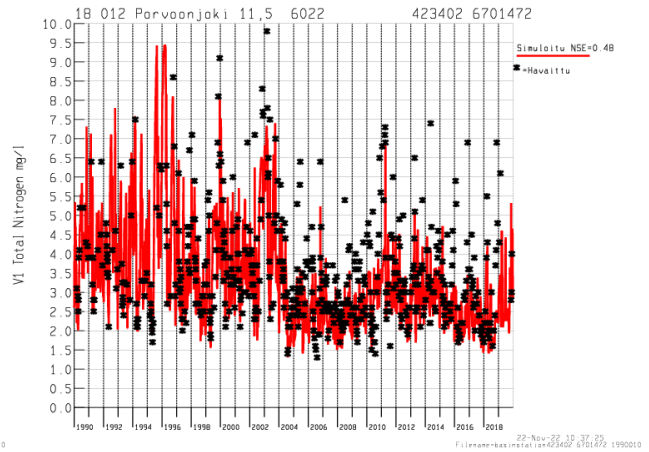
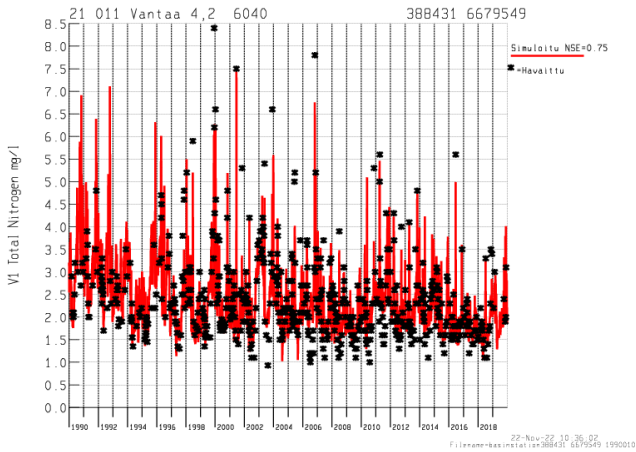
Figure S1. a) **N-NH<sub>4</sub> in manure**, surface applied and N volatilization for one example field (Aurajoki catchment), b) **N in manure** surface applied and N volatilization, mean for Aurajoki catchment fields.

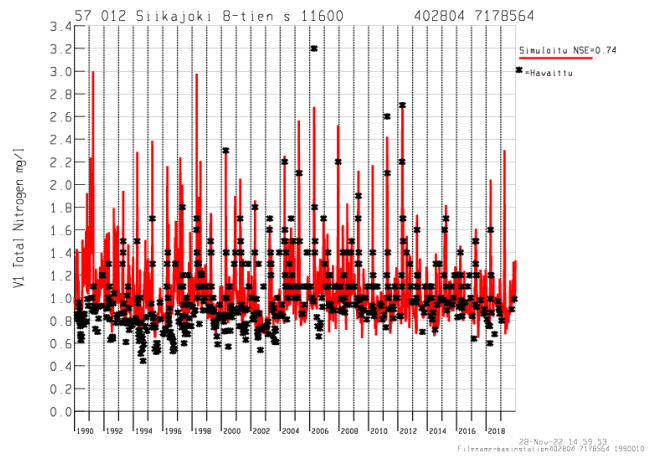
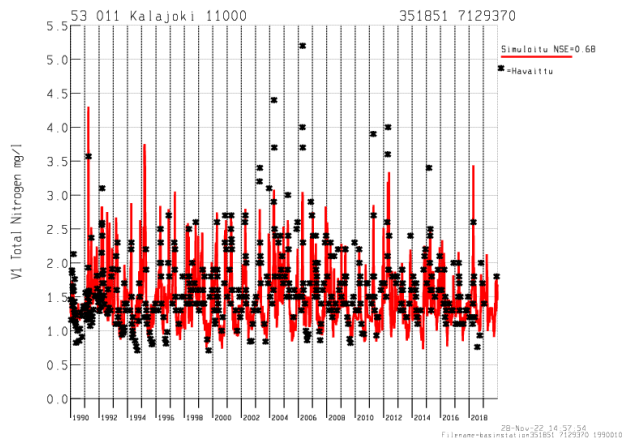
### S2. Simulated and observed mean TN concentration, Nash and Sutcliffe efficiency (NSE) criteria and source apportionment (as % of total loading) (2013-2020) for selected river points

| Observation point         | ELY centre         | Number of observations | Observed concentration, mg/l | Simulated concentration, mg/l | Difference, mg/l | NSE  | Agricultural loading | Forestry loading | Natural background loading | Scattered settlements | Loading with urban runoff | Deposition to water bodies | Point load |
|---------------------------|--------------------|------------------------|------------------------------|-------------------------------|------------------|------|----------------------|------------------|----------------------------|-----------------------|---------------------------|----------------------------|------------|
| 28_001 Aurajoki outlet    | South west Finland | 233                    | 2.66                         | 2.54                          | 0.11             | 0.59 | 73.8                 | 1.9              | 17.8                       | 1.6                   | 3.1                       | 0.3                        | 1.5        |
| 21_011 Vantaanjoki outlet | Uusimaa            | 147                    | 2.06                         | 2.06                          | 0                | 0.75 | 43.2                 | 1.9              | 28.6                       | 2.7                   | 9.2                       | 1.1                        | 13.2       |
| 18_012 Porvoonjoki outlet | Uusimaa            | 146                    | 3.14                         | 2.64                          | 0.5              | 0.48 | 52.6                 | 1.2              | 19.2                       | 1.8                   | 3.5                       | 0.6                        | 21.2       |
| 13_001 Summanjoki outlet  | South east Finland | 41                     | 1.02                         | 1.06                          | -0.04            | 0.46 | 34.8                 | 5.4              | 53.0                       | 1.4                   | 2.4                       | 1.6                        | 1.4        |
| 34_011 Eurajoki outlet    | Satakunta          | 138                    | 1.9                          | 1.66                          | 0.24             | 0.65 | 67.7                 | 1.9              | 15.9                       | 1.9                   | 2.8                       | 2.9                        | 6.9        |
| 44_011 Lapuanjoki outlet  | South Ostrobothnia | 100                    | 1.81                         | 1.7                           | 0.11             | 0.73 | 57.7                 | 5.9              | 24.7                       | 1.7                   | 2.6                       | 1.4                        | 6.1        |

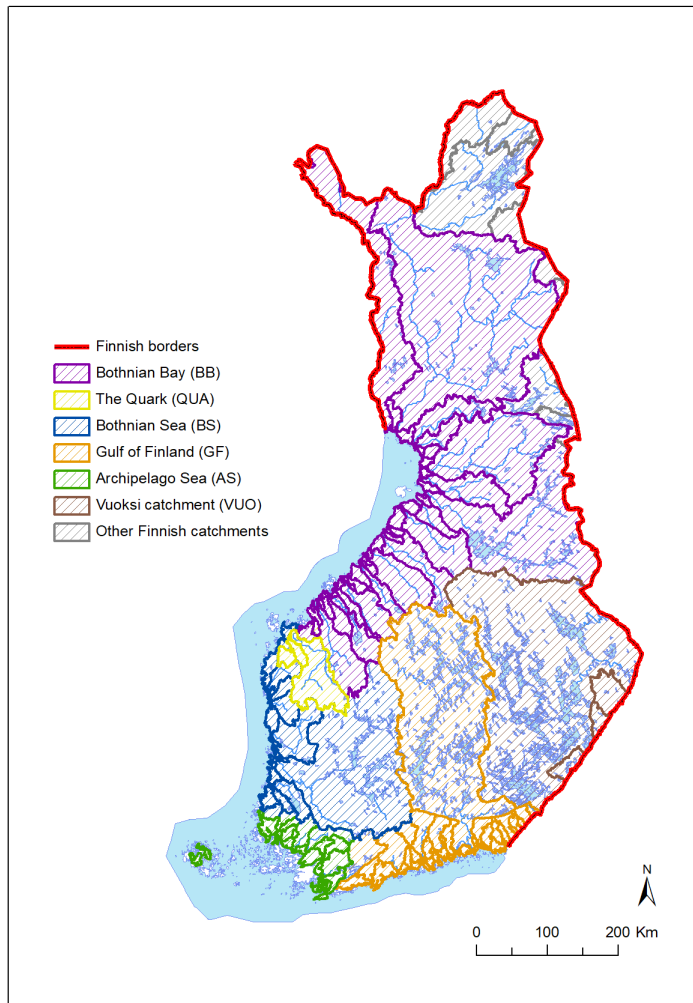
|                                |                               |     |      |      |       |      |      |      |      |     |     |     |     |
|--------------------------------|-------------------------------|-----|------|------|-------|------|------|------|------|-----|-----|-----|-----|
| 49_011<br>Perhonjoki<br>outlet | Ostro<br>bothn<br>ia          | 102 | 1.16 | 1.28 | -0.12 | 0.54 | 40.4 | 10.6 | 38.4 | 1.5 | 1.7 | 2.1 | 5.4 |
| 53_011<br>Kalajoki<br>outlet   | North<br>Ostro<br>bothn<br>ia | 93  | 1.57 | 1.55 | 0.02  | 0.68 | 52.2 | 9.1  | 28.9 | 1.7 | 2.0 | 1.1 | 5.0 |
| 57_012<br>Siikajoki<br>outlet  | North<br>Ostro<br>bothn<br>ia | 96  | 1.04 | 1.06 | -0.02 | 0.74 | 33.8 | 16.0 | 43.3 | 0.7 | 1.5 | 1.7 | 2.9 |

S3. Observed and simulated daily TN concentrations for selected test observation points.

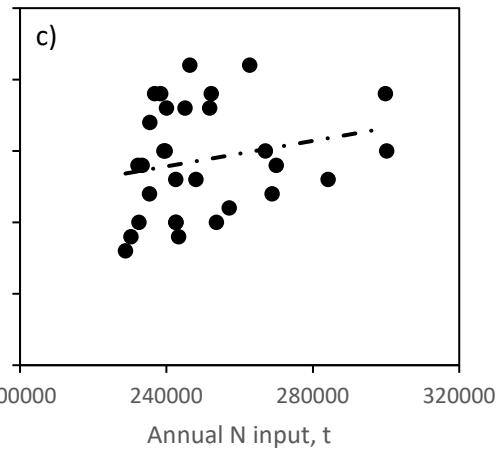
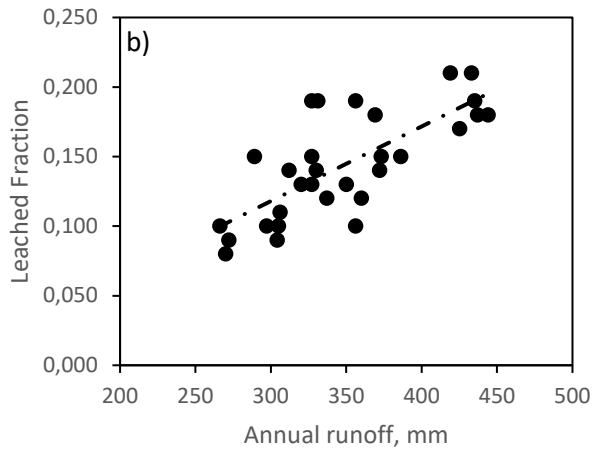
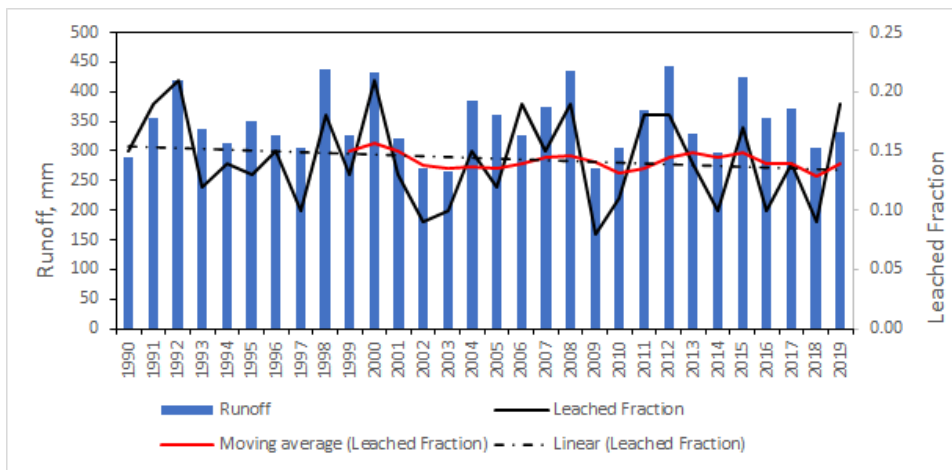




S4. Map of the Baltic Sea sub-basin catchments.



S5. a) Annual mean simulated runoff and leached N fraction for the whole Finland for years 1990-2019, and its relationship to b) annual mean runoff and c) annual mean N input.



S6. Agricultural TN loading ( $10^3 \text{ t a}^{-1}$ ) for the whole Finland for the period 1990-2019

| Year | Agricultural TN loading, $10^3 \text{ t a}^{-1}$ | Year | Agricultural TN loading, $10^3 \text{ t a}^{-1}$ | Year | Agricultural TN loading, $10^3 \text{ t a}^{-1}$ |
|------|--|------|--|------|--|
| 1990 | 43.8   | 2000 | 52.2   | 2010 | 27.6   |
| 1991 | 56.8   | 2001 | 33.0   | 2011 | 44.5   |
| 1992 | 55.7   | 2002 | 21.9   | 2012 | 44.4   |
| 1993 | 32.4   | 2003 | 24.2   | 2013 | 31.8   |
| 1994 | 37.3   | 2004 | 36.7   | 2014 | 24.7   |
| 1995 | 37.5   | 2005 | 27.5   | 2015 | 40.1   |
| 1996 | 40.1   | 2006 | 44.3   | 2016 | 23.2   |
| 1997 | 26.4   | 2007 | 35.0   | 2017 | 32.6   |
| 1998 | 44.8   | 2008 | 49.0   | 2018 | 21.6   |
| 1999 | 30.9   | 2009 | 17.7   | 2019 | 46.3   |
| Mean | 40.6   |      | 34.2   |      | 33.7   |