

## Supplementary Materials: Origin, Fate and Control of Pharmaceuticals in the Urban Water Cycle: A Case Study



**Figure S1.** Sampling location Meuse at Eijsden.



**Figure S2.** Sampling point Jeker at Maastricht.



**Figure S3.** Sampling point Geul at Meerssen.



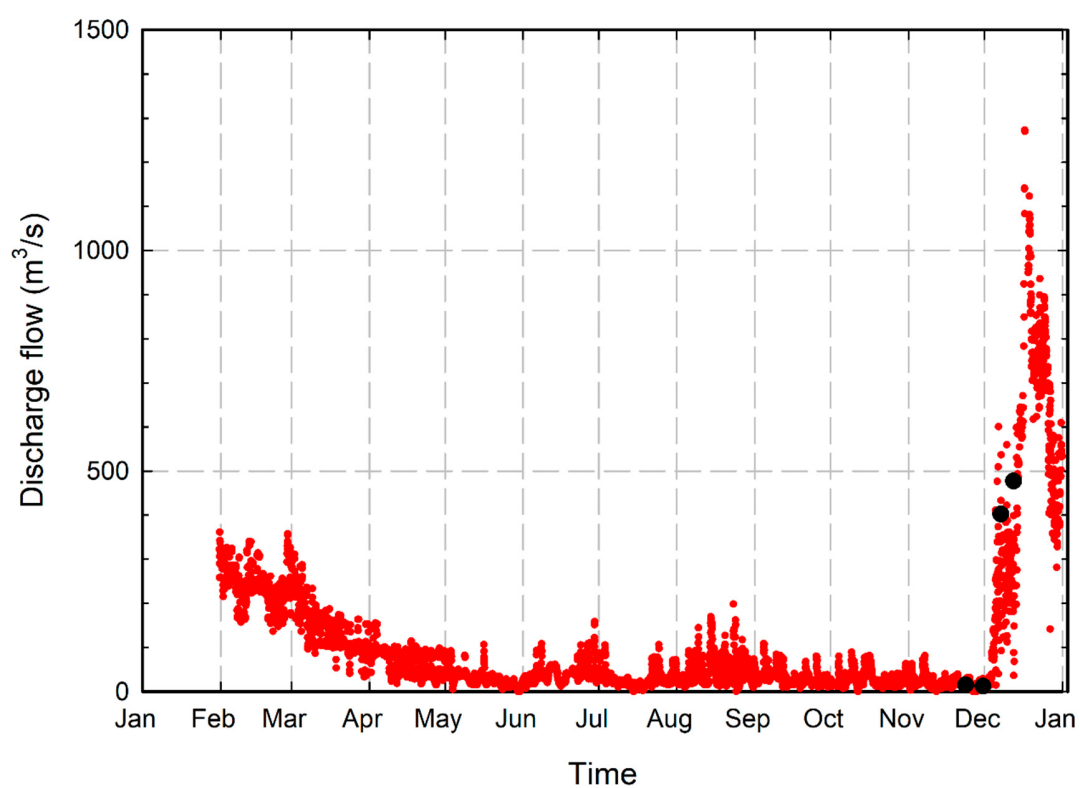
**Figure S4.** Sampling point Geleenbeek at Oud-Roosteren.



**Figure S5.** Sampling point Slijbeek.



**Figure S6.** Sampling point at intake of Water Treatment Plant Heel, Lateraalkanaal.



**Figure S7.** Discharge of the Meuse during 2011. The river showed an extreme low flow from May to early December. The black dots indicate the sampling dates for pharmaceuticals.

**Table S1.** Concentrations of pharmaceuticals in the Lateral Kanaal, measured at four different dates in 2011.

|                  | 23-11-2011 | 30-11-2011 | 07-12-2011 | 12-12-2011 |
|------------------|------------|------------|------------|------------|
| atenolol         | 0.02       | 0.02       | 0.04       | 0.03       |
| bezafibrate      | 0.01       | 0.01       | 0.01       | 0.01       |
| carbamazepine    | 0.07       | 0.07       | 0.09       | 0.05       |
| clindamycin      | 0.01       | 0.01       | 0.01       | 0.01       |
| diatrizoic acid  | 0.29       | 0.27       | 0.23       | 0.20       |
| diclofenac       | 0.02       | 0.02       | 0.04       | 0.03       |
| erythromycin A   | 0.03       | 0.03       | 0.03       | 0.03       |
| furosemide       | 0.01       | 0.01       | 0.01       | 0.02       |
| gemfibrozil      | 0.02       | 0.01       | 0.05       | 0.02       |
| guanyurea        | 4.40       | 3.40       | 4.40       | 1.50       |
| ketoprofen       | 0.01       | 0.01       | 0.01       | 0.01       |
| lincomycin       | 0.01       | 0.01       | 0.01       | 0.01       |
| metformin        | 1.50       | 1.40       | 2.00       | 2.20       |
| metoprolol       | 0.08       | 0.09       | 0.12       | 0.05       |
| metronidazole    | 0.01       | 0.01       | 0.01       | 0.01       |
| naproxen         | 0.01       | 0.01       | 0.03       | 0.03       |
| niacin           | 0.01       | 0.03       | 0.01       | 0.01       |
| paracetamol      | 0.01       | 0.01       | 0.05       | 0.01       |
| propranolol      | 0.01       | 0.01       | 0.01       | 0.01       |
| propyfenazonee   | 0.01       | 0.01       | 0.01       | 0.01       |
| sotalol          | 0.12       | 0.13       | 0.18       | 0.13       |
| sulfamethoxazole | 0.02       | 0.02       | 0.03       | 0.02       |
| tramadol         | 0.12       | 0.11       | 0.14       | 0.10       |
| trimethoprim     | 0.01       | 0.01       | 0.01       | 0.01       |
| venlafaxine      | 0.05       | 0.05       | 0.05       | 0.04       |
| <b>Sum</b>       | <b>6.9</b> | <b>5.8</b> | <b>7.6</b> | <b>4.6</b> |

**Table S2.** Concentrations of pharmaceuticals in the Slijbeek, measured at four different dates in 2011.

|                  | 23-11-2011  | 30-11-2011  | 07-12-2011  | 12-12-2011 |
|------------------|-------------|-------------|-------------|------------|
| atenolol         | 0.12        | 0.10        | 0.13        | 0.08       |
| bezafibrate      | 0.02        | 0.02        | 0.04        | 0.02       |
| carbamazepine    | 0.54        | 0.48        | 0.27        | 0.18       |
| clindamycin      | 0.02        | 0.01        | 0.01        | 0.01       |
| diatrizoic acid  | 0.16        | 0.15        | 0.09        | 0.12       |
| diclofenac       | 0.15        | 0.12        | 0.07        | 0.05       |
| erythromycin A   | 0.03        | 0.03        | 0.02        | 0.03       |
| furosemide       | 0.24        | 0.18        | 0.17        | 0.09       |
| gemfibrozil      | 0.66        | 0.53        | 0.48        | 0.27       |
| guanyurea        | 29.0        | 13.0        | 8.30        | 4.20       |
| ketoprofen       | 0.03        | 0.01        | 0.03        | 0.01       |
| lincomycin       | 0.01        | 0.02        | 0.01        | 0.01       |
| metformin        | 0.88        | 1.00        | 1.40        | 1.50       |
| metoprolol       | 0.79        | 0.66        | 0.46        | 0.30       |
| metronidazole    | 0.01        | 0.01        | 0.02        | 0.01       |
| naproxen         | 0.09        | 0.07        | 0.11        | 0.07       |
| niacin           | 0.01        | 0.01        | 0.07        | 0.01       |
| paracetamol      | 0.01        | 0.01        | 0.01        | 0.01       |
| propranolol      | 0.06        | 0.04        | 0.05        | 0.03       |
| propyfenazone    | 0.01        | 0.01        | 0.01        | 0.01       |
| sotalol          | 0.85        | 0.68        | 0.38        | 0.28       |
| sulfamethoxazole | 0.07        | 0.06        | 0.02        | 0.03       |
| tramadol         | 0.43        | 0.32        | 0.21        | 0.17       |
| trimethoprim     | 0.04        | 0.04        | 0.03        | 0.02       |
| venlafaxine      | 0.14        | 0.10        | 0.07        | 0.06       |
| <b>Sum</b>       | <b>34.4</b> | <b>17.7</b> | <b>12.5</b> | <b>7.6</b> |

**Table S3.** Concentrations of pharmaceuticals in the Geleenbeek, measured at four different dates in 2011.

|                  | 23-11-2011  | 30-11-2011  | 07-12-2011  | 12-12-2011  |
|------------------|-------------|-------------|-------------|-------------|
| atenolol         | 0.32        | 0.24        | 0.45        | 0.24        |
| bezafibrate      | 0.01        | 0.02        | 0.05        | 0.03        |
| carbamazepine    | 0.42        | 0.32        | 0.30        | 0.16        |
| clindamycin      | 0.05        | 0.03        | 0.05        | 0.03        |
| diatrizoic acid  | 0.12        | 0.11        | 0.20        | 0.13        |
| diclofenac       | 0.34        | 0.23        | 0.26        | 0.16        |
| erythromycin A   | 0.02        | 0.04        | 0.04        | 0.03        |
| furosemide       | 0.78        | 0.47        | 0.84        | 0.56        |
| gemfibrozil      | 0.11        | 0.10        | 0.23        | 0.15        |
| guanyurea        | 29.0        | 14.0        | 15.0        | 6.5         |
| ketoprofen       | 0.02        | 0.01        | 0.02        | 0.01        |
| lincomycin       | 0.01        | 0.01        | 0.01        | 0.01        |
| metformin        | 0.78        | 0.90        | 6.40        | 5.40        |
| metoprolol       | 1.18        | 0.87        | 1.01        | 0.64        |
| metronidazole    | 0.01        | 0.01        | 0.01        | 0.01        |
| naproxen         | 0.04        | 0.04        | 0.18        | 0.16        |
| niacin           | 0.01        | 0.08        | 0.21        | 0.11        |
| paracetamol      | 0.01        | 0.01        | 0.01        | 0.12        |
| propranolol      | 0.07        | 0.05        | 0.05        | 0.04        |
| propyfenazone    | 0.01        | 0.01        | 0.01        | 0.01        |
| sotalol          | 1.15        | 0.82        | 0.93        | 0.58        |
| sulfamethoxazole | 0.08        | 0.06        | 0.08        | 0.05        |
| tramadol         | 0.62        | 0.46        | 0.42        | 0.28        |
| trimethoprim     | 0.04        | 0.04        | 0.06        | 0.04        |
| venlafaxine      | 0.23        | 0.18        | 0.17        | 0.10        |
| <b>Sum</b>       | <b>35.4</b> | <b>19.1</b> | <b>27.0</b> | <b>15.6</b> |

**Table S4.** Concentrations of pharmaceuticals in the Geul, measured at four different dates in 2011.

|                  | 23-11-2011 | 30-11-2011 | 07-12-2011 | 12-12-2011 |
|------------------|------------|------------|------------|------------|
| atenolol         | 0.03       | 0.03       | 0.02       | 0.03       |
| bezafibrate      | 0.01       | 0.01       | 0.01       | 0.01       |
| carbamazepine    | 0.05       | 0.07       | 0.02       | 0.01       |
| clindamycin      | 0.01       | 0.01       | 0.01       | 0.01       |
| diatrizoic acid  | 0.12       | 0.03       | 0.01       | 0.02       |
| diclofenac       | 0.11       | 0.12       | 0.07       | 0.07       |
| erythromycin A   | 0.03       | 0.03       | 0.03       | 0.03       |
| furosemide       | 0.07       | 0.06       | 0.06       | 0.07       |
| gemfibrozil      | 0.02       | 0.01       | 0.01       | 0.01       |
| guanyurea        | 3.80       | 3.00       | 1.00       | 1.20       |
| ketoprofen       | 0.01       | 0.01       | 0.01       | 0.01       |
| lincomycin       | 0.01       | 0.01       | 0.01       | 0.03       |
| metformin        | 1.10       | 1.10       | 1.40       | 1.30       |
| metoprolol       | 0.21       | 0.20       | 0.11       | 0.14       |
| metronidazole    | 0.01       | 0.01       | 0.01       | 0.01       |
| naproxen         | 0.03       | 0.03       | 0.03       | 0.03       |
| niacin           | 0.04       | 0.03       | 0.15       | 0.04       |
| paracetamol      | 0.01       | 0.02       | 0.16       | 0.16       |
| propranolol      | 0.01       | 0.01       | 0.01       | 0.01       |
| propyfenazone    | 0.01       | 0.01       | 0.01       | 0.01       |
| sotalol          | 0.29       | 0.35       | 0.14       | 0.17       |
| sulfamethoxazole | 0.02       | 0.02       | 0.01       | 0.01       |
| tramadol         | 0.15       | 0.17       | 0.09       | 0.09       |
| trimethoprim     | 0.01       | 0.01       | 0.01       | 0.01       |
| venlafaxine      | 0.05       | 0.06       | 0.04       | 0.04       |
| <b>Sum</b>       | <b>6.2</b> | <b>5.4</b> | <b>3.4</b> | <b>3.5</b> |

**Table S5.** Concentrations of pharmaceuticals in the Jeker, measured at four different dates in 2011.

|                  | 23-11-2011  | 30-11-2011  | 07-12-2011  | 12-12-2011  |
|------------------|-------------|-------------|-------------|-------------|
| atenolol         | 0.07        | 0.07        | 0.09        | 0.10        |
| bezafibrate      | 0.01        | 0.01        | 0.01        | 0.01        |
| carbamazepine    | 0.16        | 0.16        | 0.09        | 0.10        |
| clindamycin      | 0.03        | 0.03        | 0.03        | 0.03        |
| diatrizoic acid  | 0.34        | 0.34        | 0.22        | 0.44        |
| diclofenac       | 0.15        | 0.15        | 0.10        | 0.12        |
| erythromycin A   | 0.02        | 0.02        | 0.02        | 0.02        |
| furosemide       | 0.12        | 0.12        | 0.07        | 0.12        |
| gemfibrozil      | 0.01        | 0.01        | 0.01        | 0.01        |
| guanyurea        | 4.70        | 4.70        | 1.80        | 1.90        |
| ketoprofen       | 0.05        | 0.05        | 0.03        | 0.05        |
| lincomycin       | 0.05        | 0.05        | 0.02        | 0.02        |
| metformin        | 5.50        | 5.50        | 5.80        | 5.90        |
| metoprolol       | 0.06        | 0.06        | 0.01        | 0.05        |
| metronidazole    | 0.01        | 0.01        | 0.01        | 0.01        |
| naproxen         | 0.14        | 0.14        | 0.17        | 0.16        |
| niacin           | 0.13        | 0.13        | 0.24        | 0.18        |
| paracetamol      | 0.24        | 0.24        | 0.63        | 0.94        |
| propranolol      | 0.06        | 0.06        | 0.05        | 0.05        |
| propyfenazone    | 0.01        | 0.01        | 0.01        | 0.01        |
| sotalol          | 0.53        | 0.53        | 0.37        | 0.47        |
| sulfamethoxazole | 0.04        | 0.04        | 0.02        | 0.04        |
| tramadol         | 0.67        | 0.67        | 0.40        | 0.48        |
| trimethoprim     | 0.01        | 0.01        | 0.01        | 0.01        |
| venlafaxine      | 0.25        | 0.25        | 0.18        | 0.20        |
| <b>Sum</b>       | <b>13.4</b> | <b>13.4</b> | <b>10.4</b> | <b>11.4</b> |

**Table S6.** Concentrations of pharmaceuticals in the Meuse at Eijsden, measured at four different dates in 2011.

|                  | 23-11-2011 | 30-11-2011 | 07-12-2011 | 12-12-2011 |
|------------------|------------|------------|------------|------------|
| atenolol         | 0.04       | 0.05       | 0.04       | 0.02       |
| bezafibrate      | 0.01       | 0.01       | 0.01       | 0.01       |
| carbamazepine    | 0.06       | 0.07       | 0.06       | 0.01       |
| clindamycin      | 0.01       | 0.01       | 0.01       | 0.01       |
| diatrizoic acid  | 0.37       | 0.33       | 0.19       | 0.08       |
| diclofenac       | 0.04       | 0.05       | 0.04       | 0.03       |
| erythromycin A   | 0.03       | 0.03       | 0.03       | 0.03       |
| furosemide       | 0.01       | 0.01       | 0.03       | 0.01       |
| gemfibrozil      | 0.01       | 0.01       | 0.01       | 0.01       |
| guanyurea        | 2.30       | 2.00       | 0.89       | 0.26       |
| ketoprofen       | 0.01       | 0.01       | 0.01       | 0.01       |
| lincomycin       | 0.01       | 0.01       | 0.01       | 0.01       |
| metformin        | 3.90       | 4.20       | 2.90       | 1.90       |
| metoprolol       | 0.02       | 0.01       | 0.01       | 0.01       |
| metronidazole    | 0.01       | 0.01       | 0.01       | 0.01       |
| naproxen         | 0.04       | 0.05       | 0.05       | 0.03       |
| niacin           | 0.03       | 0.03       | 0.05       | 0.06       |
| paracetamol      | 0.04       | 0.14       | 0.01       | 0.18       |
| propranolol      | 0.01       | 0.02       | 0.02       | 0.01       |
| propyfenazone    | 0.01       | 0.01       | 0.01       | 0.01       |
| sotalol          | 0.16       | 0.17       | 0.17       | 0.07       |
| sulfamethoxazole | 0.01       | 0.02       | 0.02       | 0.01       |
| tramadol         | 0.20       | 0.22       | 0.13       | 0.05       |
| trimethoprim     | 0.01       | 0.01       | 0.01       | 0.01       |
| venlafaxine      | 0.09       | 0.09       | 0.05       | 0.02       |
| <b>Sum</b>       | <b>7.4</b> | <b>7.6</b> | <b>4.8</b> | <b>2.9</b> |

**Table S7.** Concentrations of metabolites in the Lateraal Kanaal, measured at four different dates in 2011.

|                                | 23-11-2011 | 30-11-2011 | 07-12-2011 | 12-12-2011 |
|--------------------------------|------------|------------|------------|------------|
| 10.11-trans-diol-carbamazepine | 0.22       | 0.22       | 0.26       | 0.16       |
| 2-hydroxy carbamazepine        | 0.01       | 0.01       | 0.01       | 0.01       |
| 3-hydroxy carbamazepine        | 0.01       | 0.01       | 0.01       | 0.01       |
| 4-acetaminophen sulfaat        | 0.03       | 0.03       | 0.03       | 0.03       |
| 4-formylaminoantipyrine        | 0.02       | 0.02       | 0.02       | 0.02       |
| alfa-hydroxy metoprolol        | 0.01       | 0.01       | 0.01       | 0.01       |
| anhydro erythromycine A        | 0.05       | 0.05       | 0.05       | 0.05       |
| carbamazepine-10.11-epoxide    | 0.01       | 0.01       | 0.01       | 0.01       |
| hydroxy ibuprofen              | 0.50       | 0.50       | 0.50       | 0.50       |
| N4-acetyl sulfamethoxazole     | 0.01       | 0.01       | 0.01       | 0.01       |
| o-desmethyldramadol            | 0.01       | 0.01       | 0.09       | 0.08       |
| oxcarbamazepine                | 0.01       | 0.01       | 0.01       | 0.01       |
| <b>Sum</b>                     | <b>0.9</b> | <b>0.9</b> | <b>1.0</b> | <b>0.9</b> |

**Table S8.** Concentrations of metabolites in the Slijbeek, measured at four different dates in 2011.

|                                | 23-11-2011 | 30-11-2011 | 07-12-2011 | 12-12-2011 |
|--------------------------------|------------|------------|------------|------------|
| 10.11-trans-diol-carbamazepine | 1.79       | 1.42       | 0.73       | 0.48       |
| 2-hydroxy carbamazepine        | 0.10       | 0.08       | 0.05       | 0.03       |
| 3-hydroxy carbamazepine        | 0.13       | 0.11       | 0.06       | 0.04       |
| 4-acetaminophen sulfaat        | 0.03       | 0.03       | 0.03       | 0.03       |
| 4-formylaminoantipyrine        | 0.02       | 0.02       | 0.01       | 0.01       |
| alfa-hydroxy metoprolol        | 0.15       | 0.11       | 0.10       | 0.06       |
| anhydro erythromycine A        | 0.11       | 0.06       | 0.10       | 0.03       |
| carbamazepine-10.11-epoxide    | 0.07       | 0.06       | 0.03       | 0.02       |
| hydroxy ibuprofen              | 0.50       | 0.50       | 0.50       | 0.50       |
| N4-acetyl sulfamethoxazole     | 0.02       | 0.02       | 0.02       | 0.02       |
| o-desmethyldramadol            | 0.20       | 0.15       | 0.01       | 0.08       |
| oxcarbamazepine                | 0.01       | 0.01       | 0.01       | 0.01       |
| <b>Sum</b>                     | <b>3.1</b> | <b>2.6</b> | <b>1.7</b> | <b>1.3</b> |

**Table S9.** Concentrations of metabolites in the Geleenbeek, measured at four different dates in 2011.

|                                | 23-11-2011 | 30-11-2011 | 07-12-2011 | 12-12-2011 |
|--------------------------------|------------|------------|------------|------------|
| 10.11-trans-diol-carbamazepine | 1.28       | 0.96       | 0.98       | 0.55       |
| 2-hydroxy carbamazepine        | 0.08       | 0.06       | 0.06       | 0.03       |
| 3-hydroxy carbamazepine        | 0.10       | 0.08       | 0.08       | 0.04       |
| 4-acetaminophen sulfaat        | 0.03       | 0.03       | 0.01       | 0.02       |
| 4-formylaminoantipyrine        | 0.25       | 0.15       | 0.19       | 0.11       |
| alfa-hydroxy metoprolol        | 0.04       | 0.04       | 0.10       | 0.09       |
| anhydro erythromycine A        | 0.06       | 0.05       | 0.05       | 0.04       |
| carbamazepine-10.11-epoxide    | 0.05       | 0.04       | 0.04       | 0.02       |
| hydroxy ibuprofen              | 0.50       | 0.50       | 0.83       | 0.87       |
| N4-acetyl sulfamethoxazole     | 0.01       | 0.01       | 0.01       | 0.01       |
| o-desmethylnaloxone            | 0.33       | 0.24       | 0.22       | 0.14       |
| oxcarbamazepine                | 0.15       | 0.07       | 0.12       | 0.05       |
| <b>Sum</b>                     | <b>2.9</b> | <b>2.2</b> | <b>2.7</b> | <b>2.0</b> |

**Table S10.** Concentrations of metabolites in the Geul, measured at four different dates in 2011.

|                                | 23-11-2011 | 30-11-2011 | 07-12-2011 | 12-12-2011 |
|--------------------------------|------------|------------|------------|------------|
| 10.11-trans-diol-carbamazepine | 0.20       | 0.22       | 0.09       | 0.11       |
| 2-hydroxy carbamazepine        | 0.01       | 0.01       | 0.01       | 0.01       |
| 3-hydroxy carbamazepine        | 0.01       | 0.01       | 0.01       | 0.01       |
| 4-acetaminophen sulfaat        | 0.03       | 0.02       | 0.15       | 0.18       |
| 4-formylaminoantipyrine        | 0.06       | 0.08       | 0.04       | 0.04       |
| alfa-hydroxy metoprolol        | 0.01       | 0.01       | 0.01       | 0.01       |
| anhydro erythromycine A        | 0.05       | 0.05       | 0.05       | 0.05       |
| carbamazepine-10.11-epoxide    | 0.01       | 0.01       | 0.01       | 0.01       |
| hydroxy ibuprofen              | 0.50       | 0.50       | 0.50       | 0.50       |
| N4-acetyl sulfamethoxazole     | 0.01       | 0.01       | 0.01       | 0.01       |
| o-desmethylnaloxone            | 0.01       | 0.08       | 0.01       | 0.01       |
| oxcarbamazepine                | 0.01       | 0.01       | 0.01       | 0.01       |
| <b>Sum</b>                     | <b>0.9</b> | <b>1.0</b> | <b>0.9</b> | <b>1.0</b> |

**Table S11.** Concentrations of metabolites in the Jeker, measured at four different dates in 2011.

|                                | 23-11-2011 | 30-11-2011 | 07-12-2011 | 12-12-2011 |
|--------------------------------|------------|------------|------------|------------|
| 10.11-trans-diol-carbamazepine | 0.42       | 0.39       | 0.26       | 0.31       |
| 2-hydroxy carbamazepine        | 0.02       | 0.02       | 0.02       | 0.02       |
| 3-hydroxy carbamazepine        | 0.03       | 0.03       | 0.02       | 0.02       |
| 4-acetaminophen sulfaat        | 0.40       | 0.43       | 0.69       | 1.12       |
| 4-formylaminoantipyrine        | 0.08       | 0.08       | 0.04       | 0.07       |
| alfa-hydroxy metoprolol        | 0.01       | 0.01       | 0.01       | 0.01       |
| anhydro erythromycine A        | 0.05       | 0.05       | 0.05       | 0.03       |
| carbamazepine-10.11-epoxide    | 0.02       | 0.02       | 0.01       | 0.01       |
| hydroxy ibuprofen              | 1.36       | 1.43       | 1.27       | 1.44       |
| N4-acetyl sulfamethoxazole     | 0.03       | 0.03       | 0.02       | 0.03       |
| o-desmethyldramadol            | 0.32       | 0.32       | 0.18       | 0.21       |
| oxcarbamazepine                | 0.01       | 0.01       | 0.01       | 0.01       |
| <b>Sum</b>                     | <b>2.8</b> | <b>2.8</b> | <b>2.6</b> | <b>3.3</b> |

**Table S12.** Concentrations of metabolites in the Meuse at Eijsden, measured at four different dates in 2011.

|                                | 23-11-2011 | 30-11-2011 | 07-12-2011 | 12-12-2011 |
|--------------------------------|------------|------------|------------|------------|
| 10.11-trans-diol-carbamazepine | 0.18       | 0.19       | 0.17       | 0.07       |
| 2-hydroxy carbamazepine        | 0.01       | 0.01       | 0.01       | 0.01       |
| 3-hydroxy carbamazepine        | 0.01       | 0.01       | 0.01       | 0.01       |
| 4-acetaminophen sulfaat        | 0.07       | 0.13       | 0.03       | 0.47       |
| 4-formylaminoantipyrine        | 0.03       | 0.03       | 0.02       | 0.01       |
| alfa-hydroxy metoprolol        | 0.01       | 0.01       | 0.01       | 0.01       |
| anhydro erythromycine A        | 0.05       | 0.05       | 0.05       | 0.05       |
| carbamazepine-10.11-epoxide    | 0.01       | 0.01       | 0.01       | 0.01       |
| hydroxy ibuprofen              | 0.50       | 0.50       | 0.50       | 0.50       |
| N4-acetyl sulfamethoxazole     | 0.02       | 0.02       | 0.01       | 0.01       |
| o-desmethyldramadol            | 0.13       | 0.14       | 0.11       | 0.01       |
| oxcarbamazepine                | 0.01       | 0.01       | 0.01       | 0.01       |
| <b>Sum</b>                     | <b>1.0</b> | <b>1.1</b> | <b>0.9</b> | <b>1.2</b> |