TRENDS IN URBAN STORM WATER QUALITY IN TALLINN AND INFLUENCES FROM STORMFLOW AND BASEFLOW

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Abstract. Temporal trends provide a good interpretation of change in stormwater quality over time. This study aimed to analyse trends and influences due to stormflow and baseflow. Grab samples of 18–19 years from 1995 to 2014 recorded at outlets of 7 Tallinn watersheds were analysed for monotonic trend through seasonal Mann Kendall test for long-term, short-term, baseflow and stormflow. Statistically significant downward trends (P-value (p) < 0.05) were found for 6 – hydrocarbon (HC), 1 – suspended solids (SS), 3 – biological oxygen demand (BOD), 4 – total nitrogen (TN) and 2 – total phosphorus (TP) out of 7 sampling outlets over the last 10 years. Less significant decreasing trends (p > 0.05 and < 0.2) for 3 – SS, 1 – BOD, 1 – TN and 1 – TP were identified. Statistically significant long-term upward trends of pH were revealed in 5 basins, which reduced to 2 with 5 less significant upward trends over the 10 year period, indicating improvements in pH reduction. Härjapea has the highest pH without trend but it includes an upward trend of TN at p = 0.051. The highly possible causes for downward trends are street sweeping, sewer network improvement, decline in sub-urban agricultural areas, etc. The upward trend results of pH are related to increased alkalinisation due to acidic rain, weathering of carbonate rocks, sewage discharge and alkaline road dust. In most of the basins, stormflow has more influence on trends than baseflow.

Keywords: seasonal Mann Kendall test, stormwater quality, temporal trend.

Introduction

Stormwater runoff volume and pollutant load on Baltic Sea is a topic of concern for all Baltic Sea member states. Indeed, HELCOM (Baltic Marine Environment Protection Commission – Helsinki Commission) has made recommendations for restoring the ecological status of the Baltic marine environment. Valid recommendations on the reduction of discharges from urban areas through the proper management of stormwater systems focuses on the volume of runoff and first flush in the separate system and most polluted overflow in the combined system (HELCOM, 2002). One of the major problems in the Baltic Sea region, including Estonia, is eutrophication in water bodies (Itäl et al., 2010), and urban runoff has contributed substantially in raising nutrient levels (King et al., 2007). Significant efforts have been made in the past few decades in reducing the eutrophication of the Baltic Sea. Most recently, the new HELCOM Baltic Sea Action Plan (BSAP) has been adopted by all Baltic countries and by the European Union to reduce the anthropogenic nutrient load and restore the good ecological status of the Baltic Sea by 2021 (HELCOM, 2007). The objectives of the EU Water Framework Directive, HELCOM recommendation and BSAP are reflected in the Estonian Water Act (RTI, 2015b), and it has set a target of protecting all waters against pollution and achieving the good status of all waters by promoting sustainable water and wastewater management. Trends in time series can be one effective interpretation of the increase and decrease in the pollution load discharged from stormwater systems, and this can assist in the decision-making process for further planning with potential drivers for changes in loads.

The regulation of Estonia and HELCOM has set stormwater discharges and pollutant thresholds. It requires compliance verification. According to the Estonian Environmental Monitoring Act, national, local government and special permit owners are responsible for stormwater monitoring (RTI, 2015a). The collection and treatment of wastewater and stormwater are regulated by the Estonian Public Water Supply and Sewerage Act, according to which local government shall develop a plan and activities for stormwater management. Outside the buildings, stormwater and sewerage system are constructed, rehabilitated, maintained and operated according to the Estonian standard EVS 848:2013. The principle is based on returning stormwater to nature either by possible infiltration and delay at sources or reuse. Tallinn has issues with the drainage system in terms of frequent flooding during heavy rainfalls and snowmelts (RTI, 2013b). Instead, the pollution loads from stormwater outlets have a substantial part to play in coastal water degradation, especially in Tallinn Bay (Erm et al., 2014) and Kopli Bay (TCG, 2015). The study conducted in 2009 for the ecological status of coastal water in Tallinn city showed moderate ecological status in two of the three studied water bodies of Tallinn Bay (Miiduranna, Pirita) and one in Paljassaare Bay; however, it was bad in one water body of Tallinn Bay (Kalaranna-Russalka) and one of Kopli Bay (Stroomi) (TCG, 2015). Moreover, long-term negative dynamics were observed in these two water bodies. However, long-term improvement trends were found in the Paljassaare and Miiduranna sea areas. The inflow of stormwater is argued as being one potential
source for impact on water transparency and depletion of the oxygen level. Thus, the trends of stormwater pollutants are essential for effective understanding of the temporal changed share of pollutant discharges in the ecological status of coastal water. Some of the initiatives introduced in the 2000s by the City of Tallinn have begun to reduce runoff and the pollution load. Many action plans and activities are formulated in the Tallinn Development Plan 2014–2020 (RTI, 2013b), Tallinn Stormwater Strategy to 2030 (RTI, 2012) and Tallinn Water Supply and Sewerage Development Plan 2010–2021, such as reducing the pollution load by street cleaning, minimising hydrocarbon through the installation of oil filters, reducing nutrients building treatment plants, the construction of separate system and the reconstruction of the combined sewer system, etc. The compliance of these initiatives with the reduction of pollutants needs to be ensured over the long-term and short-term periods to make decisions on further planning.

The water moving through the storm drainage system between the rain events, which is called baseflow, has a substantial effect on overall stormwater runoff and quality output (Nicolau et al., 2012; Janke et al., 2014). Nevertheless, extreme rainfall events have higher impacts on the discharge of pollutants in stormwater runoff (King et al., 2007; Erm et al., 2014). Research has shown that while phosphorus is predominantly delivered by stormflow, nitrogen loading is similar between the baseflow and stormflow (Janke et al., 2014). The temporal changes in pollution require information about the contribution of the illegal connections and / or ground water source and rainfall events. The influences of these flows on overall trends are not yet well defined. Therefore, there is room to investigate how the quality output of baseflow and stormflow influence trends over the years.

The main objective of this study is to discern upward / downward trends of the pollution load in 7 watersheds in Tallinn and to investigate the possible causes behind those trends.

**Material and Methods**

**Study Area**

Tallinn, the capital of Estonia, is situated in the northwestern part of Estonia with an area of 156 sq. km. It has a temperate climate where the average air temperature between 1961 and 2010 ranges from -5.9 to -1.0 in January and 12.7 to 21.9 in July (EWS, 2015). The average rainfall is 550–750 mm and the mean runoff is 280–290 mm per year. Most of the land in Tallinn is urbanised, with impervious areas forming about 50% of the total area. There are 66 stormwater outlets (EELIS, 2015). Among them, 47 outlets discharge to the coastal sea. Major outlets that discharge to Tallinn Bay are Härjapea, Saare tee / Pirdita, Lasnamäe, Russalka, Ülemiste Polder and to Kopli Bay are Rocca al Mare and Mustoja as shown in Fig. 1. The area of the stormwater system of Tallinn is about 6,500 hectares and the length of stormwater pipeline was 478 km in 2014 (Tallinna Vesi, 2014). Altogether, these 7 outlets approximately cover total area of 4,000 ha.

**Data sources and Sampling Strategy**

Stormwater monitoring has been carried out since the late 1980s, but it only became regular in the 1990s. For this study, data from three sources have been combined. The first source of data is monitoring reports from Paulkin et al. (2005–2011) for the years 2005 and 2008–2011. In this monitoring system, grab samples were collected 4–6 times a year from the stormwater outlets by the Estonian Environmental Research Centre. The data was measured only once in 2010. The second source is monitored data provided by AS Tallinna Vesi, a special water permit owner that has measured samples each month from 1996 to 2014 in 6 outlets: Härjapea, Lasnamäe / Lauluväljak, Pirdita / Saare tee, Mustoja, Rocca al Mare and Russalka. Besides these, one other outlet is Kadriorg, which has only two years of data and is excluded for analysis. Six parameters: pH, SS, TN, TP, BOD₅ and HC are measured with grab sampling. The third source is samples measured by both the department of Environmental Engineering of Tallinn University of Technology and AS Tallinna Vesi for the period 2012–2014 in six outlets excluding Härjapea and Kadriorg, but Ülemiste polder is included.
Samples were tested for parameters such as dissolved oxygen, conductivity, pH, temperature, SS, TN, TP, BOD₅, HC, salmonella, Escherichia Coli and Enterococci using analytical methods based on ISO 10523, ISO 5667-10, EVS-EN 25814, EVS-EN 27888, EVS-EN 872, ISO 5815-1, SFS 5505, EVS 9377-2, EVS 9308-1, EVS-EN ISO 7899-2, EVS-EN ISO 6878 and EVS-ISO 6340. All of these are competent bodies according to EN ISO/IEC 17025:2005 for conducting tests in the field of water analysis (accreditation scope on the Estonian Accreditation Centre). The sampling procedure adhered to Estonian Environment Minister Regulation “Sampling methodology” as stated in the Water Act. EVS-EN 25667-2, EVS-EN ISO 5667-3:2012 or any equivalent internationally recognised standard should be followed. These grab samples of 18–19 years from 1995 to 2014 at outlets of 7 watersheds in Tallinn are used for this study. Dissolved oxygen, conductivity, temperature and microbiological parameters have data either less than 10 years or inconsistent. Therefore, these parameters are excluded from data analysis. Since 2012, the records have been available for discharge and sampling times that assist in separating data into baseflow and stormflow. The hourly consistent rainfall data from the Tallinn-Harku Meteorological Station, which is located approximately 20 km from the study area, was obtained from the Estonian Meteorological and Hydrological Institute (EMHI) for 10 years from 2005 to 2014.

Data have been split into baseflow and stormflow by looking at the antecedent rainfall of 3 dry days prior to the sampled time. Most of the dry weather flow samples are taken after a period of at least 3 days without rain (Smoley, 1993; Schiff, 1997; Francey et al., 2010) when the runoff does not exceed the minimum sampling threshold. If there is no rain within 3 days or rainfall < 2 mm/hr (also compared with available discharge data) then these data are categorised as baseflow; otherwise, they are stormflow data.

Statistical Methods

Monotonic trends in time series on watershed basis are analysed for six parameters: HC, pH, SS, BOD₅, TN and TP using seasonal Mann Kendall (SMK) trend test (Hirsch, Slack, 1984). Water quality data are usually not normally distributed and exhibit a seasonal pattern (Gilliom, Helsel, 1986). SMK tests are non-parametric tests for the detection of trends in time series. It provides a way for the accounting of ties and missing values and is defined as a sum of all signs of differences in the dataset. The test statistic is called SMK statistics (SMK-stat), which has approximately standard normal distribution according to the central limit theorem. Variance is corrected for ties and serial correlation among seasons during the test (Hirsch, Slack, 1984). The trends are analysed in four kinds of data sets: long-term data set for 18-19 years from 1995 to 2014, short-term data set for 10 years from 2005 to 2014, baseflow and stormflow for the same 10 year period. SMK statistics, 5% and 20% significance levels ($p \leq 0.05$ and $p > 0.05$ to $< 0.20$) are estimated, the differences between trends are compared and the possible causes are investigated. Significance ranks are provided as 3 for $p \leq 0.05$ indicating statistically significant, 2 for $p > 0.05$ and $< 0.20$ indicating trends can exist if $p < 0.20$ and no trend for $p > 0.20$ after performing a two-tailed test. The positive and negative sign indicate upward trend and downward trend respectively. There are three factors that drive the performance of the short-term trend test. Firstly, some data do not have 19 years of data but only 10 years, as in the Ülemiste polder. Secondly, consistent hourly rainfall data is only available for this period in order to categorise stormflow and baseflow. Thirdly, the trends observed in the long term may not necessarily appear in the short term or vice versa. Therefore, analysing both long term and short-term data may provide information on these characteristics.

Results and Discussion

The results from SMK test for long-term, short-term, stormflow and base flow are presented in Table 1. This includes SMK statistics, $P$-values and significance ranks. The concentration range of the data from 1995 to 2014 at 7 outlets are presented in Fig. 2 with statistical 10th and 90th percentile, mean, median, maximum and minimum points. The results from Table 1 is summarised in simple form and presented in Table 2 to provide the information of short-term, stormflow and baseflow trends with significance ranks, which make it easier to find the number of statistically significant trends according to basins and parameters. Generally, pollutants concentration and trends show decreasing tendency, thus indicating improved water quality. The results by each parameter are described below.

Hydrocarbons

HC has remarkable decreasing trends in Tallinn because there are statistically significant downward trends in almost all basins, 5 in 7 basins over 19 years and 6 in 7 basins over the 10 year period (Table 1 and Table 2). However, the HC downward trend in Härjapea basin for the long period is less significant ($p = 0.076$). These downward trends mainly occurred during stormflow, as in Table 1 where 5 out of the 7 basins have decreasing trends at $p < 0.20$, but baseflow has also attributed in some of the basins, e.g. Härjapea, Rocca al Mare, and Russalka. The decreasing tendencies are probably related to either the reduced oil emission from vehicles or trapping oil substances through the installation of oil trappers on the roadsides before entering drainage channels. HC has 90th percentile varied from 2–2.6 mg/l based on catchment basins during the 1995–2014 period (as in Fig. 2). They are less than the stormwater limit value of 5 mg/l, i.e. national permissible concentration of Estonia. However, the annual 90th percentile between 2002 and 2004 exceeded the limit values in the Piritu and Rocca al Mare outlets.

Hydrocarbons can cause problems in receiving water bodies as they can be toxic to aquatic life, depress the oxygen concentration and impart a foul odour. PAH, a compound of HC, even in low concentration can affect
aquatic life (Khan et al., 2006). The amount of HC in stormwater runoff highly depends on the land use and volume of runoff. In many studies, they are found in higher quantity in highways, car parks, roads and vehicle washing areas than in residential areas (Stenstrom et al., 1984, Göbel et al., 2007). Dry atmospheric deposition due to the accumulation of dust, aerosol and gas on the roofs and land surfaces form residue and are washed into drainage / waterways as concentrated pollutants during runoff events (Göbel et al., 2007). Therefore, street cleaning is highly effective in reducing the HC loads in storm runoff. Indeed, few parking areas and highways in land use can significantly decrease HC concentration. The City of Tallinn has initiated an action plan for implementing street cleaning methods in most of the areas and these practices have been increased in the last few years. Footpaths, roads and car parks are being kept clean and it is made necessary to clean the streets before the spring starts (RTI, 2013b; Tallinn Vesi, 2014). To minimise HCs, they have installed sand and oil filters on stormwater drainage on major roads and car parks (RTI, 2013b). Moreover, local oil separators have been installed around car parks and petrol stations.

Table 1. SMK Test results

| Storm outlets       | HC, mg/l | SS, mg/l | pH     | BOD5, mgO2/l | TN, mgN/l | TP, mgP/l |
|---------------------|----------|----------|--------|--------------|-----------|-----------|
|                      | SMK      | P-value  | Sig.   | SMK          | P-value   | Sig.      |
| 18–19 years data (1995–2014) |          |          |        |              |           |           |
| Härjapea            | -1.8 0.076 | -2       |        |              |           |           |
| Lasnamäe            | -2.3 0.022 | -3       |        |              |           |           |
| Mustoja             | -3.5 0.001 | -3       |        |              |           |           |
| Pirita              | -2.7 0.007 | -3       |        |              |           |           |
| Rocca-al-Mare       | -2.7 0.006 | -3       |        |              |           |           |
| Ülemiste polder      |          |          |        |              |           |           |
| Russalka            | -2.6 0.009 | -3       |        |              |           |           |
|                      | 1.4 0.164 | 2        |        |              |           |           |
|                      | 3.5 0      | 5        |        |              |           |           |
|                      | 2.2 0.0028 | -3    |        |              |           |           |
|                      | -2.5 0.011 | -3       |        |              |           |           |
| 10 years data (2005–2014) |          |          |        |              |           |           |
| Härjapea            | -2.1 0.036 | -3       |        |              |           |           |
| Lasnamäe            | -2.5 0.013 | -3       |        |              |           |           |
| Mustoja             | -2.3 0.021 | -3       |        |              |           |           |
| Pirita              | -2.1 0.034 | -3       |        |              |           |           |
| Rocca-al-Mare       | -3.2 0.001 | -3       |        |              |           |           |
| Ülemiste polder      |          |          |        |              |           |           |
| Russalka            | -2.7 0.008 | -3       |        |              |           |           |
|                      |          |          |        |              |           |           |
| Baseflow             |          |          |        |              |           |           |
| Härjapea            | -1.6 0.117 | -2       |        |              |           |           |
| Lasnamäe            |          |          |        |              |           |           |
| Mustoja             |          |          |        |              |           |           |
| Pirita              |          |          |        |              |           |           |
| Rocca-al-Mare       | -1.8 0.067 | -2       |        |              |           |           |
| Ülemiste polder      |          |          |        |              |           |           |
| Russalka            | -1.3 0.183 | -2       |        |              |           |           |
| Stormflow            |          |          |        |              |           |           |
| Härjapea            | -1.6 0.117 | -2       |        |              |           |           |
| Lasnamäe            | -1.9 0.052 | -2       |        |              |           |           |
| Mustoja             | -1.8 0.069 | -2       |        |              |           |           |
| Pirita              |          |          |        |              |           |           |
| Rocca-al-Mare       | -2.4 0.015 | -3       |        |              |           |           |
| Ülemiste polder      |          |          |        |              |           |           |
| Russalka            | -1.3 0.192 | -2       |        |              |           |           |

Table 2. Simplified trends detail with significance ranks

| Parameter     | Mustoja | Härjapea | Lasnamäe | Pirita | Rocca-al-Mare | Russalka | Ülemiste polder |
|---------------|---------|----------|----------|--------|---------------|----------|----------------|
|               | ST      | SF       | BF       | ST     | SF            | BF       | ST             |
| HC            | -3      | -2       | -2       | -3     | -2            | -3       | -3             |
| SS            | -2      | -2       | -3       | -3     | -2            | -3       | -2             |
| pH            | +2      | -        | +2       | +2     | -2            | +2       | +3             |
| BOD           | -3      | -2       | -2       | -3     | -2            | -2       | -2             |
| TN            | -3      | -3       | -3       | -3     | -3            | -3       | -3             |
| TP            | -3      | -3       | +2       | +2     | +2            | +2       | +2             |

ST – short term trend or 10 yr trend, SF – stormflow trend; BF – baseflow trend; -3 – significant downtrend (P-value < 0.05) , +3 – significant uptrend (P-value < 0.05), -2 – downtrend (P-value > 0.05 and < 0.2), +2 – uptrend (P-value > 0.05 and < 0.2), NA – data not available.
Suspended solids

About SS, the number of sites that have decreasing trends doubled from two in 19 year period to four in 10 year period indicating additional reduction in SS. Nevertheless, the levels of significance for this reduction were site specific. One significant long-term downward trend in Pirita outlet ($p = 0.012$) was found but the $P$-value has increased resulting less significant decrease in the past 10 years (Table 1). Rocca al Mare has improved in reducing SS since 2005 because it has a significant 10 year downward trend, and both storm and base flow play an equal role in this result (Table 2). Mustoja and Härjapea only show a decreasing trend at $P$-value 0.15 and 0.09, respectively. On the contrary, Russalka over the long-term reveals a less significant increase ($p = 0.164$) in SS which does not appear in the short-term, pointing no substantial reduction of SS after 2005. Only in Härjapea is a considerable contribution from baseflow as the result of a decreasing trend (at $p = 0.045$), whereas stormflow is more influential in Pirita. Overall, SS in Tallinn, though decreasing in 4 sites after 2005, neither have conclusive significant trends nor clear influence from stormflow and baseflow.

Instead, SS in Tallinn stormwater outlets varies a lot. Median and mean of 19 years of SS concentration do not exceed the national stormwater limit value of 40 mg/l in all basins except Rocca al Mare (Fig. 2). Nevertheless, the 90th percentile in most of the basins exceeds the limit, excluding Lasnamäe and Ulemiste Polder. Among them, Härjapea and Rocca al Mare have a large variation. It was noticed that Härjapea in 2001 and Rocca al Mare in 2005 and 2007 have a yearly median value of SS that exceeded the national stormwater limit value of 40 mg/l. The data of SS in all outlets except Ulemiste polder can surpass this limit during heavy rainfalls associated with the first flush and snowmelts in spring.

Suspended solids typically degrade water quality leading to different issues such as aesthetic problems, ecological degradation of aquatic environment, decline in fish resources, higher treatment cost, eutrophication, etc. (Bilotta, Brazier, 2008). They are associated with pollutants such as phosphorus, organic compounds and some heavy metals due to adsorption capacity and can have detrimental effects (Wakida et al., 2014). They are comprised of fine particulate matters suspended in storm runoff that have emanated from natural and anthropogenic sources. Natural sources are usually left unregulated because they are naturally present. These sources include forest fires, pollen, mould, etc. Anthropogenic sources are associated with air dust, street dust, salts used for deicing, poorly maintained garden beds / lawns and con-
struction activities. Therefore, one of the most influential controls of SS could be a reduction in the dry atmospheric deposition of air dust, aerosol and gas. In Tallinn, street sweeping has been initiated since a decade ago and it has reduced considerable dry atmospheric deposition. It is evident that the yearly concentration of particulate matters up to 10 µm (PM10) in South Tallinn and the city centre has decreased more than 50% during the 12 years since 2002, though this has decreased nearly 25% during the same period in the Haabersti area (EKK, 2015). This reduction in dry atmospheric deposition has probably revealed significant downward trends in Pirita and Rocca al Mare. Instead, the amount of SS is reduced in storm-water outlets due to renovation and the new construction of storm pipes. The major construction of stormwater pipes occurred in 2005 to 2010 (Tallinna Vesi, 2014). Many discharges from sewerage pipes were diverted to waste water treatment plants (WWTP), reducing the volume of possible WW connection to storm water systems and making storm water cleaner. In wintertime during the snow period, salt is used for de-icing road surfaces. It melts ice but it is also considered the main source for road dust. Chlorides keep damp road surfaces stable and the low temperature water is an enemy for asphalt. There is also more wear of asphalt surfaces if studded tyres are used for vehicles. The wear of the pavement caused by studded tyres is suggested to be ~ 47000 t/y, out of which 5–20% or 2100–10400 t/y is the spread into ambient air (Hääl, Sürje, 2006). This characteristic feature probably increases SS concentration in stormwater runoff, particularly during spring.

pH

Noticeably, pH has increasing trends in six out of seven outlets though the trends were less significant after 2005 in most of the basins. However, the pH is in between acceptable range (6–9) in all basins except Härjapea. Statistically significant long-term upward trends at p < 0.05 for pH are obtained in 5 out of 7 sites: Lasnamäe, Mustoja, Pirita, Rocca al Mare and Russalka, but except Pirita, P-values increased during last decade resulting less significant trends (Table 1). Ülemiste Polder has pH data from 2005 and SMK test for this period reveals significant upward trend. Stormflow is more influential for pH increase (3 out of 7 sites) than baseflow (1 out of 7 sites) (see Table 2). Only in Rocca al Mare, baseflow with upward trend at p = 0.051 is effective to attribute increased pH. In the stormwater outlets, the average pH over 19 years varied from 7.4–7.8 except in Härjapea where it has the highest level at 9 (Fig. 2). Moreover, 90th percentile pH ranged 7.8–10 with the highest in Härjapea.

Measurement of pH indicates acidity, basicity, alkalinity and neutrality in terms of hydrogen ions concentration in solution. The most preferable range of pH for the aquatic organisms is from 6.5 to 8, though the US EPA suggests 6.5 to 9 in freshwater as water quality criteria. The lower and higher ends of this range affected many species in terms of reproduction, growth and diseases (Ohrel et al., 2006). In Estonia, the pH of discharged water to the coastal area should be between 6 and 9 (RTI, 2013a).

A high pH has direct and indirect effects on aquatic life. As a direct effect, prolonged exposure to pH > 9.5 can damage outer surfaces such as fish gills, skin and eyes. It can also harm fish olfactory system, reducing detection capability for food, sex hormones and toxic chemicals. As an indirect effect, it has the proximity of ammonia toxicity. Since the fraction of unionised ammonia is more than 100 times greater than ionised ammonia when the pH is higher, toxicity exposure is increased during daylight hours.

The illustration for the increasing trend of pH in the respective sites is difficult to explain because there are different sources that can cause an elevated pH level. The main contributing factors in Tallinn that can be argued are storm drains from carbonate lithology, roads and impervious surfaces and sewage discharges that are primarily from industrial areas. The first contributing factor can be illustrated by increased alkalisation and acid deposition (Kaushal et al., 2013). Inorganic carbon fluxes increase due to development in urbanisation, changes in agricultural liming and mining, which contribute to bi-carbonate alkalinity in rivers and streams. Another influential factor for increased alkalinity can be related to acid rain and the weathering of carbonates in bedrock, soils and cement (Kaushal et al., 2013, Barnes, Raymond, 2009). It is because the weathering reactions produce alkalinity during acid neutralisation by geological materials. The rainfall data in Tallinn for the past 19 years indicate that the pH trend has been significantly decreasing as the SMK test result shown in Table 3. It is evident that precipitation in Tallinn is becoming more acidic. The Tallinn region is in the north-western Estonia, which belongs to Quaternary cover of a depth of 5 to 10 m. Sandy and clayey soils occur in the seashore area. Cambrian claystone, sandstone and siltstone are bedrocks in most of the basins, but the western area of Tallinn has Ordovician carbonate rocks such as Lasnamäe, Ülemiste and Russalka. The composition of these underlying rocks determines the major lithogenic element distribution of soils (Reintam et al., 1999). The high abundance of clay minerals and iron oxides are prevalent whereas oxides of Al, K, Ca and Mg are also enriched with clay minerals. Carbonate rocks from surrounding quarries have been used for limestone and dolostone for hundreds of years in Tallinn. Old churches, stone walls and building walls have been constructed with these rocks (Bityukova, 2006). Huge amounts of carbonate rocks have been mined and used for road construction material such as aggregates, gravels, fillers in asphalt and white road-marking mixture, for cement producing and for agriculture and garden liming (Notton, Söistra, 2010). The decay of these carbonate rocks can occur due to weathering from the acidity of rainfall and, as a consequence, alkalization with pH can increase in the runoff.

The volume of traffic has also increased in this region, which damages the road surfaces, particularly in winter when studded tyres are used and de-icing salts is applied. This accelerates the weathering of road surfaces.
When the volume of traffic is increased, it influences air pollution because the concentrations of NO\textsubscript{2}, SO\textsubscript{2}, CO and PM10 are highest during working hours. The road areas produces alkaline dust, which can increase the pH found in tree bark in the range from acidic to subneutral, and this dust is highest in the centre of Tallinn (Kesklinn) rather than suburbs such as Rahu (an industrial area) and Õismae (a residential area) (Marmor, Randlane, 2007). In addition, sewage discharge contains carbonate compounds because the industrial process involved in making fertilisers, plastics, ceramics, rubber, paint, glass, glass fibre and sugar uses carbonate rocks. Also, the discharge of soap and detergents most likely increases the alkalinity in water considerably.

In Härjapea, the yearly 90th percentile of pH ranged 8.4–10.8 and yearly mean ranged 8.1–9.78 over 18 years, which is the highest average pH of all basins; however, there is no significant trend observed in this outlet. From 1996 to 2002, the annual pH level increased to a maximum of 9.78, and it began to decline from 2003 onwards. The decline is not as steady as in 2007 when there was a high pH of 9.58 and in 2013, it reached 8.87. This region includes the areas of Põhjaväila and the streets of Lootsi and Ahtri along with suburbs of the central city. The Old Town with its historic buildings, medieval stone walls, churches, stone pavements and old houses have the option of partial discharge to this outlet. On the other hand, there is a significant possibility of weathering of the carbonate rocks in these areas. On the other hand, due to the increased traffic volume, dust pollution has increased in the city area; for example, in 2006, the 24h limit value of 50 μg/m³ was exceeded on 42 occasions in the Keskklinn area (Marmor, Randlane, 2007). Moreover, the factories and old sewerage system have the potential to discharge sewage containing carbonate compounds. In recent decades, the city government has made many efforts to protect and improve the Old Town environment in Tallinn. Roads were reconstructed, traffic was limited, some small enterprises were removed to the outlying areas, etc. (Bityukova, 2006). This is a probable explanation for the increases and decreases during this period.

**Biological Oxygen Demand (BOD)**

BOD concentration, that seldom exceeded the permissible value, has remarkable decreasing trend in nearly half of the studied outlets. Within the period of 19 years, the 90th percentile of BOD concentrations are below the stormwater national limit value (15 mg/l), excluding Rocca al Mare which reached 19 mg/l (Fig. 2). Mustoja and Rocca al Mare exceeded this limit value in the years before 1997, but since then the yearly median values have been within this limit. Statistically significant downward trends have been noted for BOD\textsubscript{5} in 3 basins: Härjapea, Mustoja and Pirita (Table 1). Pirita’s downward trend is associated with baseflow (\( p = 0.197 \)) and Mustoja’s with stormflow (\( p = 0.166 \)), while Härjapea does not show trends in stormflow and baseflow. Rocca al Mare has significantly decreased BOD in the 19 year period but it has not shown a trend in the past 10 years. This characteristic feature can be associated with an increasing trend in the stormflow at \( p = 0.056 \) (as in Table 1) because the trend for stormflow reveals that during this period only storm events caused a rise in BOD concentration levels against the long term decreasing trend.

Naturally, fallen leaves and decaying vegetation increase the oxygen demand in runoff. In addition, anthropogenic activities such as grass clippings, human, birds and animal excreta, hydrocarbons, engine coolants and antifreeze containing ethylene glycol and propylene glycol can exert high BOD in stormwater (Bingham, 1993, Erickson et al., 2013). BOD in urban runoff can be directly correlated with watershed development and percentage impervious surfaces (Erickson et al., 2013). Göbel et al. (2007) found in many research works that BOD is higher in runoff in traffic areas than in runoff from gardens, lawns and cultivated areas. However, a significant load originates from roofs due to bird excretion and dry atmospheric deposition. In Tallinn, a significant decrease in BOD is mainly observed in three watersheds, such as Härjapea, Mustoja and Pirita. According to the environmental reports of AS Tallinna Vesi (Tallinna Vesi, 2014), sewerage networks were constructed at Mustamäe (region of Mustoja watershed), Pirita and the city centre (regions of Härjapea watershed). In 2004, the sewerage connections for the residential area of Lilleküla and also catering for the Tondi area were constructed under Mustamäe road. In the districts of Merivälja, Lilleküla and Mustamäe, some streets were directly linked to combined sewers leading to WWTP in 2005. This has considerably reduced BOD and other organic pollutants in those areas. The large volume of sewerage extension occurred between 2005 and 2010. Moreover, the municipalities initiated street sweeping, which has remarkably prevented organic matters from mixing with runoff. The Pırıta district has mainly improved the sewerage connections to reduce the pollutant load in the outlet. However, in Rocca al Mare, BOD concentration is not well controlled and it showed increasing volumes during storm events, indicating possible wash-off of animal waste from upstream Tallinn Zoo.

**Nutrients**

In the case of nutrients, the results reveal nearly half of the basins have decreasing trends and two basins considerably improved TN reduction over the last 10 years. From 2005 to 2014, 4 statistically significant downward trends of TN (at Mustoja with \( p \approx 0.05 \), Härjapea, Pirita and Russalka) and 2 statistically significant downtrends of TP (at Mustoja and Pirita) have been observed (Table 1 and Table 2). However, the TN trend for the long-term period in Pirita does not appear and in Härjapea it is less significant. It indicates that there have been considerable reduction activities for N in these two basins since 2005. On the contrary, Lasnamäe’s long-term trend is less significant but does not appear to be a short-term trend, suggesting no considerable improvements in N concentration after 2005. Nevertheless, it is interesting to know that Härjapea has begun to see a rise in TP for the past 10 years and baseflow (at \( p = 0.089 \)) is mainly contributing to this result. In most cases, the decreasing trend in nutri-
ents is formed during stormflow.

Nutrients’ 90th percentile concentrations, at 5.1–16.9 mg/l of TN and 0.2–1 mg/l of TP, do not exceed the stormwater limit value. However, the concentrations are over levels causing eutrophication (Fig. 2). The mean TN and TP in the inlet of WWTP Tallinn in the last 10 years were 44.6 and 6.8 mg/l respectively (Tallinna Vesi, 2004–2014), whereas stormwater runoff TN and TP in stormwater outlets have varied between 3.5–8.4 mg/l and 0.1–0.6 mg/l respectively, as in Fig. 2. According to Estonian regulations, the stormwater limit values of TN and TP are 45 mg/l and 1 mg/l (RTI, 2013a) which is far higher than coastal water class 5 limit values (0.97 mg/l of TN and 0.67 mg/l of TP) and river water limit values (6 mg/l of TN). Sometimes, the concentrations during heavy rainfall with first flushes almost reached the mean concentrations of inlet WW with yearly 90th percentile of 36.9 mg/l (TN) and 4.8 mg/l (TP). Some years, the areas of Härjapea and Pirita reached the stormwater TP limit value (1 mg/l) with first flushes almost reached the mean concentrations of inlet WW at 90th percentile of 0.9 mg/l and 1 mg/l, respectively. Similar to river and streams, natural and anthropogenic sources contribute to nutrients in stormwater runoff. Natural sources for nutrients in the urban area include ground water, atmospheric nitrogen and vegetation, e.g. some N-fixing plants. Natural biochemical processes during the decomposition of plant and animals can occur in the watershed to contribute to nutrients.

Table 3. SMK-test results for trends analysis of air quality and precipitation in Tallinn (Statistics Estonia, 2015, EEA, 2015).

| Parameters                  | Period          | Annual average range | SMK Stat | P- Value | Sig. rank |
|-----------------------------|-----------------|----------------------|----------|----------|-----------|
| Tallinn Air Quality Monitoring |                |                      |          |          |           |
| SO₂, µg/m³                  | 1995 to 2014    | 1.1–8.0              | -4.546   | <0.001   | -         |
| NOₓ, µg/m³                  | 1995 to 2014    | 14.4–101.6           | -4.385   | <0.001   | -         |
| CO₂, mg/m³                  | 1995 to 2014    | 0.2–1.7              | -5.086   | <0.001   | -         |
| PM₁₀, µg/m³                 | 2006 to 2014    | 12.3–29.5            | -2.488   | 0.013    | -         |
| Tallinn Precipitation Pollutants |            |                      |          |          |           |
| pH                          | 1995 to 2014    | 5.6–7.4              | -3.943   | <0.001   | -         |
| SO₂-S, mg/l                 | 1995 to 2014    | 0.4–9.6              | -4.207   | <0.001   | -         |
| NOₓ-N, mg/l                 | 1995 to 2014    | 0.2–1.1              | 1.693    | 0.091    | 2         |
| Cl, mg/l                    | 1995 to 2014    | 0.7–4.2              | -0.905   | 0.365    | -1        |
| NH₄-N, mg/l                 | 1995 to 2014    | 0.1–0.6              | 1.111    | 0.267    | 1         |
| Precipitation mm            | 1995 to 2014    | 39.2–78.0            | 0.062    | 0.951    | 1         |
| HCO₃, mg/l                  | 2003 to 2014    | 0.1–56.4             | -2.09    | 0.037    | -3        |

Overflows from sewer systems and the leaching of sewerage due to poor drainage systems can be expected in some areas of Tallinn. This century, there has been extensive stormwater and sewerage network construction and renovation in Tallinn, which has lasted almost a decade (Fig. 3). According to environmental reports from AS Tallinna Vesi (Tallinna Vesi, 2014), 97% of Tallinn area was connected to the public sewerage system by 2006 and, in collaboration with the City of Tallinn, the company had covered 99% with the public sewerage network by the end of 2010.

In the urban environment, dissolved inorganic nitrogen (NOₓ-N and NH₄-N) is emitted from vehicle exhausts and the combustion of fossil fuels such as coal and oil. These emissions are accumulated as wet and dry deposition in the atmosphere. Wet forms are rain, snow, fog and hail, while dry forms are particulates, gases and droplets. They are either infiltrated into the soil or washed off into the drainage system. The air quality monitoring records of Tallinn as in Table 3 show that there is a significant decrease in the dry deposition of nitrogen dioxide in the atmosphere. However, the wet deposition does not show the same behaviour because there has not been a significant decrease or increase trend for NOₓ-N and NH₄-N in precipitation in the past 19 years (see Table 3). Instead, the nitrogen compound has tended to increase during this period (Table 3). The wet deposition source of dissolved nitrogen is comparatively small in quantity, which cannot contribute to the nitrogen level in stormwater runoff. Similarly, wet deposition in the precipitation of dissolved phosphorus is negligible at the median concentration of 0.005 mg/l during the record period from 1995 to 2002 (EEA, 2015). The atmospheric deposition varies with land use patterns and the volume of emissions. It is not valid in every area of Tallinn and the nitrogen load is high where the number of vehicles and impervious surfaces is significant (Hou et al., 2012, Shen et al., 2014).

Another potential source for stormwater runoff nutrients can be related to fertiliser application in lawns and gardens. Runoff drainage from agricultural and residential lands has much higher nutrient yields than runoff drainage from forested land (Roberts, Kolosseus, 2011), especially during storm events. Lawns and gardens are larger contributors of N and P than streets. Indeed, streets supply particulate nutrients because they are large source of suspended solids (Waschbusch et al., 1993). In agriculture, there is a lowered application of organic and inorganic fertilisers in Estonia (Iital et al., 2010). Instead, the utilisation of organic fertiliser has reduced while usage of inorganic fertiliser has increased (Statistics Estonia, 2015). The constituents that nitrate from them are readily

Fig. 3. Cumulative stormwater and sewerage network extension and reconstruction in Tallinn. (Tallinna Vesi, 2014)
soluble and can be reduced through denitrification. It is one of the likely reasons why the trends of nutrients in Ulemiste Polder and in Russalka have decreased. The upstream basin of the Ulemiste polder sites has agriculture fields and the overflows during heavy rainfall and snowmelt are diverted to the Russalka outlet. Both the Mustoja and Pirita basins have private yards covering approximately 18% of the basin area where the roof runoff as well as rainfall water flows over the yards, with some infiltrating and the remainder flowing into drainage inlets.

There are other possible anthropogenic activities for the diffuse sources of nutrients in stormwater runoff. Rainfall is more concentrated with nutrients once it flows through the surfaces e.g. roofs where there are organic pollutants like leaves, animal or bird excreta, flowers and pollen (Göbel et al., 2007). Vegetation in drains and ditches enhances decay and the decomposition of organic compounds to increase nutrients.

Stormflow is more susceptible to a decrease in nutrient trends than base flow, as shown in Table 2. Compared to the past, the overflow frequencies and volume that divert to storm pipes during storm events are less in quantity, and the reconstruction and construction of sewer pipes have been mainly attributed to this result. It is likely to be a reason that significantly enhances or considerably decreases the volume of nutrients. However, the nutrient concentration in Tallinn stormwater requires study due to the unusual exceeding of limit values, and further investigation can be directed towards determining such patterns.

Conclusion

Downward trends for HC were detected; 6 out of 7 investigated basins have statistically significant downward trends over the past 10 years. The possible reason is street sweeping to keep the roads and parking areas clean and sand and oil trappers on the drainage inlets, which prevent HC from entering the drainage pipes.

SS downward trends are observed in 4 sites but statistically significant downward trends include one in Rocca al Mare for the past 10 years and one in Pirita for the last 19 years. Reduced particulate matter in the air, the connection of WW to sewerage pipes and reduced salt de-icing practice have probably reduced SS in stormwater.

Increased pH trends in most of the basins, except Härjapea have likely been caused by increased alkalisation due to acidic precipitation, the weathering of carbonate aggregates, discharges and alkaline dust from roads. However, Härjapea stormwater outlet has shown the highest average pH level at 9. One potential reason might be sewage discharge and requires further investigation.

Statistically significant decreasing trends for BOD of three outlets (Pirita, Härjapea and Mustoja), TN of four outlets (same as of BOD and Russalka) and TP of two outlets (Pirita and Mustoja) were observed from 7 outlets over the last 10 year period. Two stormwater outlets, Mustoja and Pirita have shown significant decrease for BOD, TN and TP. The improvement in sewer networks, street sweeping, the decline in the use of agricultural land, and in turn fertilisers have favourably influenced the result.

In general, most of the studied parameters are in decreasing trends except pH which requires more attention to ensure the potential causes and most of the basins’ trends have been influenced by stormflow rather than baseflow. Stormflow has a greater influence on HC, SS, pH, BOD, TN and TP trends than baseflow.

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