

Undeclared Sources: Identification and Characterisation of Diffuse Pollution from Urban Stormwater

1 INTRODUCTION

Densification of urban areas due to growing population contributes to risk of increasing the pollutant loads in stormwater and concomitant contamination of lakes and water bodies in the urban environments, in a direct conflict with the European Water Framework Directive (WFD)(1). This was recently further stressed by the verdict of the European Court regarding the Weser dredging case (2). Roads, traffic and building materials in urban areas, exposed to precipitation and weathering, are the sources of such pollutants. Upstream source control is one opportunity to improve stormwater quality and protect receiving waters. However, to apply such controls, comprehensive information on pollution contributions from these surfaces is needed. Furthermore, a deeper knowledge of the stormwater chemistry will increase the predictability of the pollutants transport and changes, which may be essential for the development and application of effective stormwater treatment technologies serving to reduce pollutant discharges and contribute to complying with the WFD. Sources, transport and fate of urban contaminants have been pointed out by the scientific community as a field where a better understanding is needed (3).

Stormwater runoff represents a source of diffuse pollution. Roofing materials, façade cladding/paints, roads, and other surfaces present in the urban environment leach out the pollutants, which are characteristic for individual materials, and end up in stormwater runoff (4-6). One can hypothesize that the diverse origin of these pollutants and their sources, as well as the challenges in conducting quality measurements in stormwater from specific sources, led to the lack of systematic pollution source controls. Although roads and traffic are recognized as the most potent source of urban runoff pollution (7), building materials and structures are also emerging as causes of high pollution impacts (4, 8, 9). Extensive past efforts have focused on zinc, copper and lead roofing materials (10-12), and roof runoff quality as a potential source of potable water (13). With respect to organic micro-pollutants and their sources in the urban environment, the research is still in the initial discovery stage. Thus, a systematic approach is missing, in which each material and surface in the urban environment would be seen as a potential source contributing to the overall stormwater quality.

The distribution of trace metals among different physicochemical fractions ("truly" dissolved = free metal ions; colloidal = dissolved particles; particulates) and species determine the metal fate in BMP (best management practice) treatment facilities and in the receiving waters. Many of the pollutants and contaminants in stormwater runoff are associated with particles, and thus the particulate phase in stormwater runoff has been thoroughly investigated and characterised in numerous studies (e.g., 14, 15). Furthermore, particles, with the associated pollutants, are more easily retained in BMP facilities compared to the dissolved species. However, the dissolved phase (operationally defined as passing through a 0.45 µm filter), often contributes to over 50% of the total metal concentration, which may help explain the some poor treatment efficiencies (16). Thus, the knowledge of fractionation and speciation characteristics of the stormwater dissolved phase is essential for further development of improved high-

performance stormwater treatment techniques that are aiming at reducing the metals also in the dissolved phase.

Summary: There is a lack of knowledge of pollutant sources and the corresponding loads, originating in the urban environment, and the effects of various types of surfaces on, and contributions to, the chemistry of urban stormwater runoff.

2 PROPOSED RESEARCH FRAME

2.1 HYPOTHESES

- Upstream characterisation and source control of the major and minor sources of pollutants contributing to the deterioration of the urban stormwater quality is essential for protecting the receiving waters.
- Knowledge of specific sources that contribute the most to the total pollutant load in stormwater runoff creates an opportunity to prevent deterioration of receiving water quality, which is the risk associated with densification of urban areas.
- Thorough knowledge of the amounts of pollutants that can be expected to be released from typical surfaces in the urban environment will improve process modelling.
- Better understanding of the urban stormwater chemistry will facilitate the development of high-performance stormwater treatment techniques.

2.2 Aim

The overall aim is to increase the knowledge of the contributions of organic and inorganic pollutants from specific sources in the urban environment to stormwater, and identify potential environmental risks on the basis of element speciation, to enable informed decision making in planning and building urban structures and BMP treatment facilities.

2.3 WORKPLAN

To achieve the stated goals, the work has been divided into five work packages (WPs), several of which run in parallel and depend on the same core activities, such as laboratory experiments, pilot roofs and facades, and field measurements and modelling. The WPs are described below; connections of different activities to individual WPs are summarised in Appendix J.

WP1 –ORGANIC MICRO-POLLUTANTS IN THE BUILT ENVIRONMENT

Background: The current knowledge of the release of organic pollutants from the urban environment is limited. Material flow analyses have been conducted and identified building materials and structures as potential sources (17), but relatively little has been published on lab and field studies. Andersson-Wikström (18) conducted a lab study, screening a range of roofing materials, and the results indicated a range of organic pollutants (e.g. nonylphenol, biocides, naphthalene), from various materials. The study also revealed that materials with similar composition exhibited rather different substance release profiles.

Objective: Identify diffuse sources of organic pollutants released to the environment via stormwater and quantify the fluxes of these pollutants from well-characterised catchments with homogenous land use.

Method: *Field measurements* will be carried out in homogenous catchments of different characteristics, e.g. commercial area, car park and a road. The sampling will cover a full-year cycle, including the spring (snowmelt), summer and autumn. Automatic flow-weighted sampling will be applied to collect high quality data. The analysed parameters will include phthalates, biocides, nonylphenol and -etoxilates, PAHs, etc. These measurements will indicate, which land uses contribute more of these pollutants than others.

A range of materials found in the urban environment will be tested in leaching experiments in a **lab study**. These include roofing materials (e.g., coated steel, clay and cement tiles, bitumen sheets), sidings/façades (e.g. painted wood, stucco, bricks), and other common materials. The selected materials will complement the ones already analysed by Andersson-Wikström (18). Small pieces of the materials will be exposed to synthetic rainwater, which will be prepared on the basis of precipitation chemistry data. The materials will be analysed for a broad group of organic pollutants (see above). A thorough literature review and database searches (e.g. bastaonline.se, sundahus.se, byggvarubedomningen.se) will help select materials to be studied and parameters to be analysed.

Based on the lab study, the number of materials will be reduced to about 5 roofing materials and 5 façade coatings applied in a **pilot scale field study** conducted outdoors. Runoff from the miniature roofs and sidings will be analysed with focus on the pollutants identified in the lab study. Precipitation and dry deposition will also be measured over a period of one year to cover seasonal changes.

Outcome: 2 journal, 1 conference articles

WP2 – INORGANIC MICRO-POLLUTANTS IN THE BUILT ENVIRONMENT

Background: Characterisation of the dissolved fraction in stormwater is only sparsely reported in the literature (19, 20), even though such information is essential for estimations of metal toxicity and mobility. The increased concern about the diffuse pollution conveyed by stormwater compels stakeholders to consider stormwater treatment more often. The knowledge of the chemical speciation of metals in stormwater from specific surfaces, as well as at the catchment scale, is therefore necessary to advance the systematic approach to the development of more effective stormwater treatment systems.

Objective: Characterise the size fractionation of metals in the dissolved phase in stormwater originating from specific surfaces and materials in the built environment, and at the catchment scale.

Method: Speciation/fractionation measurements of the dissolved phase ($<0.45\ \mu\text{m}$) will be conducted on stormwater samples from catchments with well-defined land use (commercial areas, roads, parking lots), collected during the **field sampling** campaigns described in WP1 for all seasons of the year. Separate sets of automatic samplers, with glass or plastic bottles, will be used for organic (WP1) and inorganic parameters, respectively. 5-10 samples from each rain/snowmelt event and site will be collected and analysed, throughout the whole year, on 10-15 occasions. Speciation/fractionation measurements will also be carried out on runoff from **pilot roofs** with copper and zinc sheets. Size fractionation of metals in the dissolved phase will be determined applying e.g. ultrafiltration on single samples and diffusive gradients in thin films (DGT) passive samplers deployed in situ.

Outcome: 1 journal and 1 conference article

WP3 – EFFECTS OF AGING AND CLIMATE ON POLLUTANT RELEASE FROM BUILT STRUCTURES

Background: The releases of metals as well as organic pollutants are known to be affected by aging and weathering. The effect of pH and material age on the release of metals from copper and galvanised iron has previously been studied by Wicke et al. (21) in both the field and laboratory simulations. Corrosion and release of copper and zinc from roofs depends on pH and sulphate concentration in the precipitation (21-23), but other ambient factors were only studied to a smaller extent. The releases of organic pollutants (such as phthalates and nonylphenol/-etoxilates) are known to be affected by temperature, and UV radiation is assumed to contribute to aging and breakdown of materials. A better understanding of the processes affecting the release of organic and inorganic micro-pollutants is important for estimating the current and future stormwater quality.

Objective: Evaluate the importance of climatic factors and materials aging on the release of micro-pollutants from roofing and façade materials.

Method: Based on WP1, materials will be selected for further studies of organic micro-pollutants. In a **lab study**, a climate chamber, commonly used, e.g. , in the paint and coatings industry, will be used to accelerate the aging of the selected materials and study the variations in pollutant release over time. In addition, various climates will be applied to predict the effects of future climate changes in different parts of the world. The effects of rain intensity and temperature on pollutant release will also be studied. Measurements from **pilot roofs and facades** made for WP1 and WP2 will be conducted in several sampling campaigns over the duration of the proposed project to complement the lab results on the aging effects.

Outcome: 2 journal, 1 conference articles.

WP4 – THE EFFECT OF SEWER PIPE MATERIAL ON CHEMICAL SPECIATION AND TREATABILITY

Background: Initial investigations (unpublished results) have shown that during the transport of stormwater in concrete pipes, fine particles coalesce into larger particles. Analogous tests in PVC pipes have not indicated such phenomena. The storm sewers in Sweden are reaching their design lifetime and are successively replaced, in some cases by plastic pipes (e.g. PVC, PP or PE). The efficiency of the stormwater treatment will depend not only on the selected technology, but also on the chemical speciation of the metals present in stormwater. The knowledge of the stormwater geochemistry is therefore essential for the development of high-performance treatment processes.

Objective: To investigate how the choice of pipe material affects the stormwater quality and geochemistry with respect to metal speciation/fractionation.

Method: **Lab experiments** will be conducted to study the particle size distribution (PSD) and (trace) metal speciation/fractionation in semi-synthetic stormwater transported in pipes made of different materials, including concrete, plastic, corrugated steel and weathered steel. Metal speciation/fractionation will be estimated from DGT measurements and ultrafiltration with various cut-offs, two techniques commonly applied for this purpose (24). The results will be complemented with **field sampling** and measurements of stormwater in catchments having sewer pipes of various materials.

Outcome: 1 journal, 1 conference article.

WP5 – FUTURE STORMWATER QUALITY

Background: Modelling of stormwater pollutant loads, using available software, such as e.g. SEWSYS (25) and StormTac (26), is frequently applied by local authorities and consultants as a substitute for costly field sampling. The use of sometimes old or general default values in combination with the lack of field data for model calibration results in high uncertainties of the simulated pollution loads (25). Among others, the extension of the list of pollutants and testing the models against field data have been pointed out as areas, where stormwater modelling can be improved (27).

Objective: To improve the accuracy of stormwater quality modelling of organic micro-pollutants in urban catchments and to evaluate various scenarios of future quality of stormwater.

Method: The information obtained in WP1-4 will be used for *modelling* future stormwater quality. Beside the data obtained in WP1-4, datasets from extensive sampling in two other Swedish cities will also be available. These datasets cover all seasons of the year, various catchment characteristics, and a wide range of analysed parameters, and can be used for model verifications. For this approach it will be necessary to carefully characterise building materials and structures in the modelled catchments.

Outcome: 2 journal, 1 conference articles.

3 RESEARCH ENVIRONMENT

3.1 APPLICANT AND RESEARCH TEAM

The main applicant, **Prof Maria Viklander**, Urban Water, Luleå University of Technology (LTU), is one of the leading researchers in the urban water field with focus on urban hydrology and systems in cold climates. She will be main supervisor for the PhD student applied for in this project. **Prof Jiri Marsalek** is internationally recognized as one of the leading researchers in urban drainage, and will contribute his comprehensive knowledge of urban drainage and his connections to an extensive international network of researchers. **Dr Helene Österlund** is a young researcher with a PhD in geochemistry and will contribute her knowledge of analytical and aquatic chemistry. Marsalek and Österlund will be co-supervisors. **Dr Anna-Maria Perttu** is a young researcher with a PhD in water resources engineering and she will strengthen the proposed project in the area of urban hydrology and modelling. **Lic Tech Kerstin Nordqvist** is a senior laboratory engineer with more than 30 years of experience within the field of urban water engineering. She will assist in the lab and field.

3.2 LABORATORY FACILITIES

The Urban Water research group is a part of the Environmental Laboratory at LTU. The lab is equipped with clean benches promoting a low contamination atmosphere, which is safe for sample handling and preparation. The research group possess instruments for pH, conductivity, turbidity, N and P analysis, as well as a brand new particle size analyser. Equipment for field measurements and sampling of stormwater is also available but needs be supplemented. Furthermore, there is an outdoor test site which is available for the pilot scale studies.

3.3 NATIONAL AND INTERNATIONAL COLLABORATION

The Urban Water research group has established an extensive research collaboration network within the field of urban drainage, including some world-leading research groups and experts at

Monash University, Australia; Norwegian University of Science and Technology (NTNU) Norway; University of Innsbruck, Austria and INSA de Lyon, France. For the proposed project, collaboration with the following experts will be particularly valuable: Prof. Lian Lundy, Middlesex University, UK; research focus on urban stormwater management, particularly the transport and environmental behaviour of urban pollutants in both water and sediment; Dr. Louise Sörme, Project manager at Statistics Sweden, assessing material flow balances and substance flow analysis; and, Dr. Thomas Larm, StormTac, Sweden, developer of the StormTac stormwater modelling software.

3.4 STAKEHOLDER COLLABORATION

The proposed project will be carried out in cooperation with City of Stockholm Environment & Health Administration and NCC as well as the national research program Stormwater&Sewers (S&S). The Urban Water research group at LTU is responsible for S&S which is financed by the Swedish Water and Wastewater Association (SWWA) and municipalities in Northern Sweden (Luleå, Umeå, Sundsvall, Örnsköldsvik and Östersund). S&S produces newsletters, handbooks, and hold seminars and national conferences to continuously report on the activities and new knowledge resulting from this collaboration. Stockholm Environment & Health Administration will use the results from the proposed project for their preparation of guidelines to sustainable stormwater management and the action plan for polluted surface waters. In addition they contribute results from previous projects on diffuse pollution sources. NCC develops and builds future environments for working, living and communication. Their vision is to renew our industry and provide superior sustainable solutions.

4 DELIVERABLES TO SOCIETY AND USERS

4.1 SCIENTIFIC VALUE

The results presented in this application will give deeper understanding of the extent of micro-pollutants in the urban environment, applicable not only in Sweden, but worldwide. The research will increase the knowledge about the chemical speciation of metals which will be a tool in further research on high-performance stormwater treatment technologies. Some climatic factors influencing release of organic as well as inorganic contaminants from building materials and structures will be reported to complement the earlier published data, and will assist the manufacturing industry in developing new environmentally friendly products.

4.2 PUBLICATION OF RESULTS

Seven journal papers will be published in peer-reviewed journals. Open access will be assured by choosing at least Green Open Access journals for publications, and post-prints will be published in the LTU research database. It is proposed to pay open access fees for four of the articles produced. The results will also be presented at international and Scandinavian conferences (e.g. arranged by IWA Sweden, Novatech 2019 and DIPCON 2018).

4.3 DELIVERABLES TO USERS

The research results will be communicated to stakeholders within the urban water area in Scandinavia through the communication program of S&S. All research results considered of high value to stakeholders will be summarised in technical briefs, delivered through the website of the research group and advertised in the Stormwater&Sewers newsletter (published 3-4 times a year to >1000 subscribers). Another important channel for communications is the

national professional journals (e.g. Miljöaktuell, Cirkulation and Svenskt Vatten). Results will also be presented at Stormwater&Sewers conferences arranged for practitioners.

5 SOCIETAL VALUES

This project will identify and quantify diffuse sources of micro-pollutants found in building materials and structures, which are leached by precipitation and discharged into the environment and receiving waters with stormwater runoff. By developing this specific knowledge, informed decisions concerning controls of pollutant discharges from the built environment can be taken. The identified hot-spots of polluting materials can be phased out with new regulations. By considering the results of this project in life-cycle environmental certification systems for building materials, the existing buildings can be up-graded or redesigned to reduce their environmental footprint, and new buildings with reduced environmental impacts can be implemented, making a strong contribution to a non-toxic environment. This project will also identify appropriate treatment systems for the various types of pollutants identified, which should reduce pollution loads from the existing and future urban areas. Besides the environmental quality objectives and a non-toxic environment, this project will contribute to a high-quality built environment, good-quality groundwater and flourishing lakes and streams, resulting from cleaner stormwater discharges.

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