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Emissions from Building Materials—A Threat to the Environment?

Daniel Wicke [1,*], Roberto Tatis-Muvdi [1], Pascale Rouault [1], Patricia Zerball-van Baar [2], Uwe Dünnbier [2], Mirko Rohr [3] and Michael Burkhardt [3]

[1] Kompetenzzentrum Wasser Berlin, 10709 Berlin, Germany; robetatis@yahoo.com (R.T.-M.); pascale.rouault@kompetenz-wasser.de (P.R.)
[2] Berliner Wasserbetriebe, 10179 Berlin, Germany; Patricia.Zerball-Vanbaar@bwb.de (P.Z.-v.B.); uwe.duennbier@bwb.de (U.D.)
[3] Institut für Umwelt- und Verfahrenstechnik (UMTEC), Ostschweizer Fachhochschule, 8640 Rapperswil, Switzerland; mirko.rohr@ost.ch (M.R.); michael.burkhardt@ost.ch (M.B.)
* Correspondence: daniel.wicke@kompetenz-wasser.de

Abstract: A large variety of substances are used in building materials to improve their properties. In recent years, attention to organic additives used, for example, in renders, façade paints or roof sealing sheets has increased as these compounds have been detected in urban stormwater runoff and surface waters. In this paper, we show the extent of emissions induced by rain events in two study sites in Berlin. For this purpose, stormwater runoff from roofs, façades, and in storm sewers was sampled and analysed over a period of 1.5 years in two residential catchments. Results show that, in particular, the biocides diuron and terbutryn from façades, the root protection agents mecoprop and MCPA in bituminous sheeting, and zinc from roofs and façades reach concentrations in the stormwater sewer that exceed limit values for surface waters. Additionally, transformation products of the biocides were also detected. However, many other analysed substances were below the quantification limit or inconspicuous in their concentration levels. The emissions, modelled with the software COMLEAM, demonstrate that in urban areas the limit values in smaller surface waters are exceeded during wet weather. Furthermore, the orientation of the buildings to wind-driven rain is essential for the emitted load from façades. The calculated mass balances of both catchments show that a major portion of all substances remains on-site and infiltrates diffusely or in swales, while the remaining portion is discharged to stormwater sewers. For example, in one of the two study sites, <5% of diuron emissions are discharged to surface waters. Infiltration, in particular, is therefore a crucial pathway of pollution for soil and groundwater. Measures for source control are proposed to mitigate the leaching of environmentally relevant substances from construction materials.

Keywords: stormwater runoff; micropollutants; diffuse pollution; environmental monitoring

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1. Introduction

The quality of stormwater runoff in urban areas is influenced by a number of factors, such as the built environment, operational conditions, and maintenance. A long-known source of pollution is traffic, which is responsible for the emission of solids and microplastics (e.g., tire and brake abrasion), heavy metals (e.g., copper, zinc), oil residues, and organic trace substances (e.g., PAH) [1]. In addition, buildings with large surface areas in contact with precipitation may release substances to the environment by surface runoff [2,3]. For example, in Berlin, 38% of the total impervious surface area connected to the drainage system is roofs. During rain events, environmentally relevant substances can be leached, depending on the substance properties and material composition. Substances controlling root penetration, such as mecoprop and the heavy metal copper from metal sheets, have been detected in stormwater runoff for a long period [4,5]. Façades are affected by wind-driven rain that results in façade runoff [6]. Various substances can leach out of façade materials,

such as zinc used as zinc oxide pigments and biocides used as film preservatives in coatings, e.g., diuron, terbutryn, or isoproturon [7]. Although the volume of façade runoff is much lower compared with horizontal surfaces, such as roofs or roads, concentrations of these substances can be high [7,8]. Factors affecting the leaching, such as direction of exposure, rainfall intensity, or polymer-controlled diffusion, have been investigated in several studies using test specimens in the laboratory or under natural weather conditions [8–10].

Depending on the pathway of runoff from buildings to the stormwater sewer system, these substances can subsequently be conveyed directly to stormwater and aquatic systems. Consequently, trace pollutants related to building materials have been found in urban stormwater runoff of residential and commercial areas, as well as in a receiving urban stream in Berlin [11] and in stormwater runoff of residential catchments in France [12], Denmark [13], and the US [14]. These studies clearly show that building-related substances are widespread in stormwater runoff. However, the presence in stormwater runoff is mostly reported without knowing the source materials and their composition in detail. Mass balances of emitted substances are therefore lacking. For this purpose, an investigation under natural weather conditions at field sites, taking into account all potential sources (e.g., roofs, façades) and their material properties, is essential.

To gain further knowledge on the release mechanisms or to assess the environmental impact of trace pollutants from buildings, modelling is a promising tool. So far, biocide risk assessment has relied on emission scenario documents (ESDs) to provide an estimate of environmental impact [15]. ESDs are based on simple linear release concepts without reflecting dry and wet weather conditions. In combination with field or laboratory studies, leaching models that reflect the emission behaviour of buildings under real weather conditions are promising tools to derive and extrapolate the environmental impact. With assessments of roof and façade runoff, specific source-control measures can be evaluated.

In this study, the building materials, as potential sources of stormwater pollution, are linked with the pollution in urban stormwater runoff. Field investigations at two study sites in Berlin were carried out, which included event-based sampling of façade and roof runoff and corresponding stormwater runoff from the catchment area. Furthermore, leaching from these two test sites and the impact on surface water was modelled and compared to the monitoring data from the field sites to identify relevant parameters and derive mitigation measures.

2. Materials and Methods

2.1. Field Study

2.1.1. Test Sites

Two new development sites (site A and B, about 120 apartments each) were selected in Berlin (Germany), both constructed in 2017 (1–1.5 years prior to the start of the monitoring campaign). The total catchment area is 12,500 m^2 for site A and 6000 m^2 for site B. The catchments represent typical architecture for multi-storey apartment buildings with a separated sewer system. Both sites include plastered and painted façades (external thermal insulation composite system) and flat roofs covered with bituminous sheets, combined with extensive green roofs at site B. Several components are assembled by zinc or aluminium metal sheets. A schematic view of the test sites with stormwater sewers and location of sampling points is shown in Figure A1.

The compositions and applied amounts of all relevant construction products (e.g., renders, façade paints, roof materials) were known. For example, the biocides used as film preservatives in renders and paints were diuron, octylisothiazolinone (OIT) (plasters site A), terbutryn (façade paints), isoproturon, and iodopropynyl butylcarbamate (IPBC) (façade paint site A). For site B, a mineral plaster was used without film preservatives but zinc oxide. The biocides in the coatings were provided as an encapsulated formulation.

2.1.2. Monitoring Strategy

For a period of about 18 months (September 2018–March 2020), stormwater emissions from selected façades and roofs of both sites were monitored for individual storm events. An additional sampling point was located at the catchment outlet at both sites (stormwater sewer), which allowed establishing a link between relevant emission sources (façades and roofs) and total site runoff emissions.

Façade runoff was captured at four points per site covering all four cardinal directions. Runoff was collected using 1 m long aluminium gutters connected to 10–20 L glass bottles. In order to minimize direct rainfall into the gutters, the opening was covered leaving a gap to the façades of 2 mm (Figure A2). The gutters were firmly attached to the façade with a flexible tape, allowing runoff to enter the gutters despite the roughness of the plaster. For each storm event, specific façade runoff was quantified by dividing the volume collected in the bottle by the façade area above the gutter.

At both sites, roof runoff was quantified by diverting the full flow of one downpipe into a stainless steel tipping counter (Umwelt-Geräte-Technik GmbH, Müncheberg, Germany) with tipping tray volumes of 2 L (site A) and 1 L (site B). Discharge was logged in 5 min intervals. A proportion of 0.5% of each tipping volume was diverted to a sampling container in order to collect a volume-proportional sample for each storm event (Figure A3). Calibration of the tipping and sampling mechanisms was conducted in field conditions prior to installation. Specific roof runoff for each event was computed by dividing the total logged volume by the connected roof area.

The stormwater runoff was sampled near each site's catchment outlet. Flow meters (ultrasound and cross-correlation sensors, PCM4 logger, Nivus, Eppingen, Germany) and portable automatic samplers (Sigma SD 900, Hach Lange, Düsseldorf, Germany) equipped with glass bottles (8 × 2 L) were installed in a manhole receiving runoff from the entire catchment (A and B, respectively) (Figure A4). The samplers were triggered by a preassigned water level threshold. Volume-proportional composite samples were prepared based on runoff measurements (for details see [11]) to determine the event mean concentrations (EMC).

All sample containers were emptied after each runoff event. Runoff volumes were recorded for a total of 70 runoff events for both sites. Of these, 20 events with rain depths between 5 and 45 mm were selected at each site for laboratory analyses of respective samples. These 20 events covered a total rain depth of 340 mm at site A (42% of total rainfall) and 315 mm at site B (43% of total rainfall). A list of all analysed events with their corresponding rain and wind data can be found in the Supporting Information Tables S1 and S2. Samples for analysis were retrieved and cooled within 24 h after the storm event. Given the predominance of wind directions from the north and the west, only façade runoff samples from these cardinal directions were analysed.

The samples were analysed by LC-MS/MS for a set of 12 biocides and transformation products and by ICP-MS for 4 heavy metals (zinc, copper, aluminium, vanadium) (see Table A1 for limits of quantification). The biocides were analysed directly without filtration with an injection volume of 50 µL. The selection of the substances was based on the known and suspected substances used in the construction products of both sites. Furthermore, a suspect target screening was conducted prior to the start of the monitoring campaign, according to Wode et al. [16], on grab samples taken at both sites during rain events. For this, the suspect database was expanded to include biocide transformation products described in the literature. As result of the suspect target screening, transformation products of diuron (diuron-desmethyl) and terbutryn (terbutryn-desethyl, terbutryn-2-hydroxy, and terbumeton) were detected, confirmed with reference standards, and integrated into the quantitative analysis method.

A tipping bucket rain gauge (Nivus RM 200/202, Nivus, Eppingen, Germany) was installed on the roof of a building in test site A and connected to a data logger (NivuLog Easy, Nivus, Eppingen, Germany) for remote data access. Rainfall was logged at 5 min intervals. Data for wind direction and speed were obtained from the German Weather

Service for stations closest to the test sites (Schönefeld for site A, Tegel for site B, both located about 7 km from the test sites).

2.2. Simulation of Runoff and Emissions

The modelling of emission events with COMLEAM (www.comleam.com, accessed on 16 January 2022) allowed the estimation of risk exposure in the water body on the basis of product-specific leaching from laboratory tests (immersion test for coatings with 9 leachates, dynamic surface leaching test (DSLT) for roof membranes and other solid products with 8 leachates) [EN 16105:2011-12, prEN 16637-2:2021]. For this purpose, logarithmic emission functions were parametrised using laboratory leaching data by means of non-linear regression [17]. A detailed description of the software and its mathematical model is available on the website (see above).

An emission scenario was defined using the building geometries, orientation, and materials of site A and B, as well as the surface runoff into the storm sewer. A wind-driven rain calculation was carried out according to ISO 15927-3:2009. Since the main wind direction in Central Europe is west, wind-driven rain mainly occurs on this side. Surface areas of façades relevant for runoff were determined for all buildings at both sites. For the façades, the runoff coefficient was set to 0.7, which includes water absorption, rebound, and evaporation. The dilution caused by runoff from sealed or partially sealed pathways and areas was taken into consideration and included in the model.

Weather data of the German Weather Service in hourly resolution for the Berlin site A (precipitation, wind direction, wind speed) were included to model the runoff. In addition, weather data over a period of three years, starting with the completion of construction of the investigated sites, was used to investigate the significance of specific factors, such as water body size or site-specific weather, on the potential emission using different scenarios.

The accuracy of the prediction of the modelled wind-driven rain was tested by comparing the simulations directly with the measured data for individual storm events. The measurement data used refer to event-related façade runoff quantities at the test sites.

Subsequently, loads were calculated taking into account the substance concentrations in runoff and in the sewer. The total potential emission includes all façades and represents the maximum substance emission in the area. From the difference between the total emission and the emission into the storm sewer, the material flows for direct discharge and retention and/or infiltration in the area could be estimated.

In the following presented results, the focus is placed on the biocide diuron from site A, which is long-lived in the environment. Furthermore, diuron used in façades coatings is a major substance present in urban runoff.

3. Results and Discussion

3.1. Field Study

The specific runoff volumes (L/m^2) from surfaces connected to the storm sewer (site, roofs, and façades) are shown in Figure 1 for all analysed events. The lower specific site runoff at site A reflects the presence of grass grid stones for parking lots with much lower runoff. The specific roof runoff at site B is lower compared with site A due to the water retention of the green roofs in site B. The higher specific façade runoff at site B is caused by a larger fraction of façade area facing west (main wind direction) compared with site A. However, façade runoff volumes are very low, comprising <1% of the site runoff measured in the storm sewer.

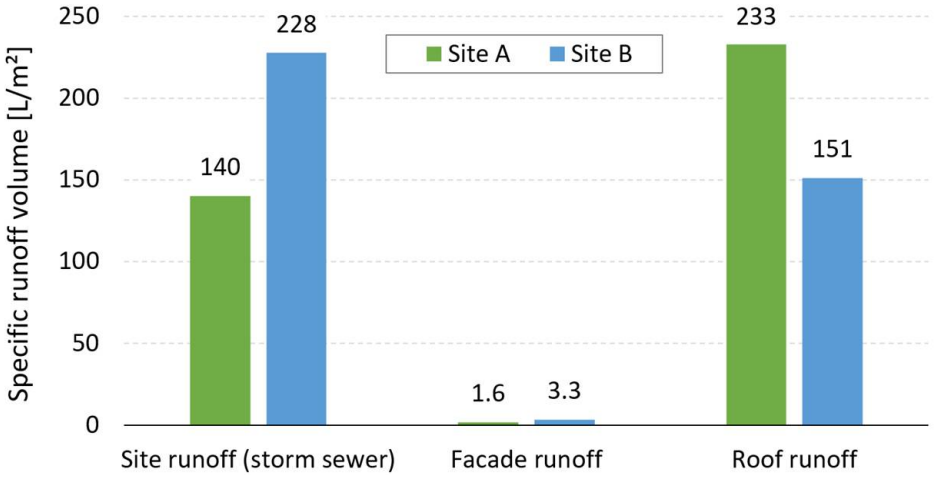

Figure 1. Specific runoff from surfaces drained to the storm sewer resulting from analysed storm events in test sites A and B.

The comparison of the specific façade runoff (L/m^2) for all storm events with stormwater volume data (n = 70) shows that the cumulated specific runoff from façades oriented to the west (site A: 8.1 L/m^2, site B: 12 L/m^2) is about twice as high compared with the runoff from façades facing north and south (see Figure A5). This is clearly associated with westerly wind directions during rain events (Figure A6). Higher specific runoff from the west façades at site B can mainly be attributed to a larger distance of the building to interfering structures (i.e., buildings, trees). The lowest cumulated specific runoff was measured at façades directed to the east (about 2 L/m^2 for both sites).

The concentrations in façade runoff are shown in Figure 2. For site A, only concentrations in the runoff from north façades are shown, as west façades were not painted and consequently did not contain biocides (Figure A7). However, the concentrations of diuron and OIT (film protection agents in plaster product) were similar in both runoff from the north and west façades (see Figure A7 for results of west façade). For site B, runoff from the west and north façades show the highest concentrations for diuron (mean: 900 µg/L), its transformation product diuron-desmethyl (DCPMU; mean 375 µg/L), and OIT (mean: 350 µg/L). No trend towards a decrease in concentration was evident in façade runoff during the time of field investigation.

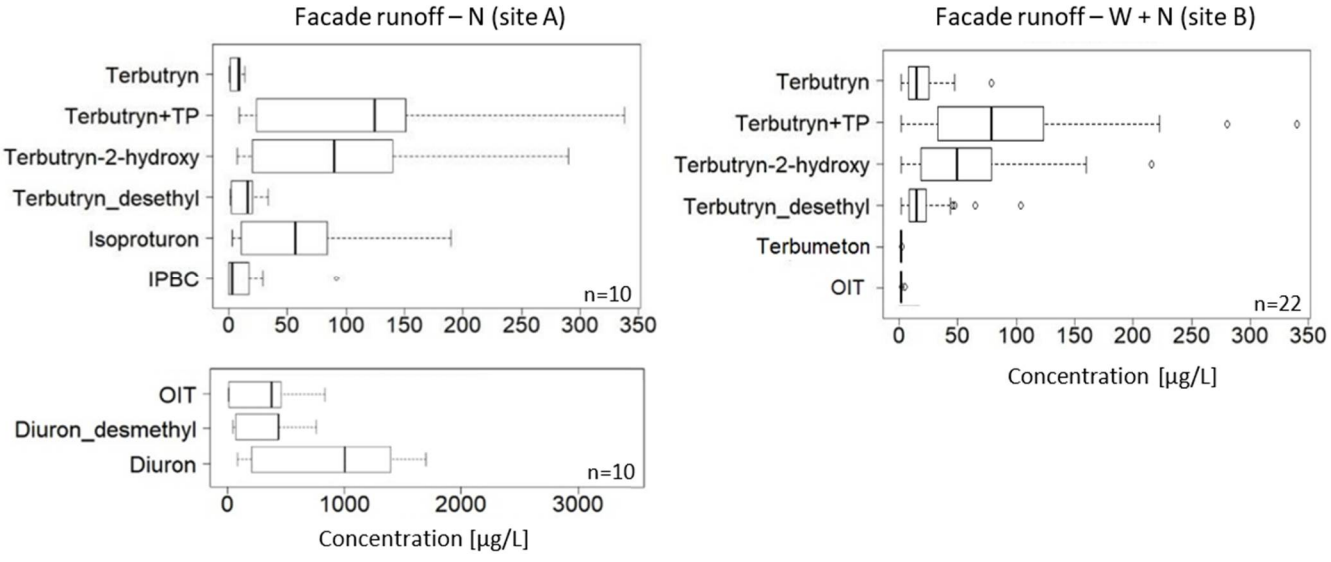

Figure 2. Concentration of biocides in façade runoff of painted façades at site A (north) and B (west and north).

Concentrations of diuron in façade runoff 1.5–3 years after application as measured in this study correspond well to concentrations of façade runoff from test façades after one year of exposure (1–3 mg/L) reported by Burkhardt et al. [8]. Diuron-desmethyl (DCPMU) has been detected in façade runoff of site A as a degradation product of diuron at concentration levels approximately one third of the diuron concentrations. Transformation of diuron to diuron-desmethyl has been described in previous studies [8,18].

Concentrations of terbutryn, isoproturon, and IPBC contained in the façade paint were about 1–2 orders of magnitude lower compared with diuron. Noticeable is the high concentration of the terbutryn transformation product terbutryn-2-hydroxy, exceeding the terbutryn concentrations by a factor of 3 (site B) and 10 (site A). Terbutryn-2-hydroxy is known to be a photodegradation product of terbutryn, induced by UV radiation. Terbutryn-desethyl (also known as M1) was detected at concentrations similar to terbutryn.

Results for façade runoff show that, although the runoff volumes were low compared with runoff from roofs and pervious surfaces, the concentrations of active ingredients were orders of magnitude above maximum allowable concentrations (MAC) of environmental quality standards applicable for surface waters (diuron—1.2 µg/L; terbutryn—0.34 µg/L; isoproturon—1.0 µg/L).

Roof runoff at site A was clearly dominated by the root protection agent mecoprop, resulting in a mean concentration of 20 µg/L (Figure 3). This is remarkable, as site A has no green roof and therefore does not require a protection against root penetration. Furthermore, recommendations by the Berlin authorities for the application of bituminous roofing membranes suggest the application of products with root protection only when necessary (e.g., for green roofs) [19]. The concentrations measured in the roof runoff of site A are in the same range as those reported by Bucheli et al. [5] in the roof runoff of two flat roofs with bituminous roofing membranes (average 3.3 µg/L, max. 35 µg/L). The concentrations of the root protection agent MCPA were low and close to the limit of detection, as it is not applied in the existing membrane. Low concentrations of diuron, terbutryn, and their transformation products were also detected (<0.5 µg/L, Figure 3, left), but at much lower concentrations than in façade runoff. Small, coated surfaces in the roof area could be potential sources.

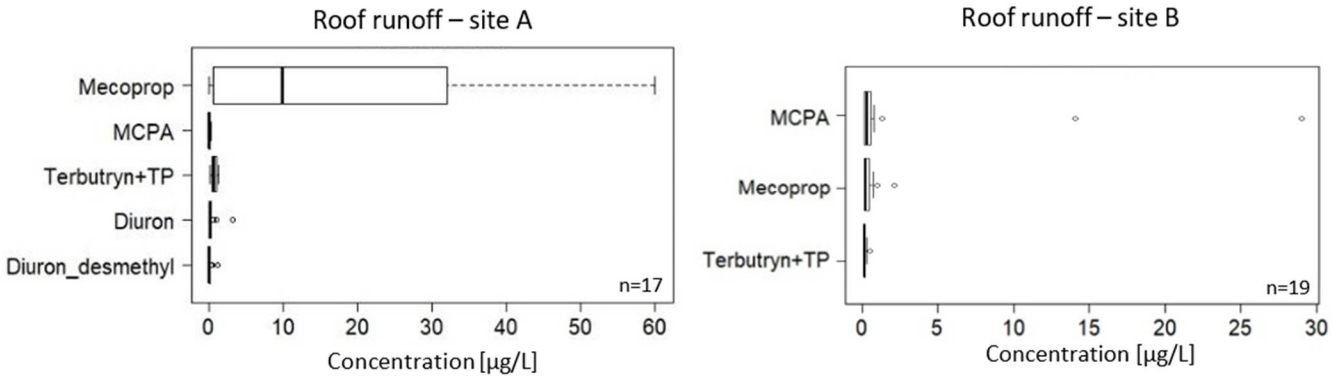

Figure 3. Concentration of organic trace pollutants in roof runoff at site A and B.

MCPA present in bituminous sheets at site B was detected only in two rain events at concentrations >5 µg/L (15 and 30 µg/L, Figure 3, right). Similar to site A, low concentrations of terbutryn and its transformation products (<0.3 µg/L) are likely due to small painted surfaces in the roof area.

Concentrations of organic micropollutants in stormwater runoff of the entire site (storm sewer) are shown in Figure 4. At site A, the highest concentrations were measured for mecoprop with a maximum concentration of 62 µg/L. Mecoprop concentrations were in the same order of magnitude as in the roof runoff (Figure 3), as a large proportion (67%) of the site runoff is contributed by the roof drainage (Figure 1). In contrast, the MCPA

measured in the storm sewer cannot be explained by emissions from the sampled roof. Therefore, much higher emission of MCPA from the other roofs of site A are expected.

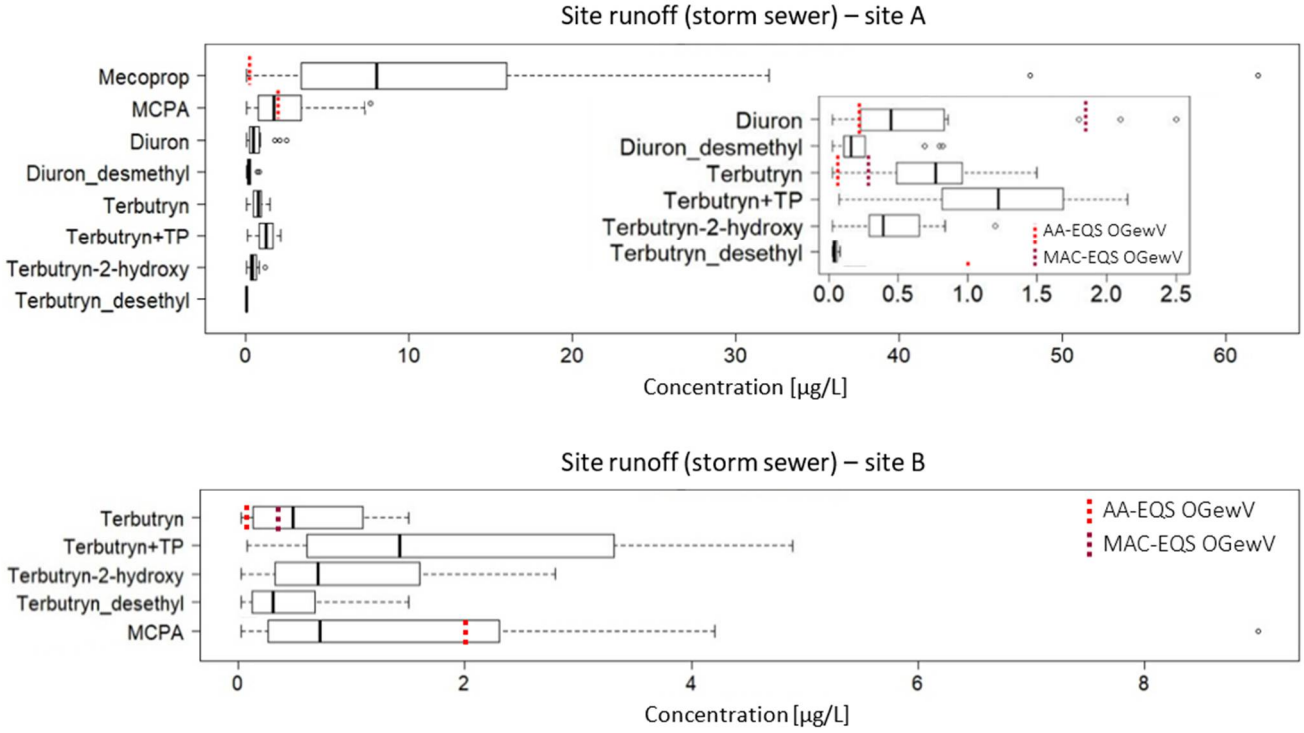

Figure 4. Concentration of micropollutants in site runoff (storm sewer) of site A and B.

Concentrations of diuron and terbutryn varied between 0.2 and 2.5 µg/L. These are higher compared with stormwater runoff from larger urban catchments of different land uses [11]. This is plausible as stormwater runoff in our study originates exclusively from sites with new buildings constructed about 1.5 years before monitoring began; whereas, larger urban catchments comprise of buildings of different age containing different construction materials. On the other hand, concentrations of terbutryn in stormwater runoff are well in line with concentrations of Bollmann et al. [13], measured in the stormwater runoff of a residential catchment in Denmark (mean: 0.1 µg/L, max 1 µg/L). In contrast to diuron and terbutryn, OIT, IPBC, and isoproturon, occurring in the façade runoff (Figure 2), were hardly detected in the storm sewer samples. In the case of OIT and IPBC, this has also been described by Burkhardt et al. [20], who concluded that OIT and IPBC released from façades do not lead to surface water pollution due to the rapid transformation of both biocides (half-life DT_{50} < 3 days).

Although concentrations of biocides in storm sewer samples are several orders of magnitudes lower than at the source (façade runoff), the results of this study show that environmentally relevant concentrations can be reached in stormwater runoff of entire residential sites despite very low façade runoff volumes. A comparison with environmental quality standards (EQS) for surface waters applicable in Germany (OGewV 2016) shows that both the annual average (AA) and the maximum allowable concentrations (MAC) for terbutryn, diuron, mecoprop, and MCPA were exceeded in the stormwater runoff of the investigated sites (Figure 4). It should be noted, however, that the EQSs apply to surface water and not to stormwater runoff. The dilution of the stormwater runoff in surfaces water or removal in soil during infiltration is pivotal in determining whether the emissions exceed the water quality standards and impact aquatic organisms. Nevertheless, observed concentrations in runoff of the test sites indicate that during wet weather the water quality of small urban streams may exceed the EQSs.

In addition, groundwater could be polluted if runoff is infiltrated, and the compounds are not retained in the unsaturated soil. In fact, the presence of diuron and terbutryn in urban groundwater has been demonstrated in a residential area with infiltration swales in Freiburg (Germany) and was related to emissions from façades [18].

The concentrations of heavy metals are shown in Figure 5. For both sites, the highest concentrations were found for zinc and aluminium. Surprisingly high zinc concentrations occurred in the façade runoff, with mean values of 610 µg/L (site A) and 460 µg/L (site B) and a maximum value of 3700 µg/L. Therefore, a release of zinc from plaster and paint is likely caused by the use of zinc oxide. In roof runoff, zinc can be emitted by gutters, galvanized smoke ventilation pipes, and roof edge cover sheets made of zinc. Higher roof runoff concentrations for zinc at site B (mean 500 µg/L) in comparison with site A (mean 180 µg/L) are attributed to the different materials used for the roof edge coverings (site B—zinc; site A—aluminium), highlighting the relevance of roof materials for zinc loads in stormwater runoff. For site runoff (storm sewer samples), zinc inputs resulted in mean zinc concentrations of 240 µg/L for site B (max: 640 µg/L) and 135 µg/L (max: 370 µg/L) for site A. These are comparable to the zinc concentrations in stormwater runoff of residential areas in Berlin (150 and 260 µg/L in runoff from catchments comprising of concrete slab buildings and one family homes [11]) and in France (210 µg/L [12]).

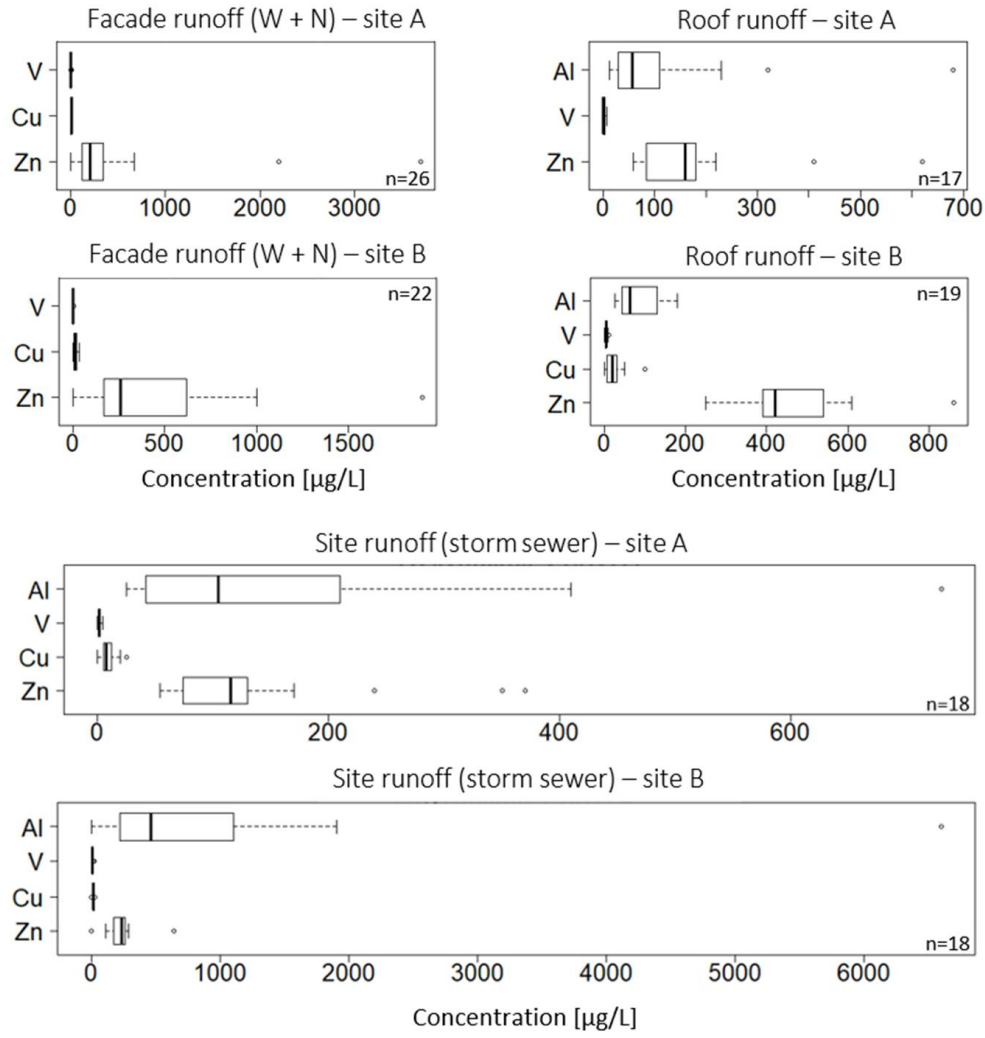

Figure 5. Concentrations of heavy metals in runoff of façades and roofs and in site runoff of site A and site B.

The dissolved fraction of zinc in this study is in average 60% and of aluminium 15%. Thus, zinc concentrations in the site runoff clearly exceeded the EQS proposal for dissolved zinc of 33 µg/L [21]. Load estimations indicate that zinc in roof runoff contributes >90% (site A) and 50% (site B) to the total zinc mass in the stormwater sewer. At site B, in addition to roof inputs, zinc sheet windowsills and possible traffic-related zinc inputs from a small residential street are additional plausible sources. At site B, high concentrations of aluminium were detected with a mean concentration in site runoff of 960 µg/L (max: 6600 µg/L). The sources for aluminium could not be fully clarified. However, in contrast with zinc, the ecotoxicological relevance of this metal is significantly lower.

3.2. Modelling

First, façade runoff was modelled considering the local weather data for the monitoring time period and the same façade area and orientation as the sampled façades in the field study. The predicted runoff from all building façades shows high agreement with the measured pattern of events, both in terms of variability and runoff amounts (Figure A8). For site A, the simulated runoff, cumulative over 1.5 years, is 12.5 L/m^2 compared with the measured runoff of 8.4 L/m^2. The overestimation in the range of 30% is mainly associated with 3 heavy rain events of >30 mm each. If these data points were not considered, the deviations would amount to <10%. During these rain events, the effective wind field was different from that at the weather station about 1 km away. Shading by trees, bushes, or neighbouring buildings and local turbulences could have caused further deviations, which can only be roughly described with hourly resolved measurement data.

The simulated runoff volume in the storm sewer, draining the associated surfaces of the entire catchment area, corresponds to about 120% of the measured volume and originates for 30% from open spaces and paths and >60% from roofs (Figure 6). The percentage of area connected to the storm sewer was estimated in the field study and adopted in the model accordingly. Façade runoff is negligible in terms of volume, accounting for 0.02% of the total runoff, with the east façades (2200 m^2) contributing 65%, followed by the south (1850 m^2) and west (1250 m^2) façades, each contributing 8%. The overestimation of the runoff volumes in the stormwater sewer is based firstly on two large individual rain events, and secondly on the generic runoff coefficients defined in the model for all surfaces, which are likely to deviate from reality (Figure 6).

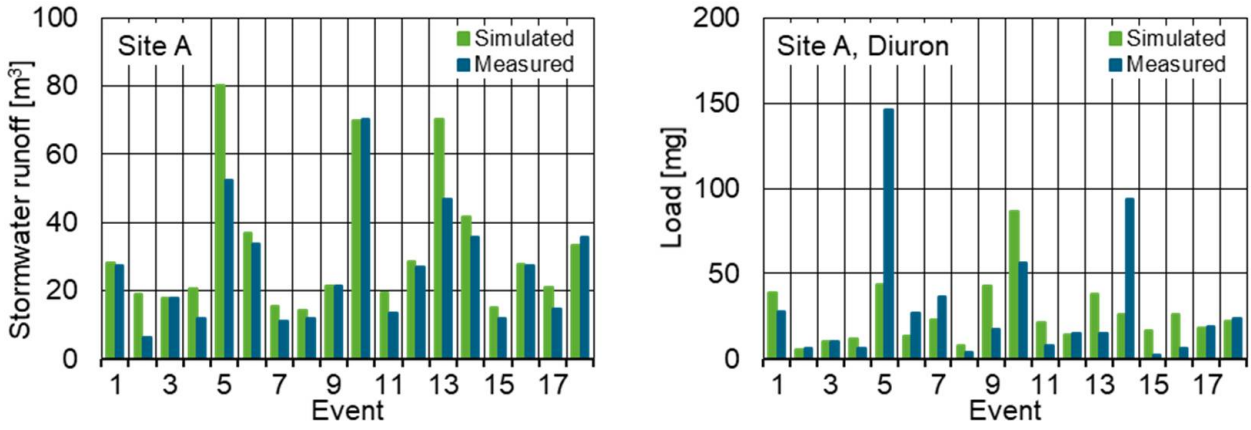

Figure 6. Measured and simulated stormwater runoff (**left**) and diuron load (**right**) in the stormwater of the entire catchment of site A for 18 analysed events.

For the modelling of the substance emission, lab data (prEN 16637-2:2021) of the same render used at site A was used to parametrize the emission function. The modelled emission underlines the strong relevance of the façade orientation. In fact, 75% of the total emitted diuron load of the catchment originates from the west and south façades (Figure 7, right). The modelled diuron load of all façade surfaces draining into the stormwater channel is only about 9% below the load estimated from measured data (476 vs. 522 mg).

The difference can again be attributed to two rain events with measured values being about five times higher than the simulated concentrations (Figure 6). However, diuron entered the storm sewer primarily from the east façades, as these have the highest percentage of area being in connection with the storm sewer through sealed and drained surfaces (Figure 7).

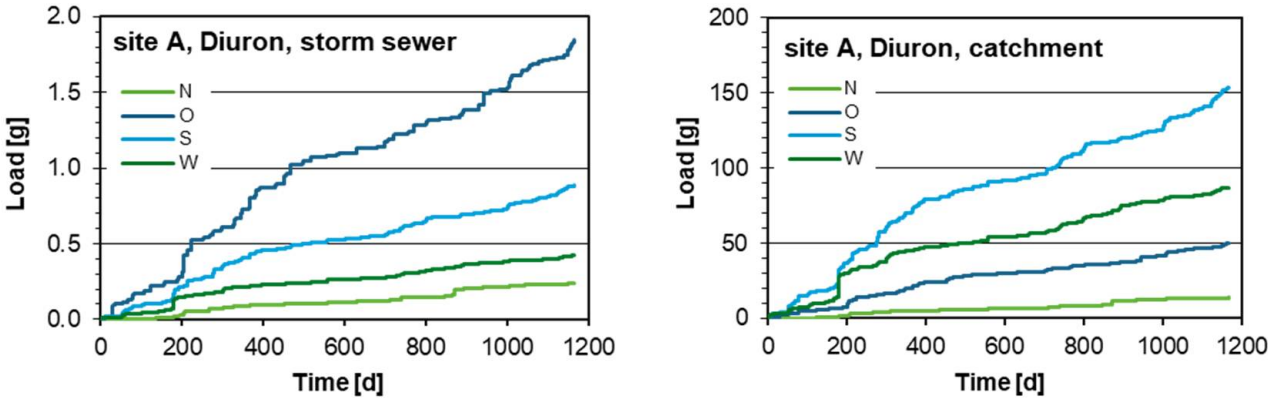

Figure 7. Cumulative exposure-dependent diuron load of all building façades in connection with the storm sewer (**left**) and of all façades of site A (**right**). The scenario-based simulations cover 3 years starting with the construction of the site.

The total catchment emissions of diuron were calculated for a time period of three years by cumulating emissions of single events for all façades, regardless of their connection to the storm sewer. This model-based, exposure-dependent emission estimation at site A shows that the modelled diuron load (3.4 g) in the storm sewer is about 100 times smaller than the total emission from all façades of the site (305 g) (Figure 7). Thus, the difference between the emission into the sewer and the total emission indicates that only 1% of the amount of biocide washed out enters the stormwater sewer, which discharges into River Spree, while 99% of the load remains on site. The 305 g diuron emission corresponds to 6950 m^2 of façades, resulting in an average release of 43.9 mg/m^2 in 3 years with decreasing emission rate over time.

To estimate the possible impact of the diuron emission on a receiving surface water, another scenario was calculated, assuming that 100% of the façade runoff is directly discharged into a surface water with a flow of 50 L/s (worst case scenario). Results show that the total diuron emissions are significant as these result in concentrations that exceed the EQS for surface waters (Figure 8). Based on the presented scenario, it is clear that peak concentrations occur during most wet weather flows at least within the first year after construction. Especially in small water bodies (50 L/s), the duration and number of possible exceedances increase disproportionately than in larger ones (500 L/s).

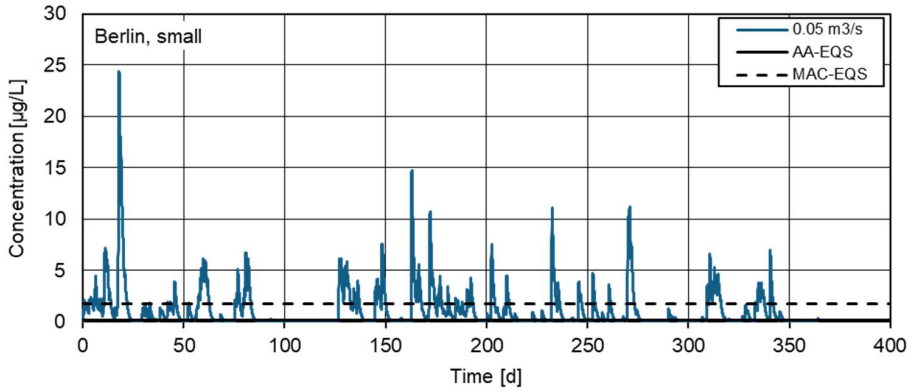

Figure 8. Simulated concentration of diuron in a small brook (0.05 m^3/s) in Berlin over a period of 1 year.

4. Conclusions

Results of field investigations have shown that emissions of several substances from building materials may enter stormwater runoff resulting in concentrations above the thresholds for surface water quality. In particular, leaching of the biocides diuron and terbutryn, used as film preservatives in façade coatings, the root protection agents MCPA and mecoprop, and zinc resulted in excessive concentrations. The concentrations of the analysed transformation products for terbutryn were, in some cases (especially for terbutryn-2-hydroxy), significantly higher than those of terbutryn itself; however, their ecotoxicity is lower (e.g., AA-EQS for desethylterbutryn is more than three times higher than that of terbutryn [22]). It should be considered that the peak concentration occurring during a storm event lasting several hours is higher than the respective event mean concentration presented here, as this represents an average concentration over several hours. An EQS exceedance after discharge into large water bodies is unlikely due to dilution effects. However, there is a risk for small water bodies if stormwater is not sufficiently treated before discharge or not sufficiently diluted in the receiving surface waters. If stormwater runoff is routed through infiltration ponds or ditches, the groundwater might be affected if the substances are not adsorbed or degraded in the unsaturated soil.

The modelling of two sites in Berlin using COMLEAM illustrated the most relevant emissions and transmission factors, such as the influence of site-specific precipitation patterns and receiving surface water dimensions. In addition, a mass balance was determined over a period of several years. Results showed that >90% of façade emissions can remain on site and diffusely percolate around buildings. These diffuse inputs may also contribute to potential soil and groundwater contamination.

The key to preventing or mitigating emissions is to limit or avoid material pollution at the source. As substances that are added to building materials serve a function (i.e., application of biocides in façade paints to prevent algae growth), this prevention or mitigation is challenging. In addition, methodologies are still being developed to better assess the impact of environmentally relevant substances applied in building materials. However, a variety of mitigation measures are capable of reducing the impact of substances in building materials already known to exert adverse effects on the environment. For example, it is recommended to construct façades with roof overhang to keep them dry, to use building materials without environmentally problematic substances, to use certified reduced leaching products, and to carry out proper maintenance of the building envelope. In addition, the reduction in runoff through the greening of roofs and façades and through retention measures further contributes to the reduction in emitted loads. Finally, technical end-of-pipe solutions are available for removing organic micropollutants from stormwater using filters with specialized substrates. The German environmental agency published guidelines to support planners and architects in regard to measures mitigating the pollution of stormwater from building products [23]. To prevent negative effects for surface waters and/or groundwater due to the leaching of substances from building materials, mitigation measures need to be incorporated already in the planning stage of construction projects. Considering micropollutants in urban planning processes is crucial in the journey towards water-sensitive cities.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w14030303/s1, Table S1: Rain and wind data of all analysed samples of site A; Table S2: Rain and wind data of all analysed samples of site B.

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Data Availability Statement: The data set of concentrations measured at test sites A and B (façade runoff, roof runoff, and stormwater runoff in storm sewer) is available as a repository under https://doi.org/10.5281/zenodo.5792938.

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Conflicts of Interest: The authors declare no conflict of interest.

Appendix A

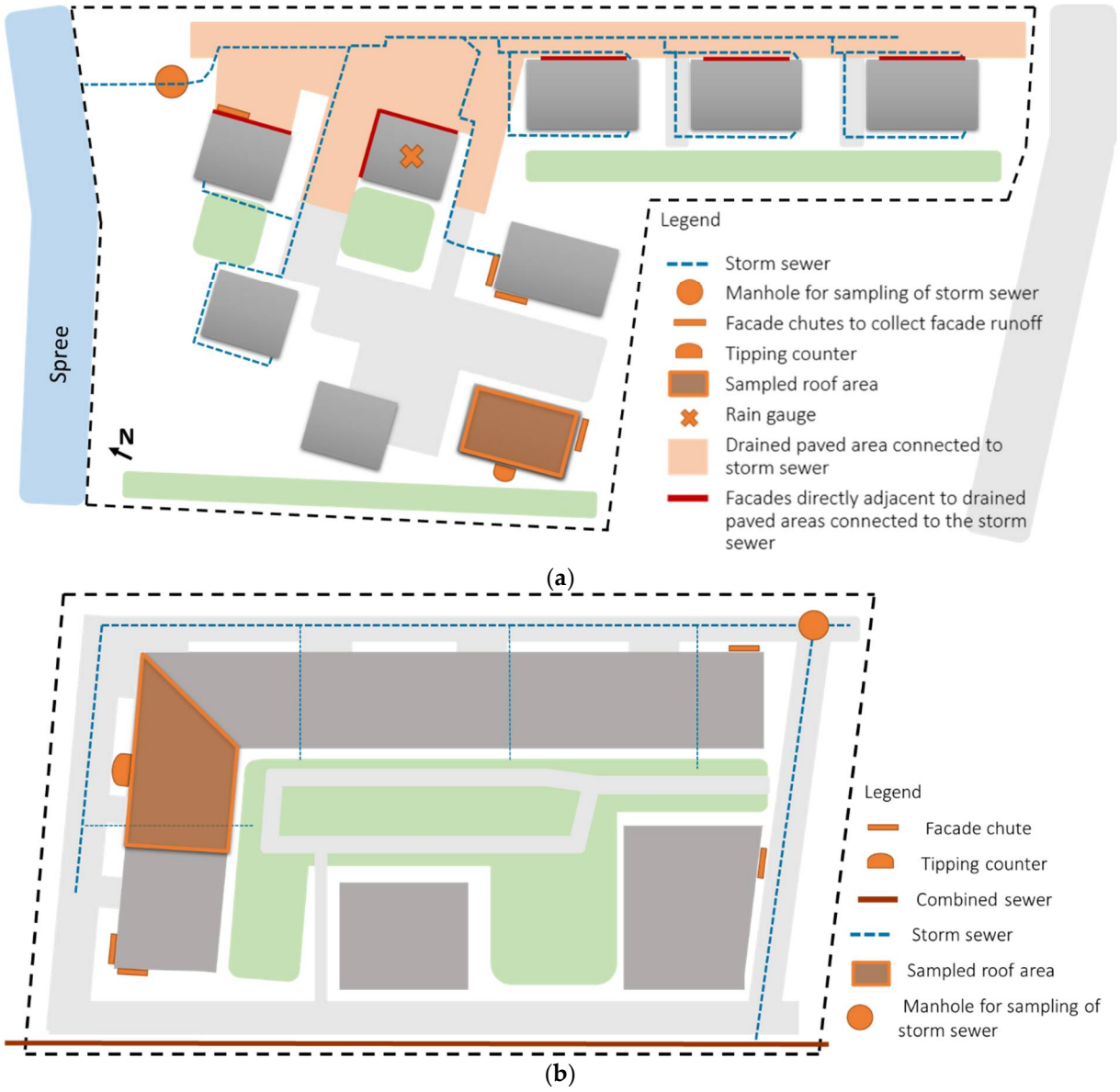

Figure A1. Schematic view of test site A (**a**) and test site B (**b**).

Figure A2. Installation of a gutter to capture façade runoff.

Figure A3. Tipping counter for measurement and sampling of roof runoff.

Figure A4. Installation of an automatic sampler and flow measuring device in a storm sewer and a manhole.

Table A1. Limits of quantification (LOQ) of analysed compounds; TP—transformation product.

Compound	Limit of Quantification [µg/L]
Diuron	0.025
Diuron-desmethyl (DCPMU; TP)	0.025
Isoproturon	0.025
Iodocarb (IPBC)	0.05
MCPA	0.025
Mecoprop	0.025
Octhilinone (OIT)	0.1
Terbutryn	0.025
Terbumeton (TP)	0.025
Terbutryn-desethyl (TP)	0.025
Terbutryn-2-hydroxy (TP)	0.025
Aluminium (Al)	5.0
Copper (Cu)	5.0
Vanadium (V)	2.0
Zinc (Zn)	5.0

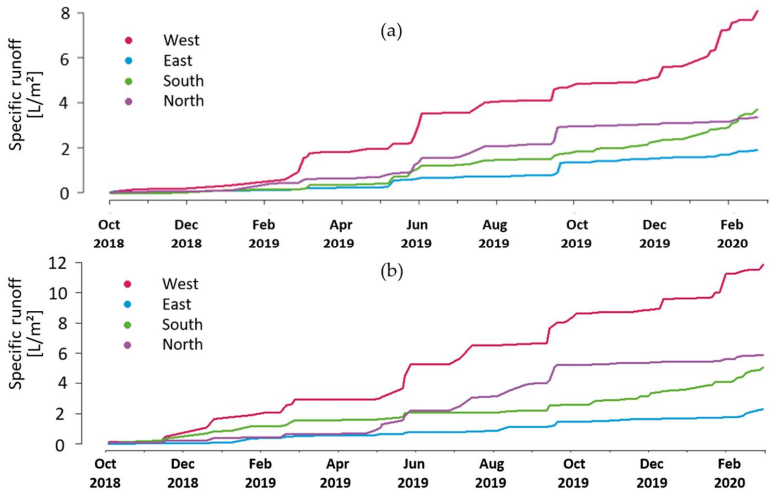

Figure A5. Cumulated specific façade runoff for site A (**a**) and site B (**b**).

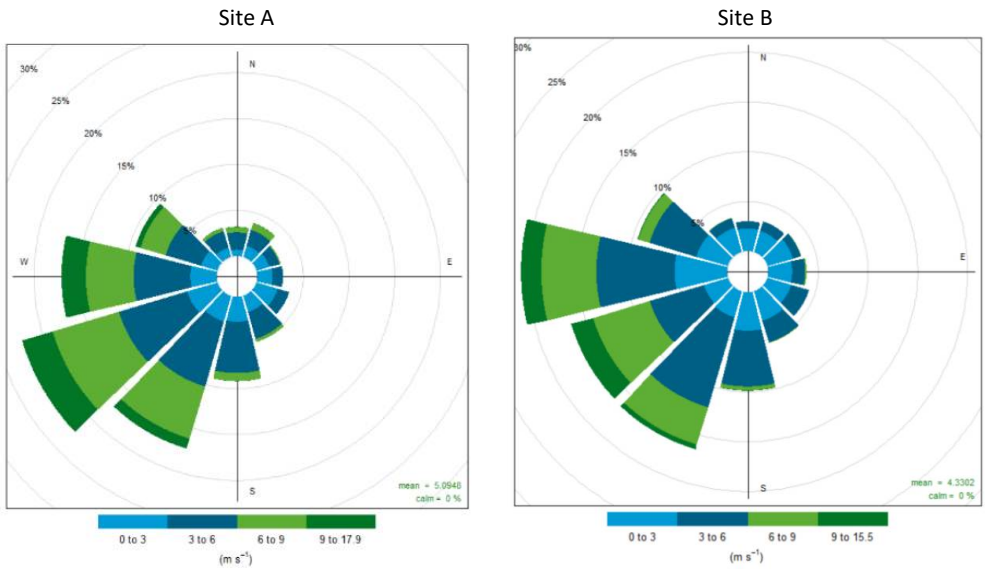

Figure A6. Wind speed and wind directions for all sampled events at test sites A and B.

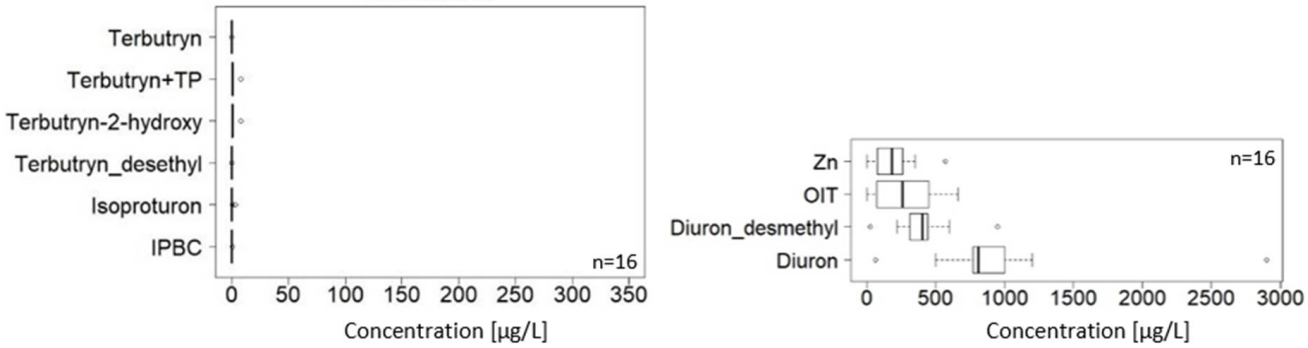

Figure A7. Concentrations of biocides in façade runoff from west façades at test site A.

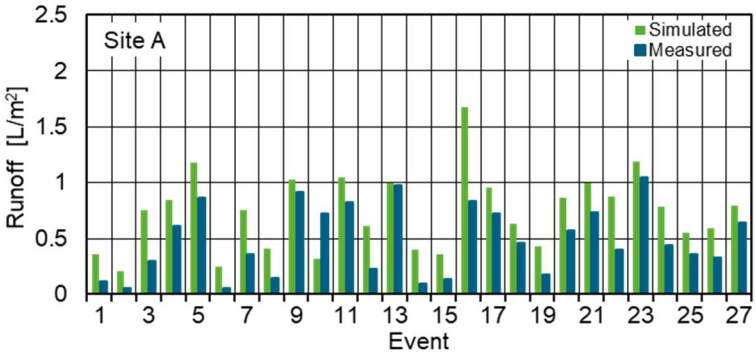

Figure A8. Measured and simulated façade runoff of buildings at test site A and site B for 27 and 26 events, respectively. Events with more than 10 mm rainfall were taken into account as a basis for the prediction.

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