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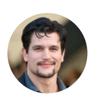

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Leaching of Biocides from Façades under Natural Weather Conditions

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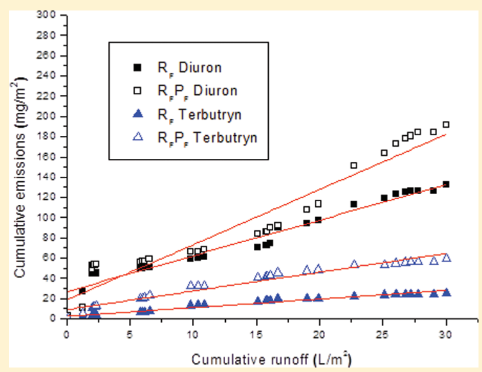
Supporting Information

ABSTRACT: Biocides are included in organic building façade coatings as protection against biological attack by algae and fungi but have the potential to enter the environment via leaching into runoff from wind driven rain. The following field study correlates wind driven rain to runoff and measured the release of several commonly used organic biocides (terbutryn, Irgarol 1051, diuron, isoproturon, OIT, DCOIT) in organic façade coatings from four coating systems. During one year of exposure of a west oriented model house façade in the Zurich, Switzerland area, an average of 62.7 L/m^2, or 6.3% of annual precipitation came off the four façade panels installed as runoff. The ISO method for calculating wind driven rain loads is adapted to predict runoff and can be used in the calculation of emissions in the field. Biocide concentrations tend to be higher in the early lifetime of the coatings and then reach fairly consistent levels later, generally ranging on the order of mg/L or hundreds of μg/L. On the basis of the amount remaining in the film after exposure, the occurrence of transformation products, and the calculated amounts in the leachate, degradation plays a significant role in the overall mass balance.



■ INTRODUCTION

At real buildings, façade coatings become moist due to wind driven rain and dew depending on characteristics of the site (latitude, altitude), architecture (height of façade, roof overhang, position on the façade), exposure (orientation), and weather conditions (wind speed, wind direction, precipitation, temperature). External thermal insulation composite systems (ETICS) are a mature building cladding technology that promote high energy savings but typically are offered with coatings containing biocides. ETICS are layered systems typically consisting of an insulation panel, upon which a ca. 5 mm mineral render, or mortar, is laid with an embedded fiberglass mesh for mechanical support. Upon this a finishing layer of a hydrophobic textured render with a polymeric binding phase (e.g., 2 mm thickness due to 2 mm grain size of certain fillers) is placed, and sometimes two paint layers of a few hundred micrometers are added. Organic biocides are an integral part of the organic renders and paints of ETICS by providing protection from growth of algae and fungi. They are typically added to the renders and paints in the wet state and are meant to slowly migrate to the surface of the dry coating during wetting events.[1] However, they also have the potential to enter the environment, at levels where they sometimes exceed water quality standards or create ecotoxicological concern.[2,3] Studies have demonstrated that some of the biocides used in façade coatings still exist in surface waters or urban stormwater sewers despite a ban on their agricultural or public use indicating that another source besides agriculture is relevant.[4−6]

Regulators ask for knowledge of the fundamental release mechanism to provide more accurate source inputs for risk assessments. Furthermore, technical stormwater treatment systems might be designed for release conditions expected in urban catchments.[7] Producers can benefit with smarter direction in product design for reducing emissions while maintaining effectiveness and service life.

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What is most important, however, is the emissions behavior in the field under real weather conditions. In one of the few published field studies of organic biocides leaching from coatings, residual amounts of diuron in a paint exposed to laboratory irrigations and to field conditions are measured finding diffusion controlled release in the lab and relating it to the field results.[8] Recently, Burkhardt et al. measured biocides in runoff and urban stormwater subcatchments for a newly coated façade and an older façade and reported biocide concentrations at levels consistent with high release behavior early in a façade's lifetime and early in a rain event.[2] Additionally, release behavior of the inorganic biocide nano-silver used in façade paint was demonstrated under field conditions.[9] Whereas studies have been performed characterizing levels of organic biocide in surface waters and in lab irrigation studies, knowledge of how wind driven rain leads to façade runoff is important in characterizing transport of pollutants from building façades to surface waters, and is introduced in the following subsection.

Wind Driven Rain and Façade Runoff. Wind driven rain (WDR), rainfall given a horizontal velocity component by the wind, is the most important moisture load on building façades. The topic as it relates to building façades was recently extensively reviewed by Blocken and Carmeliet,[10] and can be explained by parts a and b of Figure 1. Rain falls through a horizontal plane at a particular intensity R_h and at its terminal velocity V_t. The wind gives the rain a horizontal velocity component that can push it against a vertical plane such as a building façade. Whereas horizontal rainfall intensity and wind speed are easy to measure, the rainfall terminal velocity is not and it is instead captured in a coefficient α, which also captures other parameters. In general, the relationship used to calculate the WDR intensity, R_{WDR} (in L/m^2·h), on a façade is given as

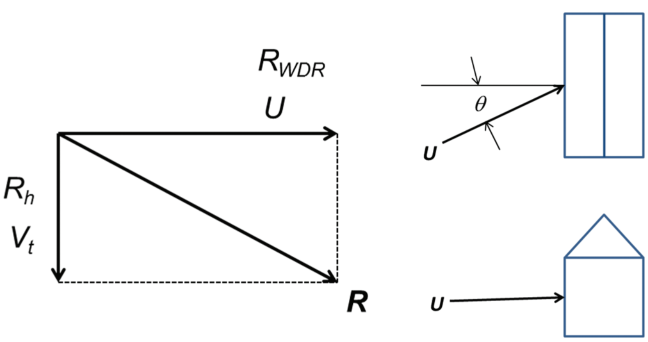

Figure 1. a-b. Figure 1a (left) gives the vector relationship of wind and rain, where R_{WDR}, the wind driven rain intensity in L/m^2·h, can be calculated if the wind speed U, horizontal rainfall intensity R_h, and terminal velocity V_t are known. Part b (right) shows how the angle of the wind direction to the building façade affects the driving rain index.

$$R_{WDR} = \alpha R_h^{0.88} U \cos\theta \tag{1}$$

where R_h is the horizontal rainfall intensity in L/m^2·h, U is the wind speed in m/s, θ is the angle formed by the wind direction and the normal to the façade, and the coefficient α is known as the wind driven rain coefficient, with the units s/m. The wind speed U varies with height and can lead to some confusion and, for the purposes of this article is taken as the wind speed measured at 10 m, which is typical for meteorological measurements. The quantity

$$R_h U \cos\theta \tag{2}$$

is called the driving rain index (DRI), and here has the exponent 0.88 removed, as is common in driving rain research.

It can be seen by inspecting part b of Figure 1 and (2) that the DRI <0 if cos θ is negative, that is if the wind direction is unfavorable, then there is no rain being driven against the façade. The driving rain coefficient α has been shown to be approximately $^2/_9$ in free field conditions, but changes when a building is placed in the wind flow field and raindrop trajectories are altered. The driving rain coefficient is calculated to account for the variation in wind speed and flow pattern due to topography, building size, and building shape, and it varies across the building façade, and with rainfall intensity. In particular, the wind blocking effect, where raindrop trajectories are altered due to the fact that wind flows around and over, but not under, a building, leads to a wetting pattern in which rain is directed toward the tops and corners of a building.[10] There are a number of methods to calculate this coefficient, two of which are the semiempirical ISO Standard method [11] and the experimentally validated, computational method of Choi,[12] which was extended by Blocken and Carmeliet.[13] Although accurate, the computational method is too cumbersome for routine analysis. Blocken and Carmeliet recently compared the two methods[14] and, while there are some deficiencies in the ISO method, it is considered an acceptable method for estimation of wind driven rain loads, and it is the method used in this study.

The ISO method takes the driving rain coefficient α and turns it into the following:

$$\alpha = \frac{2}{9} C_R C_T O W \tag{3}$$

where C_R is the roughness coefficient, C_T is the topography coefficient, and O and W are the obstruction and wall factors, respectively. The roughness coefficient accounts for change in mean wind speed due to upstream terrain roughness and height above the ground, and the topography coefficient accounts for mean wind speed increase due to hills or escarpments. The obstruction factor accounts for an obstacle in the immediate vicinity, and the wall factor accounts for the change in the wind flow pattern due to the flow around the building. Each coefficient except for C_T is less than or equal to one, so in general, environmental factors reduce the wind driven rain intensity. To give typical values, for a small (<4 m in height) building, C_R, the roughness coefficient can vary from 0.95 (wide, unobstructed terrain such as open water or fields) to 0.47 (dense urban area with tall buildings). The topography coefficient, C_T, can vary from 1 to 1.6, while the obstruction factor O can vary from 0.2 (obstruction 4−8 m in front of wall) to 1.0 (obstruction over 120 m away). The wall factor W can vary from 0.2 for tall buildings to 0.4 for smaller buildings but only for particular building types and sizes as defined by the standard.

Calculation of actual runoff is a more difficult task, as there are a number of interactions to consider when the rainfall reaches the façade, such as splashing, bouncing, absorption, adhesion, and evaporation. Carmeliet and Blocken[15,16] and Abuku et al. [17] performed in-depth studies of these phenomena for different materials, and noted the difficulties in this calculation. At best, it is possible to calculate the maximum possible runoff using WDR models because each of the noted phenomena theoretically leads to a decrease in runoff.

To understand and begin steps toward predicting field behavior in leaching of biocides from façades, it is necessary to clarify the hydrological conditions that lead to release and combine them with an understanding of the release mechanism. In this study, the runoff and leaching of typical organic biocides are measured in a model building exposed for one year.

■ MATERIALS & METHODS

Model House. A model house was constructed near Zurich, Switzerland (8°36′45″E, 47°24′10″N), with coated test expanded polystyrene (EPS) panels 1.75 m high × 0.75 m wide × 0.16 m thick placed in the west facing, 3.5 m long façade. The model house was 2.5 m in depth and had a 1 m high, symmetric sloping roof. As seen in Figure 2, there is no roof overhang and impact of runoff from the sloped roof was controlled by a rain gutter at top position of the coated panels. The orientation and construction of the façade was intended to sample the worst case scenario for driving rain in the Zurich area, where the prevailing wind direction is west to northwest with an average wind speed (measured at 10 m height) of about 2.3 m/s, and average precipitation is approximately 1100 mm per year. Runoff and weather data were monitored over the course of 369 days, from mid January 2008 to mid January 2009. There was a standard 10 m weather station positioned within 15 m of the model house taking data at 10 min intervals. The surroundings of the model house were typical of a flat, suburban zone of the greater Zurich metropolitan area, with fairly evenly spaced buildings no taller than 10−15 m in height. The immediate area of the model house had no obstructions directly in front of the measured façade for 60 m. Throughout the course of the study, the event specific runoff was captured from each individual panel in 7 L glass bottles using alloy gutters with a drain pipe mounted at the bottom of every panel. The bottles were located under the building, connected to the gutters via plastic tubing, and protected from the sun, so evaporation errors and UV degradation of compounds in the leachate are minimized. The water volume was gravimetrically measured about one day after the runoff event occurred. Runoff from panel 2 was continuously monitored by an electronic balance. The first sample accounted for was taken at January 18. The distance between the gutter and the coating was set to less than 3 mm, thus it is unlikely that precipitation influenced the sampled volume directly. Further details are described by Kägi et al.[9]

Figure 2. Model house installed in Dübendorf, Switzerland facing due west and without roof overhang. Panels (1.5 m tall × 0.75 m wide) installed are 1 − R_FP_{Dx}, 2 − R_F, 3 − R_FP_F, 4 − R_BP_B.

Coatings and Biocides. The four EPS panels were coated, in accordance with standard construction practice, by a mineral sub render with an embedded fiberglass supporting mesh followed by an organic top render. Three panels had two additional layers of paint added. The four panels were coated with 1) reference render formulation + market product paint without biocides (R_FP_{Dx}), 2) reference render (R_F), 3) reference render + reference paint (R_FP_F), 4) market formulation render + market formulation paint (R_BP_B). The marked numbers correspond to those in Figure 2. The reference render and paint formulations have been previously published,[18] and as thin hydrophobic layers, do not have large uptake relative to runoff amounts. The reference render and paint formulations are based on a styrene−acrylate and silicone binder respectively and are typical of products used in Switzerland and Germany.[1] Market formulation renders and paints were provided by industrial partners.

Biocides used in this study include diuron (3-(3,4-dichlorophenyl)-1,1-dimethylurea), isoproturon (3-(4-isopropylphenyl)-1,1-dimethylurea), terbutryn (*N*2-tert-butyl-*N*4-ethyl-6-methylthio-1,3,5-triazine-2,4-diamine), Irgarol 1051 (*N*2-tert-butyl-*N*4-cyclopropyl-6-methylsulfanyl-1,3,5-triazine-2,4-diamine), IPBC (3-iodoprop-2-ynyl *N*-butylcarbamate), OIT (2-Octyl-3(2H)-isothiazolinone), and DCOIT (4,5-dichloro-2-*n*-octyl-4-isothiazolino-3-one), which are biocides that have been used in recent years or are currently being used in façade applications.[1] The biocides were added, similar to previous lab studies[18,19] and typical of industry practice, in the final step of mixing as formulated market products (i.e., not as pure active ingredients) at uniform proportions of 750 ppm a.i. for renders and 1500 ppm a.i. for paints in the wet state. Depending on the product and biocide compound, the concentrations in practice are in the range of 200 to 2000 ppm, however, paints often typically contain twice the biocide concentrations of the render. With corresponding application amounts of each biocide of approximately 2.5 g/m^2 for renders and 0.5 g/m^2 for paints, this again corresponds to biocide area concentrations of approximately 2250 mg/m^2 for renders and 750 mg/m^2 for paint, which gives approximately 2250 mg/m^2 and 3000 mg/m^2 for render and render/paint systems, respectively. The drying time under room temperature in the laboratory was 14 days for the mineral render, 5 days for the top-render, and 1 day within the double application of the topcoat. The applied mass, as well as the loss in weight by drying, was determined after application with a balance. The final dry weights varied between 2.7 kg/m^2 for the top-renders and 0.25 kg/m^2 for the paints in average.

After rain events, samples of the runoff were taken and analyzed using liquid chromatography followed by tandem mass spectroscopy (LC-MS/MS), similar to lab studies [19] and published by Bester et al.[20] In addition to the biocides, the major degradation product of diuron, DCPMU (*N*-(3,4-dichlorophenyl)-*N*-methylurea), and the major degradation product of terbutryn and Irgarol 1051, M1 (*N*-tert-butyl-6-(methylsulfanyl)-1,3,5-triazine-2,4-diamine), were also analyzed in the runoff.

After the exposure time, the residual amount of biocide remaining in the film after the leaching studies was determined in ca. 1 g coating material (top render, paint) representing an area of 40 cm^2. The material was scraped from the substrate, milled, extracted with methanol and acetic acid (90/10 v/v) using sonification (30 min), and filtered by 0.45 μm membrane filter. An aliquot of ca. 1.5 mL of filtrate was transferred to vials for quantification by high pressure liquid chromatography coupled with UV (HPLC-UV). The limit of detection of the HPLC was 1 mg/L, with an uncertainty of 10%. The HPLC system included a reversed-phase C18-column (PerfectSil Target 5 μm, 150 × 4.6 mm). The eluent was methanol/potassium dihydrogen phophate 20 mM (65/35 v/v) at pH of 3.2 at a flow rate of 1.5 mL/min. The extraction method was tested on fresh wet coatings and freshly dried coatings, with extraction efficiencies found ranging e.g. from 80% for DCOIT to 95% for terbutryn and diuron.

Wind Driven Rain Calculations. For all ISO method calculations, $C_R = 0.72$, $C_T = 1$, and $O = 0.6$, corresponding to the roughness of suburban terrain, no upwind slope in the terrain, and an obstruction between 40 and 60 m away, respectively. The ISO method does not have the wall factor W for a one story house with a pitched roof, so the value of 0.4 was chosen, as it is the value for a two story building with a flat roof, the closest example given in the method.[11]

RESULTS

There were a total of 67 rain events taken into account over the time of the field experiment giving a cumulative horizontal rainfall amount of 993 mm. For the purposes of this study, a rain event is defined as a 24 h period in which it rains at least once, and can include small intervals during this time period where there is no rainfall. Of the 67 rain events, only 2 failed to produce runoff. The precipitation on the weather side is not always associated with runoff because driving rain is a phenomenon of precipitation, wind speed, and wind direction. Part a of Figure 3a shows a plot of runoff vs horizontal rainfall for each rain event, and part b of Figure 3 shows a plot of runoff versus WDR load predicted by the ISO method for each rain event. During the course of the year, the façade runoff fraction showed that between 5.6% (R_F) and 7.2% (R_FP_{Dx}) of the horizontal precipitation covered the vertical façade. Averaging across the four panels, 6.3% of the precipitation was collected at façades. Single runoff events yielded an average of 1.0 L/m^2 on painted coatings and less than 0.8 L/m^2 on the system without a topcoat (R_F). Irrespective of the coating system, 1.0 L/m^2 water was measured in one-third of all runoff events and less than 0.5 L/m^2 in nearly half of all runoff events. The runoff fraction of single events varied between 0.1% and 90% at four panels with a mean value of 9%. In about 40% of the runoff events the runoff fraction was less than 4%. Figure 4 gives the cumulative runoff for each panel of the model house versus cumulative horizontal rainfall, and also shows the cumulative predicted amount of potential runoff from the ISO method. Depending on the panel, the runoff amount cumulated over the events at the façade panels varied between 56 L/m^2 for the non painted panel R_F and 70 L/m^2 for the painted system R_FP_{Dx}. The façade runoff yielded an average of 61 L/m^2 water for four panels.

In this study, phenylureas and triazines (notably diuron and terbutryn) are the main focus of the release data. Figure 5a shows measured concentrations of diuron and terbutryn in runoff water from panels R_F against date. Due to constraints in analytical over the course of the study, approximately 65% of the total runoff was analyzed. However, 95% of the total runoff of the first 6 months of the study was analyzed, and the cumulative release versus cumulative runoff is shown in part b

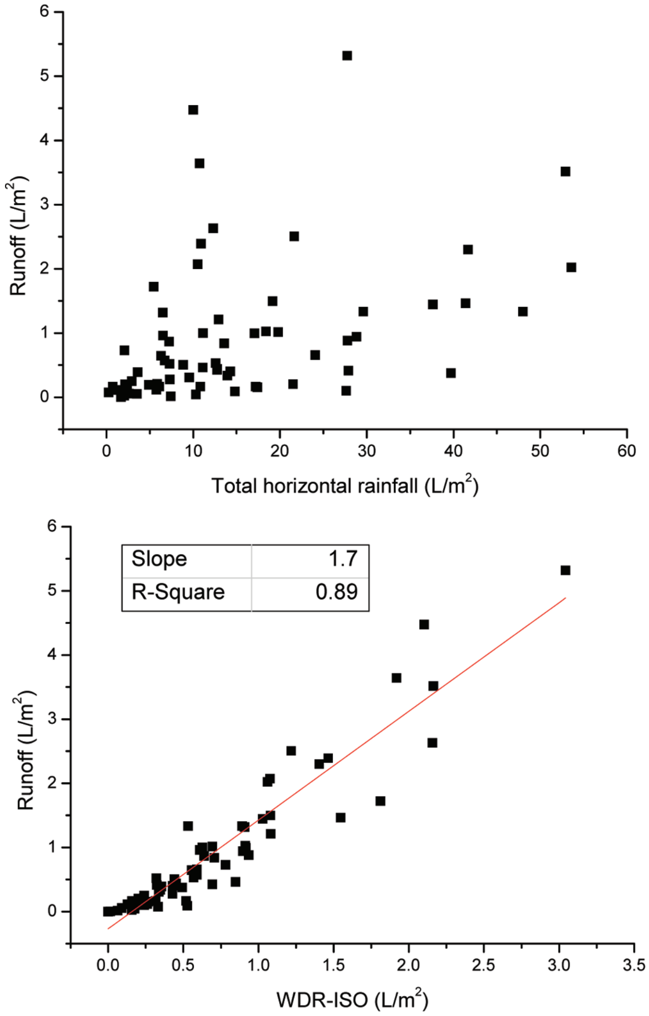

Figure 3. a-b. Plot of a) event façade runoff vs event horizontal rainfall and b) event façade runoff vs event wind driven rain amount predicted by the ISO method. Plot is for panel R_F with a hydrophobic top render containing biocides.

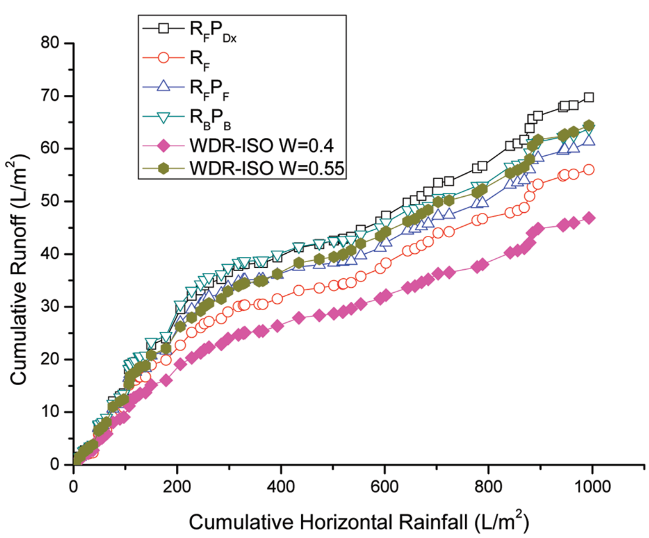

Figure 4. Plot of cumulative façade runoff vs cumulative total horizontal rainfall for all four panels and amounts predicted by the ISO method. Adjustment of the wall factor brings WDR-ISO prediction closer to measured amounts.

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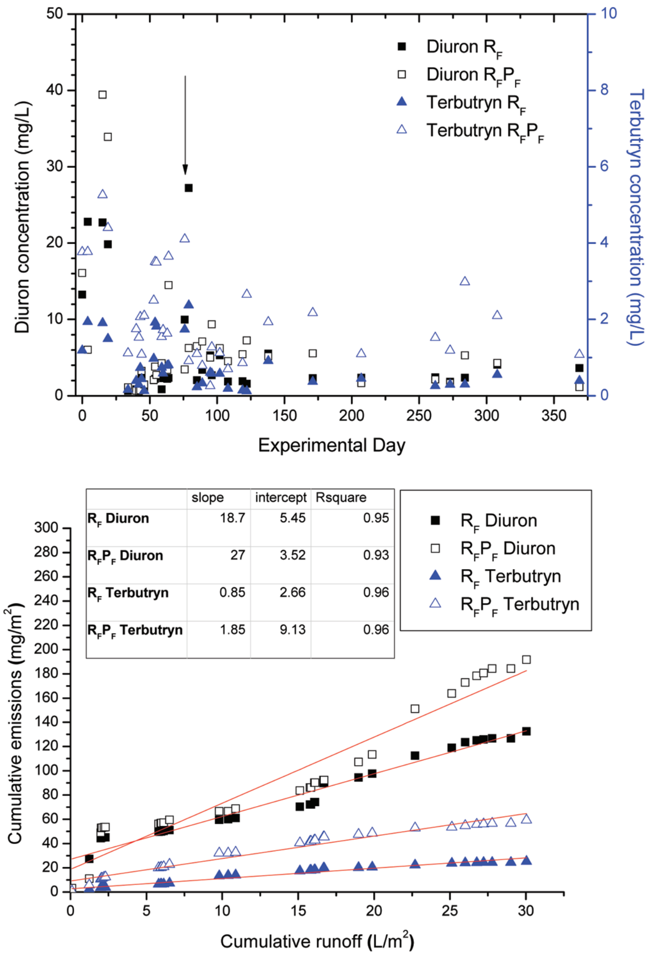

Figure 5. a-b. Plot of a) diuron and terbutryn concentration vs experimental time, and b) cumulative diuron and terbutryn emissions vs cumulative runoff. Concentrations tend to begin high and level out over time. Arrow marks rain event on April 3rd, 2008, 76 days after experiment start date of January 18, 2008. Linear fit in b) used to estimate total leached amount.

of Figure 5. Part b of Figure 5 shows a rough linear trend, which was extrapolated to estimate the total leached amount. Table 1 shows the estimated amount leached and measured amount remaining in each coating after the year of exposure.

DISCUSSION

Hydrological Results. The hydrological results unambiguously demonstrate the relationship between wind driven rain and runoff. In Figure 3, runoff clearly correlates to wind driven rain and not to total horizontal rainfall. The ISO method underpredicted the amount of runoff by at least 15%, as seen in Figure 4. The reason for this discrepancy likely lies in the selection of the wall factor *W*. There is no prescribed value for a one story building, so the selected value of 0.4 was probably too low. If the value is adjusted to 0.5–0.6, then the entire range of runoff volumes collected from the façades is captured. Future studies should take this discrepancy into account for single story buildings, especially against the background of the required risk assessment for biocides released from one story building façades demanded by the expected Emissions Scenario Document.[21] It should be borne in mind, however, that the ISO method is calculating potential runoff, with other mechanisms such as adhesion, absorption, and evaporation leading to decreases in runoff. In particular, the effect of adhesion water and evaporation based on the coating surface properties could be a source of error in the case of low runoff events, as described by Blocken and Carmeliet, where PMMA surfaces adhered an average of 0.06 L/m^2 of water.[16] In some cases, the recorded runoff amount exceeds the WDR load predicted by the ISO method, as seen in part b of Figure 3. Normally, one would expect to see runoff less than predicted WDR load due to various loss mechanisms. In these cases, higher than expected runoff totals could be due to weather fluctuations greater than the measurement frequency of 10 min, higher rainfall intensities, or wind patterns not captured by the ISO method.

When comparing runoff between panels, the two center panels, R_F and R_FP_F, showed lower runoff totals compared to the two outside panels. This is more a result of their location rather than their material properties, as validated computational studies by Blocken and Carmeliet show that the center of a façade receives less wind driven rain than the outsides due to wind flow directing rain to the top and edges.[13] The ISO method does not predict the edge effect, although it does predict the vertical change in wind driven rain amounts. Irrespective of this, for the purposes of calculating runoff totals for an entire façade, an average value is appropriate and is used for this study. Approximately 6–7% of the total horizontal rainfall came off the building as runoff. The measured runoff at the model house still exceeds the amount at real buildings. For example, at a west-oriented building façade of 10.5 m height without roof overhang a fraction of less than 0.7% of the precipitation amount was measured as façade runoff and in nearly 70% of all events even less than 0.2% (n = 72) (Supporting Information). This is very likely, in large part, due to the wind blocking effect, in which larger façades more easily impede wind flow and direct wind driven rain away from the

Table 1. Leached and Residual Amounts as a Percentage of the Applied Amount for All Panels, with Initial Amounts Given in mg/m^2; the Sum of the Leached and Residual Amounts Gives the Total Biocide Mass Recovery, with the Deficit Likely Coming from Losses Due to Degradation

panel	initial	recovery	diuron	isoproturon	terbutryn	irgarol	IPBC	OIT	DCOIT
R_FP_{Dx}	Render: 2388	% released	7.5	18.4	0.63	0.62	3.5	2.0	0.05
	Paint: 0	% remaining	46	34.8	73	73	34.8	32.5	18.6
R_F	Render: 2365	% released	9.5	23.0	2.1	2.3	6.2	2.5	0.1
		% remaining	42.8	33.6	62.5	57.9	20.8	19.7	16.2
R_FP_F	Render: 2325	% released	10.0	15.9	3.4	5.1	7.1	2.6	0.1
	Paint: 813	% remaining	45.2	39.5	71.5	71.5	30.1	30.1	17.9
R_BP_B	Render: 2357	% released	13.4	19.2	3.5	4.1	10.4	3.9	0.5
	Paint: 629	% remaining	46.3	46.3	61.7	68	22.6	18.5	30.8

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façade, although the difference is greater than would be expected from only a wall factor difference based on the ISO method. There are currently no methods being used to estimate runoff from a building façade in the determination of emissions functions, and it is advised to adapt simple wind driven rain relationships such as the ISO method to refine current regulatory strategies regarding vertical surface emission functions.

Leaching results. As can be seen in Table 1, biocides of the same class (phenylureas, triazines, isothiazolinones) show similar leachability and when comparing the render vs the render/paint systems in the two center panels, the render/paint system shows higher emissions in absolute terms due to its higher concentration of biocide in the thin paint layer. The outside panels had higher runoff amounts, so it is no surprise that the emissions from system R_BP_B tended to be a little bit higher, but the R_FP_{Dx} system generally showed the lowest total emission in spite of the high runoff amount due to the biocide free paint layer again acting as a diffusion barrier, similar to a controlled release system.

Leached biocide concentrations tend to be high early in the coating's lifetime, and then decay with time. This is consistent with most published lab studies.[18,19] There are not order of magnitude variations in the concentrations in the leachate after the initial "burst" period, however, so a likely mechanism of release is one of partition or solubility control.[22] The overall influence of weather factors in between rain events should be understood, as there could be potential for higher release dependent on these conditions, in spite of the generally constant concentrations. For example, there was a relative increase in the temperature as well as a long dry time before the first rain event of April 2008, marked by an arrow in part a of Figure 5. A recent study showed higher emissions with higher temperatures, which could be what drove this somewhat higher concentration for this particular rain event by increasing diffusion to the surface.[19] It is also interesting to note how diuron for the R_FP_F system behaves by having very low values after the initial burst, followed by a small rise before it maintains a consistent level. Temperature, increased times between rain events, UV irradiation exposure, and condensation events are all expected to have a significant influence, and will not necessarily affect each system in the same way or degree. While a detailed look at all of these factors for all rain events is beyond the scope of this study, it is worth pointing out that the estimated leached amount and the amount remaining for the biocides do not come close to the total amount for the biocide. This highlights the role that degradation plays, especially in comparison to lab studies.

Figure 6 shows the measured concentrations of M1, a degradation product of both Irgarol 1051 and terbutryn, during the course of the study. The levels begin low but increase significantly over time, even growing larger than the amounts of terbutryn and Irgarol 1051 found in the water. The levels of the degradation products measured do not account for the overall mass balance; for example, the total amount of DCPMU, the main degradation product of diuron, could only account for an estimated 0.4% of the diuron losses for panel R_F, with similar results for other panels. Combined with the diuron emissions from panel R_F of 9.5%, this does not come close to the total losses of 57%. This result points to a full degradation of the active ingredient or other loss mechanisms. The leached amounts of the quickly degrading active ingredients (IPBC, OIT, DCOIT) were also very low in this study, and they also had a very significant mass deficit, showing that degradation also occurs very quickly in the field for these biocides. All this highlights the importance of understanding loss mechanisms so that a true mass balance can be achieved under lab and field conditions.

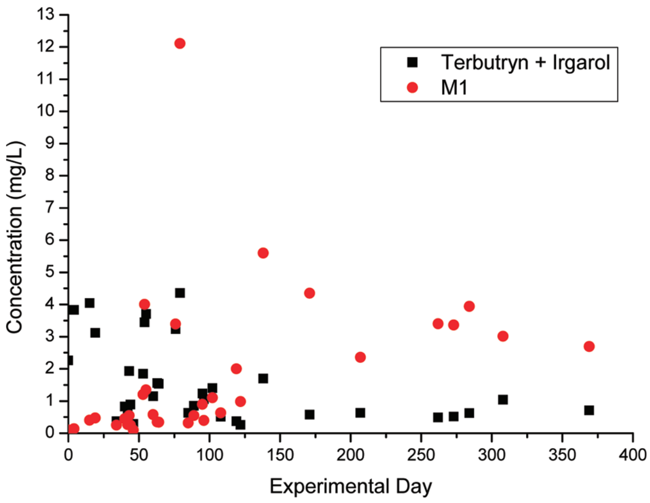

Figure 6. Concentrations of terbutryn plus Irgarol 1051 in façade R_F runoff water and M1, their main degradation product, vs experimental time.

In summary, it seems that after an initial burst release phase with high concentrations, biocide release from building façades in the field result in concentrations that are not highly variable, consistent with a solubility or partition controlled release mechanism. With knowledge of runoff volumes from the building façade provided by methods such as the ISO method used in this study, simple predictions of future emissions can be made, although this study spanned only a year and the service life of a coating is much longer. It is also apparent that biocide degradation is a major factor in the overall mass balance and should properly be taken into account. Products containing encapsulated biocides might exhibit longer service life due to smaller amounts released and protection from degradation. Future studies should work at understanding the differences between apparent release mechanisms in the lab and the field. In particular, it could be useful to use laboratory studies to understand how conditions between rain events influence emissions, as well as understanding degradation and controlling release to aid in product improvement and reduced environmental impact.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Supporting Information is available containing data from a runoff study performed on a building façade in the Zurich area, Switzerland. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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Supporting Information

Leaching of biocides from façades under natural weather conditions

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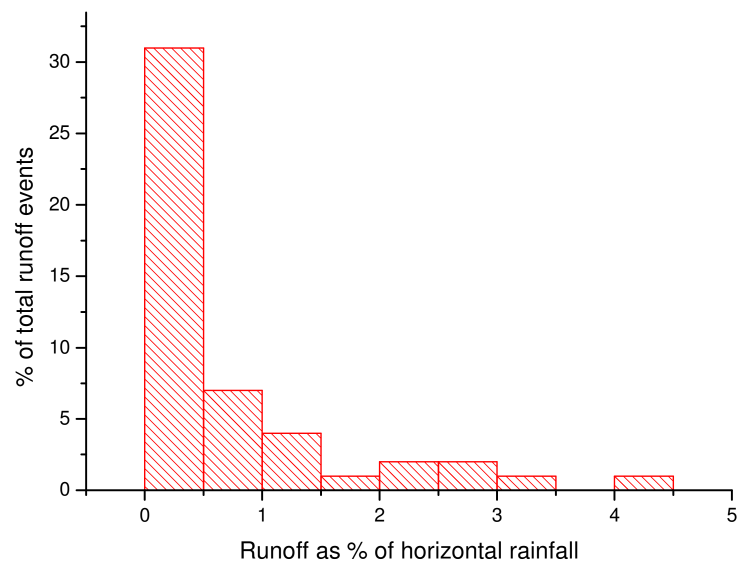
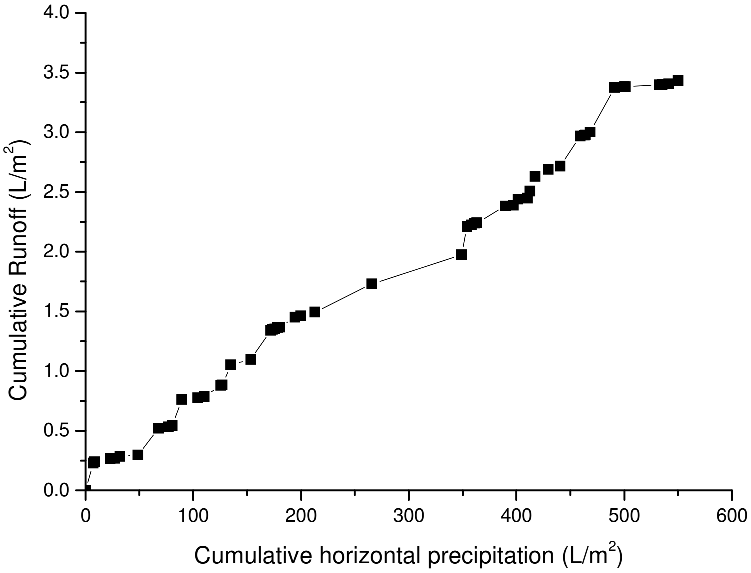
Number of Pages: 2 pages (SI-1 to SI-2)

Number of Figures: 2 figures

Facade runoff for west-facing 10.5 m high facade in Volketswil, Switzerland. Data taken from May 2007 until December 2007. Histogram (first plot) shows the distribution of the runoff percentage of horizontal rainfall for each rain event with 0.5 percentage point bins, and plot (second plot) shows cumulative façade runoff vs. cumulative horizontal precipitation for rain events causing runoff. Runoff was greatly reduced compared to the model house study due in large part to the wind blocking effect, and also due to a different experimental timeframe and site variations such as obstructions and surface roughness.




Cumulative Runoff (L/m2)
Cumulative horizontal precipitation (L/m2)
0.0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
4.0
0
100
200
300
400
500
600