water

Article

Management-Oriented Modelling of Tire and Road Wear Particle Fate and Transport in the Terrestrial and Freshwater Environment with a Global Perspective

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Abstract

Tire and road wear particles (TRWPs) are formed at the frictional interface of the tire and road surface and consist of polymer-containing tread with pavement mineral and binder encrustations. Their detection in various environmental compartments globally sparks increasing societal and regulatory interest. Solid quantitative information as a basis for managing and mitigating TRWPs in the environment is lacking however. This paper presents and demonstrates a model approach that produces catchment-scale terrestrial and aquatic TRWP mass balances anywhere in the world. A spatially and temporally explicit modelling method was used that builds on publicly available global datasets and process-based open-source modelling frameworks to describe hydrological processes, TRWP releases, fate and transport under a wide range of climatic conditions. High-resolution (<1 km) models were implemented and evaluated by demonstrating consistency with available field data for three watersheds on different continents. The approach provides comprehensive mass balances to underpin management of TRWPs that account for socio-economic, climate, geography and stormwater management gradients. Case study results revealed strong climate-induced differences: the fraction of vehicle-generated TRWPs exported to the estuarine environment varied between 2% (Seine watershed, France) to 18% (Yodo River watershed, Japan), corresponding to an increase in the fraction released to freshwater ecosystems from 20% to 36%, respectively. The modelling framework provides a consistent comparison between watersheds across the world. Limitations of the approach are its lack of local details and the uncertainties stemming from the still-developing scientific knowledge base.

Keywords: tire and road wear particles; mass balances; emissions; environmental fate and transport; modelling; water management

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1. Introduction

The U.N. Sustainable Development Goals (SDGs) established a global vision for a sustainable future organized around 17 goals and 169 targets across environmental, social, and economic systems. Integrated computational models and simulations are being increasingly recognized as a central tool advancing our progress towards many of the identified targets and their associated indicators [1]. Integrated models are being used in the context of SDGs concerning watershed management of a wide range of traditional and emerging pollution issues. In particular, multimedia models provide flexibility in assessing progress towards SDGs by combining the strengths of sub-models over a wide range of scales from local to global, and thus are one of the most frequently used approaches in SDG evaluations [1].

Tire and road wear particles (TRWPs) illustrate sustainability considerations at the intersection of product stewardship, environmental management, and sustainable infrastructure. As an unavoidable consequence of tire–road friction necessary for vehicle safety, TRWP generation connects UN Sustainable Development Goal (SDG) 12 (Responsible Consumption and Production) with watershed-scale environmental fate questions. Specifically, SDG Target 12.4 calls for environmentally sound management of chemicals throughout their lifecycle, while Target 12.6 encourages companies to adopt sustainable practices and sustainability reporting [2]. The global tire industry has recognized TRWPs under these targets as a key sustainability priority in their SDG sector roadmap [3].

Among the many environmental challenges addressed through integrated modelling, there is the sustainability of elastomers in tires. Elastomers are cross-linked viscoelastic materials, natural and synthetic, that exhibit the unique property of returning to their original shape after deforming under stress. These materials enable modern mobility, serving as key components in automobile and airplane tire tread. When considering automobile tire sustainability, there are design trade-offs caused by the conflicting demands on tire tread viscoelastic properties often described as the "magic triangle" of rolling resistance (fuel efficiency), wet traction (safety), and wear emission (tread wear emission) [4].

Mechanical energy dissipation during tire–road surface contact has been shown to generate tire and road wear particles (TRWPs) consisting of polymer-containing tread with pavement mineral and binder encrustations [5]. The tire tread fraction constitutes a blend of elastomers including natural and synthetic rubbers [6]. The material picked up from pavements is very diverse, dependent on road characteristics, traffic and environmental conditions [7]. TRWP sizes range from several hundreds of μm to less than 0.1 μm [8]. TRWP density is variable and was reported to range from 1.3 to 2.2 g cm^{-3} [6,9,10], dependent on the ratio of lower-density polymers, higher-density minerals and any other road dust components like organic matter, road marking material, and brake wear [7].

Coarse TRWPs (>10 μm), which represent the largest fraction of TRWPs by mass and volume, generally deposit on the road surface or nearby areas such as medians, curbs or sidewalks. Even if such particles are resuspended from these surfaces by wind or traffic turbulence [11], they rapidly settle again and stay on or close to the road [12]. Their potential for long-range transport via rivers is mainly governed by stormwater management infrastructure, unless direct surface runoff connects roads to nearby water bodies [13]. Field studies have shown that TRWPs occur in aquatic sediments (concentrations locally exceeding 10 mg/gDW; [6]). TRWPs are also found in estuaries and receiving coastal waters [14–16], where ecological impacts have been observed [17].

The challenge of managing the environmental impacts of automotive transport leads to the question of quantifying TRWP mass balances. Unice et al. [18] were the first to address this question by an integral model-based catchment-scale terrestrial and aquatic TRWP mass balance. This work presented a useful framework for integrated source and distribution modelling of TRWPs combining a hybrid mass-balance and hydrodynamic modelling approach [19]. The present study embeds and enhances the Unice et al. conceptual model and investigates the representativeness of the earlier findings for other watersheds across the globe. The study builds on harmonized open global datasets and high resolution, distributed modelling techniques that have become available in recent years to support the development of reproducible models of homogeneous quality across the globe [20,21]. A specific innovation in the present study is to investigate how watershed characteristics influence TRWP mass balances. Case studies are carried out in three contrasting watersheds from diverse geographic and socio-economic contexts: the Seine River (France), Chesapeake Bay (USA), and Yodo River (Japan). Field studies in these three watersheds have become available or are in progress [16,22–24] to support model evaluation.

The smaller TRWPs (<10 µm) are a suspected contributor to human health effects via exposure to fine dust by inhalation [25]. This fraction was reported to undergo atmospheric dispersion and long-range transport [26]. This study targets the quantification of aquatic and terrestrial mass balances. The contribution of long range atmospheric transport of TRWPs to these balances is not considered. This would add significantly to the complexity, without real benefit: though dominant in numbers, the contribution of TRWPs < 10 µm to the overall mass balance is small. The long-range transport by aquatic pathways and by atmospheric pathways can by good approximation be seen as separate questions.

The refined modelling framework presented in this paper supports management-oriented planning by mapping key environmental distribution determinants, parameterizing diverse case studies, and comparing model outputs to available field data. The paper concludes with a discussion of applicability, limitations and future research needs.

2. Materials and Methods

Spatially and temporally explicit emissions, hydrology and water quality models were constructed using the open-source distributed modelling software *wflow* (version sbm v0.7.3, [27]) and Delft3D (version 5.10.00, [28]). *wflow* was used to configure the hydrology models, which are coupled with a Delft3D-based TRWP emission model that quantifies the terrestrial pathways and calculates the emissions to surface waters. A coupled Delft3D-based water quality model simulates TRWP fate and transport in surface waters (Figure 1). The model approach and the parametrization of the model to case study watersheds on three continents are discussed below.

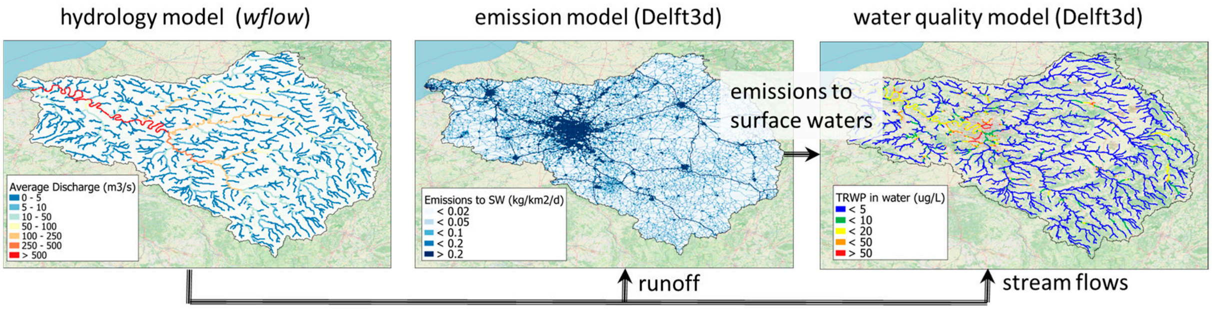


Figure 1. Overview of spatially and temporally resolved modelling components.

2.1. Hydrology and (Natural) Suspended Particulate Matter

The hydrological modelling uses the vertical simple bucket model (SBM) approach [27]. The software solves the water balance equation for every grid cell as a result of precipitation, evapo-transpiration and storage in the soil system. River, overland and subsurface flow laterally route water in a downstream direction (Figure 2).

Earlier applications demonstrated that high-quality hydrological models can be established across the globe using open-access datasets, provided that a sufficiently small grid size is applied [27]. In this study, hydrological models were built for three watersheds using global spatial inputs (elevation, soil properties, landcover, lakes) and spatio-temporal meteorological data (Table 1). The reproducible model-building tool HydroMT [29] was applied in this process. The simulation period was 2018–2020 with a grid cell size of 0.008333 degrees (750 m to 850 m).

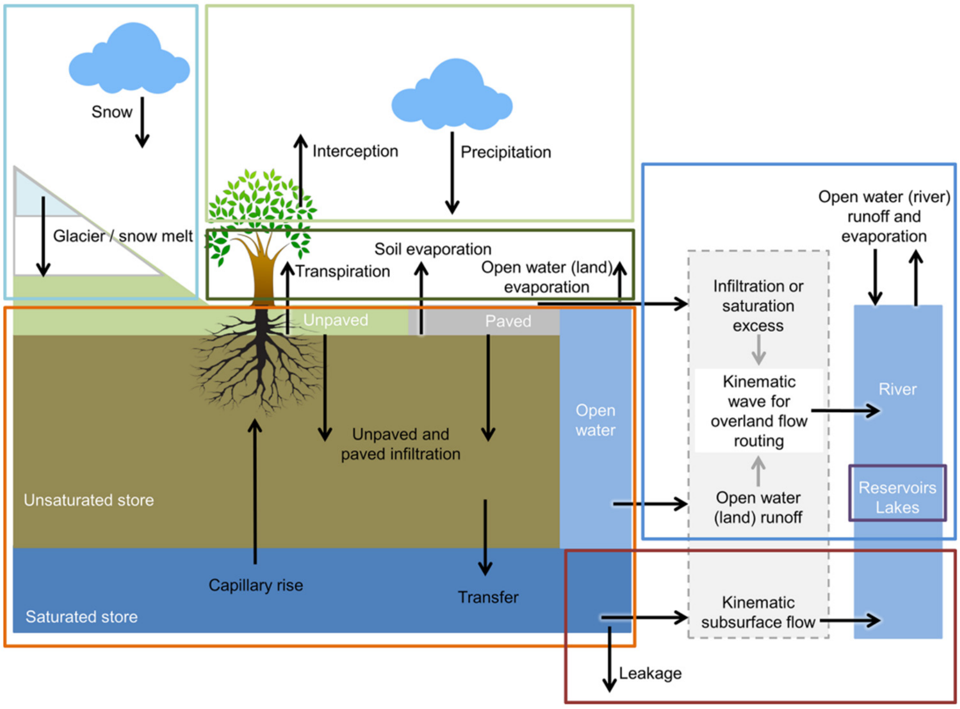


Figure 2. An overview of the different processes and fluxes in the wflow_sbm model [27].

Natural suspended particulate matter (SPM) is included in the modelling due to its interaction with TRWPs during in-stream transport [18]. SPM is separated in clay, silt particles and small aggregate fractions, reaching the river network after mobilization by natural erosive processes. These particles are either deposited in aquatic sediments of floodplains and riparian zones or transported downstream to estuarine environments [30]. Using the same framework and the input data as for the hydrology model, the delivery of SPM to streams was calculated using existing concepts implemented in the *wflow* sediment model [31]. Soil mobilization resulting from rainfall and overland flow is calculated as a function of slope, vegetation and soil properties, while delivery to streams is calculated by a transport capacity equation for the overland flow.

Table 1. Overview of global spatial data used.

Hydrology Model	Source	Processing
Elevation	MERIT Hydro Adjusted Elevations dataset [32]	Projection to grid, upscaling of the flow direction, derivation of river network and catchment delineation
Soil density Soil type	250 m resolution SoilGrids database [33]	Projection on grid, derivation of hydraulic conductivity, decline of hydraulic conductivity, porosity and residual porosity using pedo-transfer functions
Landcover	300 m resolution GlobCover map for 2009 [34]; 100 m resolution Copernicus Global Land Service 2015 land cover map [35]	Projection to grid, derivation of surface roughness, rooting depth of the vegetation, fraction of paved areas

Table 1. *Cont.*

Hydrology Model	Source	Processing
Lakes and reservoirs	GranD [36] and HydroLAKES [37]	Mapping outlet points to river network and main properties (area, average depth)
Precipitation Air temperature Potential evapotranspiration	ERA5 [38]	Downscaling to grid, correction of temperature using lapse rate correction, computation of potential Evapotranspiration using De Bruin formula from temperature, pressure, downwards radiation and incident solar radiation
TRWP Emission Model	**Source**	**Processing**
Population	Global Human Settlement Layer [39]	Aggregation on model grid
Settlement typology	Global Human Settlement Layer [40]	Projection on model grid
Road length	GRIP4 dataset [41]	Aggregation of road length per model grid cell, divided into highways and other roads

2.2. TRWP Terrestrial Pathways

Terrestrial pathways were simulated by a material flow analysis approach. TRWP mass balances were compiled per compartment in all grid cells with a daily time step (see Figure 3).

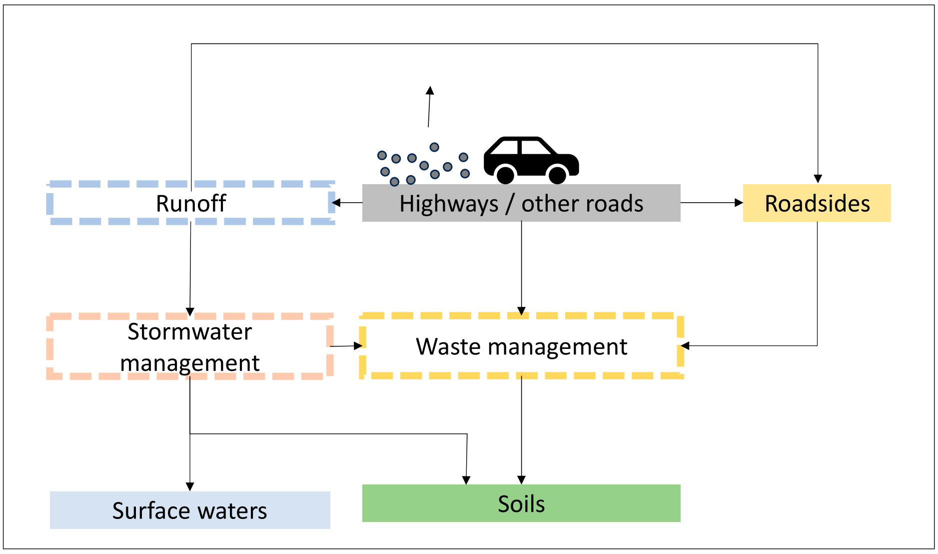


Figure 3. Schematic representation of the compartments included in the terrestrial model component. Solid boxes represent compartments with temporal or permanent storage. Boxes with a dashed outline represent compartments without storage (sum of inflows equals sum of outflows plus removal).

The mass balance equation for TRWPs on road surfaces M on two types of roads j (highways and other roads) is expressed as

$$\frac{dM_j}{dt} = E_j(1 - f_{PM10}) - F_{aeol.,j} - F_{runoff,j} - F_{por.,j} - F_{sweep,j} \quad (1)$$

where E represents TRWP releases (g s^{-1}); f_{PM10} (-) is the fraction of TRWP releases emitted to the atmosphere as PM10; $F_{aeol.}$ is the transport away from road surfaces by aeolian mechanisms (traffic induced turbulence and wind, Equation (S3-2)); F_{runoff} is the transport away from road surfaces by road runoff (Equation (S3-1)); F_{sweep} is the removal from road surfaces by road sweeping; and $F_{por.}$ is the removal of TRWPs from road surfaces by regular cleaning of porous asphalt. Note that all fluxes F are g s^{-1}. Aeolian transport is continuous, while runoff-mediated transport is intermittent, controlled by the occurrence and intensity of rainfall events. This creates a spatial and temporal distribution between both mechanisms. It also creates a variable TRWP concentration in road runoff, with higher concentrations after a longer dry period ("first flush").

The mass balance equation for TRWPs along roadsides R of roads of category j is

$$\frac{dR_j}{dt} = F_{runoff,j}\left(1 - f_{int.,j}\right) + F_{aeol.,j} - k_{soil}R_j \tag{2}$$

where F_{aeol}. and F_{runoff} are defined as above; f_{int}. (-) is the intercepted fraction of road runoff; and k_{soil} (s^{-1}) is the degradation rate of TRWPs by various processes. In rural areas, a fraction of the roads is assumed to have run-off management infrastructure in place that intercepts the runoff, depending on the road category. In urban areas, all runoff is assumed to be intercepted.

The stormwater management (SM) compartment is defined by the following equation:

$$\sum_{j=1,2} F_{runoff,j}\, f_{int.,j} = F_{SM,rem} + F_{SM,leak} \tag{3}$$

where the inflows are defined as above; $F_{SM,rem}$ is the TRWP flux removed; and $F_{SM,leak}$ is the TRWP flux leaked to surface waters. This latter flux is passed to the surface water quality model component. In rural areas, management infrastructure can be simple (swales or grassed ditches) or more elaborate (settling ponds, detention basins, retention basins). The TRWP fraction removed depends on the infrastructure applied and is input to the model. The non-removed fraction is leaked to the nearby river. In urban areas, runoff is routed either to a wastewater collection system (combined sewer system) or a separate stormwater collection system. TRWPs in runoff collected in a combined system is removed in wastewater treatment plants (WWTPs) or leaked via effluents or combined sewer overflows (CSOs). TRWPs in runoff collected in a separate system is removed by specific measures (filters, infiltration) or leaked via effluents. The removed TRWPs are transferred to the waste management system (or remain in soils if infiltrated). See SI-3.3 for details.

The waste management (WM) compartment is defined by the following equation:

$$\sum_{j=1,2} \left(F_{por.,j} + F_{sweep,j}\right) + F_{SM,rem} = F_{WM,stor} + F_{WM,leak} \tag{4}$$

where the inflows are defined as above; $F_{WM,stor}$ is the TRWP flux safely stored and thus isolated from the environment; and $F_{WM,leak}$ is the leakage of TRWPs to soils away from road surfaces by distribution of sewage sludge, waste dumping, etc. (See SI-3.4 for details).

The mass balance equation for TRWPs in soils away from roadsides S is

$$\frac{dS}{dt} = F_{WM,leak} - k_{soil}S \tag{5}$$

Hydrological forcing is derived from the output of the hydrological model component.

2.3. TRWP In-Stream Fate and Transport

TRWP in-stream fate and transport is represented by the one-dimensional advection–dispersion equation [42], with additional source terms accounting for hetero-aggregation, settling and resuspension:

$$\frac{\partial}{\partial t}(AC) = \frac{\partial}{\partial x}\left(AD\frac{\partial C}{\partial x}\right) - \frac{\partial}{\partial x}(QC) + IS \tag{6}$$

where C is concentration (g m^{-3}); A is cross section (m^2); t is time (s), x is distance (m); Q is longitudinal water flux (m^3 s^{-1}); D is longitudinal dispersion (m^2 s^{-1}); and IS is the in-stream source term (g m^{-3} s^{-1}).

The equation is solved for 3 size fractions of natural particles N_k, 4 size fractions of TRWPs T_i and $3 \times 4 = 12$ aggregates NT_{ik}, using the output from the hydrological model to derive A and Q. The source term IS is defined separately for natural particles, TRWPs and hetero-aggregates:

$$\frac{\partial N_k}{\partial t} = D_k - F_{set,k} - \sum_{i=1,4} F_{agg,ik} \tag{7}$$

$$\frac{\partial T_i}{\partial t} = F_{SM,leak,i} - F_{set,i} - \sum_{k=1,3} F_{agg,ik} \tag{8}$$

$$\frac{\partial NT_{ik}}{\partial t} = -F_{set,ik} + F_{agg,ik} \tag{9}$$

where $F_{SM,leak}$ represents the emissions of TRWPs to surface waters as discussed above, D_k. represents the delivery to streams of natural particles as calculated by the *wflow* sediment model, F_{set} represents net settling of particles and $F_{agg,ik}$ represents hetero-aggregation between a natural particle N_k and a TRWP T_i. The settling velocity of all particles was estimated based on their size, density and shape. Net settling of all particles was implemented by using the empirical deposition law from Krone [43]. The critical shear stress for deposition was expressed as a function of the settling velocity and a single critical Rouse number, independent of particle properties (SI-4). This critical Rouse number was then calibrated to obtain a realistic representation of the retention of natural particles, the latter derived from an analogy to the retention predicted by an empirical relationship between specific runoff and normalized phosphorus retention (a pollutant strongly associated with natural particles) (SI-5). Hetero-aggregation was modelled using the formulas as reported by Unice et al. [18].

The equilibrium sediment concentration of TRWPs $C_{s,eq}$ was estimated by assuming a steady state between the net settling on one hand and burial and degradation on the other hand (SI-6):

$$C_{s,eq} = \frac{F_{TRWP}}{(k_b + k_e)\delta(1-\varphi)\rho} \times 10^6 \tag{10}$$

where F_{TRWP} is the deposition of TRWPs in g m^{-2} s^{-1}, k_b is the burial rate, k_e is the degradation rate (both s^{-1}), δ is the top sediment layer thickness (m), φ is the porosity (-), and ρ the solid matter density (g m^{-3}). The burial rate was determined by the net settling of natural particles (SI-6). The time needed to reach 95% of the equilibrium concentration equals (SI-6)

$$T_{95} = \frac{-ln(0.05)}{k_b + k_e} \tag{11}$$

2.4. TRWP Model Input and Parameterization

Table 2 provides an overview of case-study-specific model input, generic model input, and parameterization, respectively.

Table 2. Overview of model input and parameterization.

Parameter	Number	Source	More Information
Country traffic volume (vehicle km yr^{-1})	**	From national statistics data, downscaled by using global spatial data on road length and settlement type (Table 1)	Section 2.4.1; SI-1

Table 2. *Cont.*

Parameter	Number	Source	More Information
TRWP release per vehicle km	146 mg vkm^{-1}	Based on values by vehicle type and by road type [18,44]	Section 2.4.1; SI-1
Fraction to PM10	2% (0–10%)	[6,18]	Section 4
TRWP wash-off	0.2 mm^{-1}	Wash-off coefficient parameterization as in [18]	SI-3.1
TRWP aeolian transport	0.15 d^{-1}	First order approximative rate constant, estimated in this study	SI-3.2
Porous asphalt	**	Various	SI-8
Street sweeping	**	Various	SI-8
Share of runoff intercepted	1.0 in urban areas, ** in rural areas	Various	SI-8
TRWP degradation in soils	Half life 490 d	[45]	Section 4
Share of combined sewers	**	Various	SI-8
Combined sewer overflows	**	Various	SI-8
Trapping in separate stormwater collection systems	20%	Estimate based on particle trapping data [46]	SI-3.3
Removal in WWTPs	95%	[18]	SI-3.3
Hetero-aggregation efficiency	0.01 (-)	[18]	
TRWP diameter in µm	10–50: 11%; 50–100: 38%; 100–150: 32%; 150–250: 19%	[10] (supporting information), [18]	
TRWP density	1.8 g cm^{-3} (1.6–2.0)	[18]	Section 1
TRWP aspect ratio	0.64 (-)	[18]	
Critical Rouse nr	**	Determined by calibration of fate of natural particles in present study	SI-5; SI-6
TRWP degradation in sediments	Half life 4900 d	[18]	

Note(s): ** Case-study-specific input was used.

2.4.1. TRWPs Released on Road Surfaces

TRWP releases to road surfaces were calculated as the product of country traffic volume statistic data (vehicle-kilometre per year, vkm y^{-1}) multiplied with an emission factor (EF; mg vkm^{-1}). EF depends on (a) tire technology, (b) vehicle type, (c) road conditions and (d) driving behaviour [6]. EFs were available by vehicle type and by road type (urban roads, rural roads, highways), the latter serving as a proxy for road conditions [18,44], assuming average tire technology and driving behaviour (SI-1). Where traffic volume data differentiated over vehicle and road types were not available, country-based traffic volume estimates were combined with a road-type and vehicle type averaged EF of 146 mg vkm^{-1} (SI-1).

TRWP releases were downscaled by using global spatial data on road length and settlement type (Table 1). TRWP releases were allocated in accordance with road presence in each grid cell, using weight factors for road types (highways vs. other roads) and settlement types (7 classes from "very low density rural" to "urban centre"). The weight factors used were validated by benchmarking to a very detailed spatial TRWP releases inventory for the whole of Germany [12] (SI-2).

2.4.2. TRWP Properties

The model distinguishes four size classes between 10 and 250 µm, with a triangular distribution centred around 100 µm (Table 2). While this distribution was first derived from older research [5,18] and references therein, the validity of this assumption was confirmed by a recent study [10] that analyzed the particle size distribution of artificially produced fresh TRWPs and the changes thereof under various laboratory-simulated ageing processes. Almost all fresh TRWP samples tested strongly resembled this triangular distribution (supporting information to [10]). The mean density of TRWPs was found to be 1.8 g cm^{-3}, equal to the earlier central estimate [18]. As earlier the model results were shown to be sensitive to the particle size distribution and the particle density [47], the sensitivity of the model to the assumptions made was tested, guided by information on the effects of ageing of TRWPs [10,48] (SI-11).

2.5. Watershed Models

Models were implemented for three watersheds on different continents: the Yodo River and Lake Biwa watershed in Japan, the Seine watershed in France and the drainage basin of the Chesapeake Bay in the United States. Some characteristics of these watersheds (size, population density, degree of urbanization, road density and climate) are collected in Table 3. These watersheds were selected to demonstrate the applicability across the globe and were considered sufficiently different (e.g., 11-fold difference in low to high population density) to provide insights in the factors affecting variability of TRWP mass balances. For the parameterization of the stormwater management infrastructure, no global data exist. A representative watershed characterization was determined using available data from various sources (SI-8).

Table 3. Main characteristics of current model applications (model domain aggregates derived from input data (Table 1).

	Yodo-Biwa	Chesapeake Bay	Seine
Area (km^2)	8246	171,210	76,135
Population density (cap km^{-2})	1191	106	221
Road density (km km^{-2})	4.0	1.1	1.6
Share of urban area (-)	15.1%	1.9%	2.0%
Annual rainfall (mm/y) (2018–2020)	1901-1603-1912	1526-1201-1233	843-869-815
Area-specific river runoff (mm/y) (2018–2020)	1115-841-1185	709-455-478	325-283-350
Population centres	Osaka, Kyoto	Washington DC, Baltimore, Richmond	Paris, Rouen

2.6. Model Evaluation

Model evaluation was conducted by comparing observed water concentrations at 8 stations along the Seine [23] to the range of simulated water concentrations. In addition, observed sediment concentrations in all three case studies [22,23] were compared to simulated equilibrium sediment concentrations. Details about the field data and the details of the comparison are collected in SI-7.

2.7. Indicative Management Scenarios

A set of scenarios was simulated to show the capabilities of the model to quantify the impact of mitigation measures (Table 4). A conceptual combined sewer (CS) systems optimization scenario was defined by doubling the fraction of combined sewers and reducing the overflow volume by 80%. A conceptual separated sewer (SS) systems optimization scenario was defined by halving the fraction of combined sewers and implementing green and grey infrastructure to purify and infiltrate the stormwater.

Table 4. Overview of stormwater management scenarios.

Scenario	Share of Combined Sewers	Combined Sewer Overflows	Fate of Collected Stormwater
Baseline	Best estimate	Best estimate	Best estimate
Combined sewer systems optimized	Double	−80%	Unchanged
Separated sewer systems optimized	Half	Unchanged	80% locally managed

3. Results

3.1. Model Implementation

The simulated spatial distribution of TRWP releases in the Seine watershed clearly reflect the population centres and road network (Figure 4). More illustrations are available in SI-9.

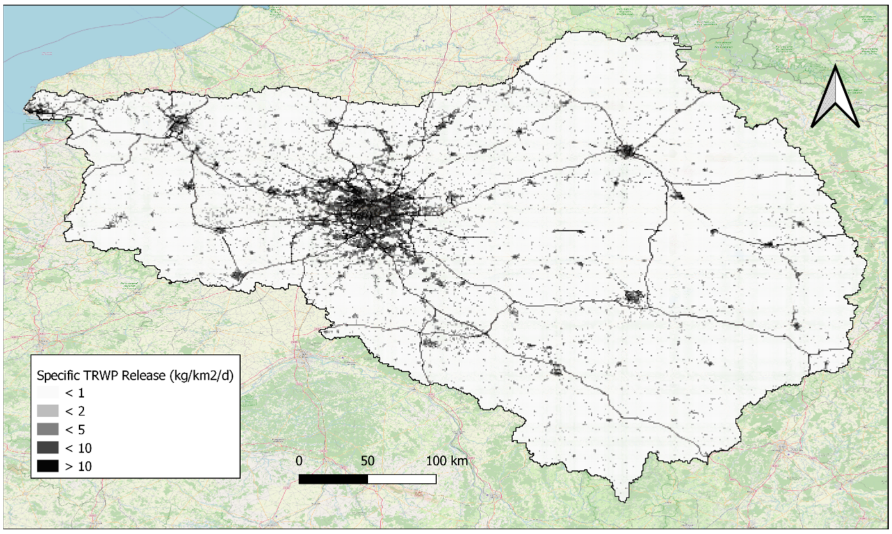


Figure 4. Simulated area specific releases of TRWPs to road surfaces (kg km^{-2} d^{-1}) in the Seine watershed (background from OpenStreetMap).

3.2. Model Evaluation

Water concentrations observed in 2021 (expressed as µg/L of tread) were well within the simulated 10–90 percentile range (Figure 5). The model reproduces the observed increase between km 398–332 (Paris input; +81% in survey, +146% in simulated mean). It underestimates the observed decrease between km 332–114 (downstream of Paris towards Rouen; −67% in survey, −26% in simulated mean). This is potentially attributable to the assumptions regarding the 10–50 µm TRWP size fraction, which constitutes 11% of the total releases and is characterized by slow settling and limited trapping in sediments. The modelled result fails to capture the observed 4.2-fold increase between km 114–8 (downstream Rouen to the estuary mouth), likely due to estuarine circulation and/or resuspension processes not included in the modelling concepts.

The 10- and 90-percentiles of model results in Figure 1 show the strong temporal variability of the simulation results. This variability is not reflected in the single field survey used for evaluation. The Seine River discharge during the 2021 survey period was verified to be within the normal range of conditions. Similar conditions occurred regularly during the 2018–2020 simulation period. Therefore, the apparent temporal mismatch between the simulation and survey periods is not expected to affect this evaluation.

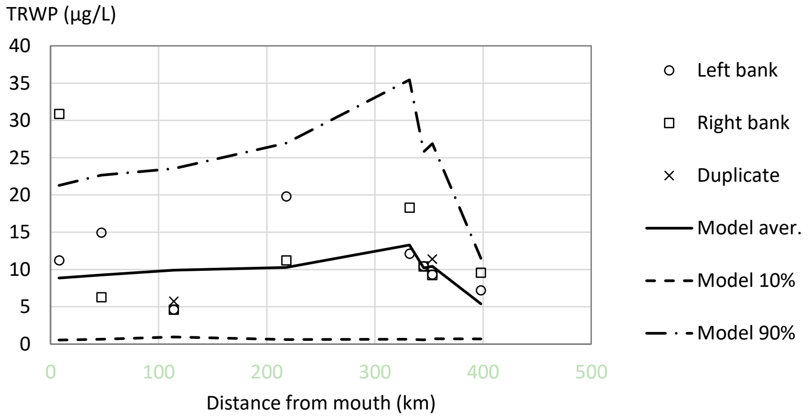


Figure 5. Comparison of simulated water concentrations of TRWPs (2018–2020) and observed concentrations at eight stations along the Seine River (May–June 2021). Analysis results are shown from sampling at the left bank and the right bank for all stations plus two duplicates.

Simulated TRWP deposition fluxes were used to calculate TRWP equilibrium sediment concentrations (Equation (10)). These were compared to measured sediment concentrations, expressed as mg/kg of tread and normalized to 2% sediment TOC (Figure 6). It is emphasized that the model does not directly calculate sediment concentrations. The simulation results over a period of 3 years were interpreted and extrapolated to equilibrium, assuming constant emissions. As this presumes that sediments respond to water fluxes of fine particles (depositional environment, as opposed to an erosive environment; SI-6), analysis results from samples with a low (<10%) share of fines are shown in a different shade. Such samples may indicate a more erosive environment for which a comparison to model results may be less meaningful.

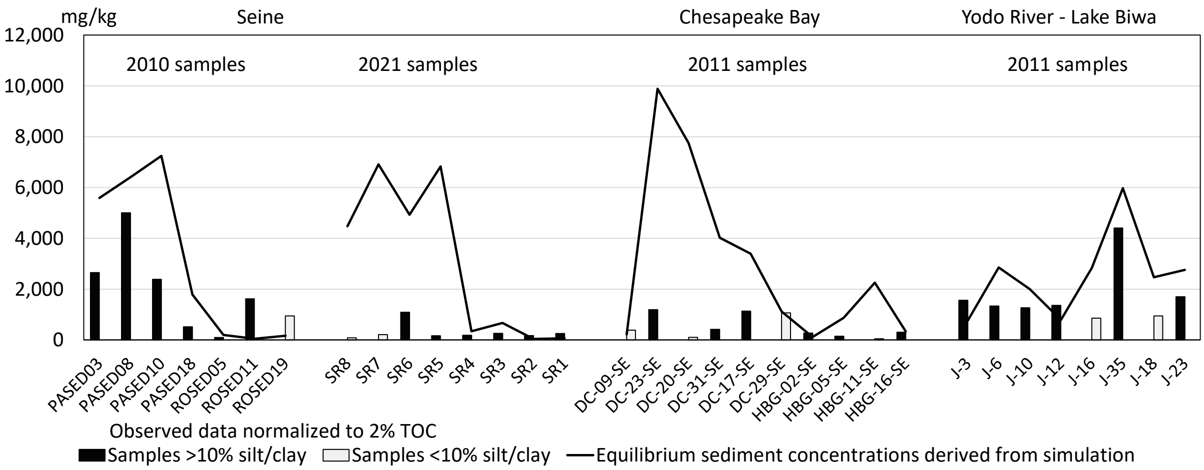


Figure 6. Station-by-station comparison of equilibrium sediment concentrations derived from simulation results for three watersheds (using 2018–2020 hydrology) and observed concentrations (from 2010/2011 and 2021). Field data are normalized to 2% of TOC in all plots. For interpretation, stations with <10% clays/silts were given a different colour.

For the Seine basin, model results reflect spatial gradients in the data reasonably well. The absolute values match the 2010 observations better than those from 2021. Near Paris (stations SR5-7, PASEDii), 2010 observations are much higher than 2021 observations. Near Rouen (stations SR2-3, ROSEDii), the differences are much smaller. The difference around Paris could reflect a true time trend, as sewer system upgrade works have been carried out in recent decades. Additionally, approximately 5 million tonnes of material are dredged from the Seine estuary annually [49]. These dredging operations, which occur on an ongoing basis, could potentially affect TRWP concentrations.

For the Chesapeake Bay basin, the model reproduces the observed concentrations and the spatial patterns only partly. The model overestimates the observations especially in two stations in the heart of Washington, DC (stations DC-23 and DC-20). The discrepancy could be caused by the temporality or frequency of dredging relative to sample collection, or erroneous input regarding the local stormwater management infrastructure and/or TRWP retention therein. For the Yodo River basin, the spatial gradients and concentration levels are well captured.

3.3. Simulated TRWP Mass Balances for Three Watersheds

Table 5 shows the total releases of TRWPs in the three watersheds, expressed by the total amount and the area-specific value (relative to total surface area). The table also shows the terrestrial and aquatic balances, relative to the total releases. Note that these results reflect a situation without any leakage from the waste management system, implying that the term "To soils" is zero. A complete uncertainty and sensitivity analysis of the mass balances presented in Table 5 was not performed. This aspect of the present results will be discussed below (also based on the results from the sensitivity tests carried out for TRWP particle size and density (SI-11)).

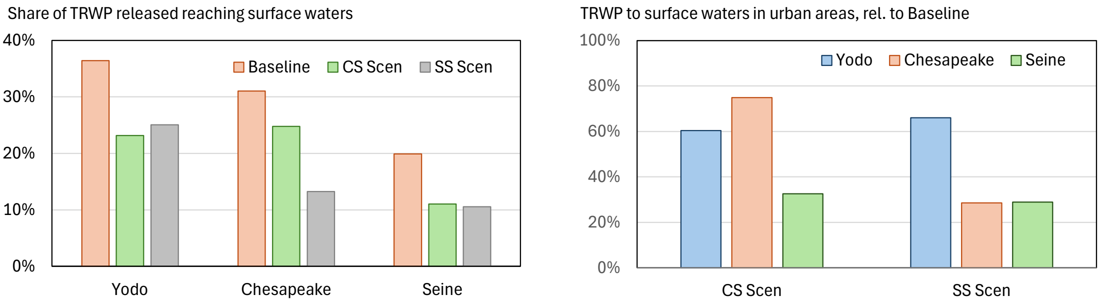
Table 5. Totals of TRWP releases, expressed as absolute values and as area specific values (total surface area in km^2), and terrestrial and aquatic mass balances, relative to total releases. Information provided for three basins on different continents, using 2018–2020 forcing.

TRWP Releases	**Yodo-Biwa**	**Chesapeake**	**Seine**
Releases of TRWPs to roads (kt a^{-1})	8.81	42.31	23.00
Releases of TRWPs to roads (kg km^{-2} d^{-1})	0.976	0.226	0.276
Terrestrial Balance (% of Releases)	**Yodo-Biwa**	**Chesapeake**	**Seine**
Releases of TRWPs to roads	100%	100%	100%
To atmosphere (small TRWPs)	−2%	−2%	−2%
To roadsides	−30%	−40%	−49%
To soils (away from roads)	0%	0%	0%
Removed and safely stored	−31%	−26%	−29%
Emissions to surface waters	−36%	−31%	−20%
Water System Balance (% of Releases)	**Yodo-Biwa**	**Chesapeake**	**Seine**
Emissions to surface waters	36%	31%	20%
To aquatic sediments	−18%	−26%	−18%
To the estuary	−18%	−5%	−2%

3.4. Indicative Stormwater Management Scenarios

The results of the scenario simulations assuming different management of stormwater and wastewater are shown in Figure 7.

These results show that the effect of infrastructure optimization as parameterized here is appreciable: up to about half of the emissions to surface water can be avoided. The Yodo-Biwa and Seine watersheds show an about equal reduction for both scenarios, while the Chesapeake Bay shows a stronger reduction for the separated sewer system scenario. The results from the CS scenario mostly reflect the share of combined sewer systems in the baseline scenario (0.3 in Yodo, 0.2 in Chesapeake and 0.43 in Seine) as a relative increase was assumed. The results from the SS scenario reflect the presumed lower efficiency of local stormwater management systems due to space limitations (SI-10).



(**a**) (**b**)

Figure 7. (**a**): Share of TRWPs released on road surfaces reaching surface waters, in the baseline scenario and in the combined sewers (CS) and separated sewers (SS) optimization scenarios; (**b**): TRWPs reaching surface waters in urban areas, relative to baseline scenario. Baseline results are as in Table 5.

4. Discussion

4.1. Basin Scale Mass Balances

The implementation of the model to watersheds situated on three different continents employed homogeneous methods and data. The results can therefore be expected to realistically represent the impact of differences between the watersheds on TRWP mass balances. The results show clear differences between the three basins. Simulated absolute releases of TRWPs are highest in Chesapeake, due to the size of the watershed. Area-specific releases are highest in Yodo-Biwa, as a result of the population density and large share of urban areas. Below, the fate of these releases is discussed and compared to TRWP balances used in global models and SDG-related policy studies. These are non-site-specific and often based on expert judgement. We are not aware of other (modelling or field) studies providing alternative watershed-scale TRWP mass balances to evaluate present results in more detail.

The three simulated basins range from relatively dry (Seine) to relatively wet (Yodo-Biwa), with Chesapeake in the middle (Table 5). The more rainfall and therefore road runoff, the lower the share of TRWPs generated on road surfaces that remains along roadsides (49% in Seine to 30% in Yodo-Biwa). A study by OECD [50] neglects this climate dependency and assumes that 45% remain in roadsides or porous pavements.

The runoff management infrastructure prevents a fraction of TRWPs in runoff from reaching surface waters. The fraction that does reach surface waters again reflects the climate (20% of generated TRWPs in Seine to 36% in Yodo-Biwa). A study by IUCN uses estimates of 12% reaching surface waters in urban areas and 2% in rural areas, independent of climate [44]. The OECD study assumes all TRWPs in rural road runoff leaked to the environment (unspecific about surface waters or soils) and complete treatment of urban road runoff in the wastewater management system (neglecting separation of wastewater and stormwater) [50].

The fate of TRWPs reaching surface waters again reflects climate conditions: the wetter the basin, the lower the trapping in the river system (90% trapped in Seine to 50% trapped in Yodo-Biwa). The trapping percentages are also affected by the distance of population centres from the estuary; a large share of urban area in Yodo-Biwa is close to the coast (Kyoto/Osaka). That also holds for Baltimore and Washington, DC, in Chesapeake. Paris on the other hand is far away from the estuary. Published global models assume 75% trapping in smaller basins and 90% trapping in larger basins [51,52]. The IUCN study neglects this trapping and assumes 100% transfer to marine waters [44]. In the present model, the share of TRWPs generated on road surfaces that reaches the estuary amounts to 2% in the Seine, 5% in Chesapeake and 18% in Yodo-Biwa.

From the comparison to existing assumptions, it appears present results can add consistency and provide insight in controlling factors. Key TRWP environmental endpoints are (1) roadside soils, especially in drier areas, (2) aquatic sediments and (3) the estuary and coastal zone. A further endpoint can be the redistribution of TRWPs removed by after-release remediation: street-sweeping, porous asphalt and storm water management infrastructure [7]. An example is the redistribution of sewage sludge containing TRWPs. The latter endpoint was outside the focus of the present work but could be important.

4.2. Uncertainty of Basin Scale Mass Balances

The balances presented above are to be interpreted accounting for uncertainty. The absolute numbers are affected by the uncertainty of TRWP releases quantification. Country-level traffic volume estimates, expected to be readily available, were used as a starting point and combined with emission factors per vehicle kilometre. It has been pointed out that emission factors reported in the literature have a very narrow empirical basis and need experimental studies to provide a better basis [53]. Country-level TRWP generation estimates from different sources, based on different methods and data, show a variation of at least a factor of 2 [6]. This uncertainty has a direct linear effect on all present model results.

A novel way of spatial downscaling of TRWP releases could be applied based on various open harmonized global datasets and a detailed study of Germany to benchmark the method [12]. The benchmark results showed that at the spatial scale of the 13 larger German federal states the present approach shows deviations from the benchmark of −33% to +48% (SI-2). Possible reasons could be the lack of spatial information on traffic density and the simple urban/rural classification of roads, neglecting for example road curvature and slope.

The simulated fate and transport expressed as a fraction of TRWPs released on road surfaces is not affected by the above uncertainty in TRWP releases, presuming that emission factors can be considered homogeneous throughout the watershed. In earlier work, a formal uncertainty assessment was conducted for the Seine watershed, accounting for likely ranges of TRWP properties, stormwater management infrastructure and model parameters [47]. As the present model has many similarities, these results still hold value. A very strong sensitivity was found to TRWP properties [47], which is expected to hold also for the present model. This was investigated here by a sensitivity assessment (SI-11). A smaller sensitivity was found to the assumptions related to stormwater management infrastructure [47]. This aspect was investigated by the present stormwater management scenarios. A strong sensitivity was found to the parameterization of the fraction of TRWPs carried away by road runoff [47]. Though this parameterization was modified in the present model, the parameter remains uncertain. The above has been quantitatively summarized in Table 6.

The information compiled in Table 6 illustrates the main achievement of this study: climate and hydrology differences play a decisive role for TRWP mass balancing, overshadowing for instance the sensitivity to TRWP size distribution and density.

The modified parameterization of the fraction of TRWPs carried away by road runoff makes this fraction time and space dependent, consistently depending on local weather. The parameterization still relies on older field studies that did not document weather conditions [54,55]. Recent detailed and well-documented field studies to parameterize this aspect in more detail are lacking. Such studies are challenging as they need to quantify a mass balance over a road section (TRWP generated, accumulating in roadside soils and carried away by runoff) and record all factors determining this mass balance (traffic, weather).

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Table 6. Summary of available sensitivity tests and uncertainty analyses for the relative fate and transport of TRWPs; top: fraction leaked to surface waters, bottom: fraction exported to estuary.

TRWPs Relative Leakage to Surface Waters	Yodo-Biwa	Chesapeake	Seine
Best estimate (this study, Table 5)	36%	31%	20%
Ranges in stormwater management scenarios (this study, Figure 7a)	23–36%	13–31%	11–20%
Formal uncertainty analysis ([47], Table 3)	n.a.	n.a.	14–29%
TRWPs Relative Export to Estuaries	**Yodo-Biwa**	**Chesapeake**	**Seine**
Best estimate (this study, Table 5)	18%	5.3%	2.1%
Ranges in stormwater management scenarios (this study, Figure 7a)	11–18%	2.2–5.3%	1.1–2.1
Ranges in particle size and density sensitivity tests (this study, SI-11)	16–24%	4.9–6.2%	1.8–2.4%
Formal uncertainty analysis ([47], Table 3)	n.a.	n.a.	1–13%

The model was evaluated using 2021 surface water samples from the Seine River with satisfactory results (Figure 5). Simulated concentrations show significant temporal variability, which could not be resolved by the field data [23]. The present results were also evaluated by a comparison to measured sediment concentrations for all three study areas. The results were acceptable, considering different additional uncertainties. The translation of simulated TRWP deposition fluxes to sediment concentrations derives a local burial rate from the simulated local deposition of natural particles. The resulting burial rates reach values up to 7.5×10^{-5} d^{-1}, comparable to parameter values used in microplastics modelling studies (2.8×10^{-5} d^{-1}, [56]; 2.7×10^{-4} d^{-1}, [57]). The resulting sediment concentrations depend strongly on the sediment degradation rate of TRWPs. A field TRWP degradation rate in soils was derived from a study published in 1980 [45], reduced by a factor of 10 to account for conditions in aquatic sediments [58]. This is uncertain because of the extrapolation and because the 1980 study is probably too old to be representative for modern tires. It is noted that the uncertain degradation rate does not affect the simulated mass balances, only the derived sediment concentrations. The sediment concentrations derived from simulation results are equilibrium values assuming unchanged releases of TRWPs, land-use, hydrology and stormwater management infrastructure during the period to reach this equilibrium (50–60 years). This assumption is probably incorrect, which could explain why the model has a tendency to overestimate sediment concentrations.

4.3. Use and Limitations of the Modelling Approach

The present model targets the watershed scale across the globe and aims to obtain homogeneous and comparable results. It therefore relies on global input data. This automatically implies that it is less suited for detailed local applications. Input data resolve the differences between urban and rural areas and separate between highways and other roads. The spatial distribution algorithm of TRWP releases exploits these differences as a proxy for traffic intensity. The stormwater management is also parameterized at this level of detail (urban vs. rural and highways vs. other roads). Consequently, the impact of stormwater management on watershed scale TRWP balances can be expected to be reasonably well represented. Halama et al. [59] present a complementary model approach that exploits an ultra-fine grid (5 m vs. 750–850 m in present study) as well as detailed traffic and infrastructure information to conduct a local assessment aimed at better understanding of mechanisms and pathways of TRWPs and associated chemicals and to optimize smart green infrastructure placement in a 5.5 km^2 study area in the city of Seattle.

Simulating the trapping of particles on the watershed scale while using just a hydrology model is challenging, because the river network wet cross sections and the associated hydrodynamic shear stresses cannot be defined in sufficient detail to fully benefit from state-of-the-art formulations for settling, deposition and resuspension [60] (SI-6.3). This can be avoided in local assessments by using a hydrodynamic model approach (as applied for micro-plastics [57]).

The size fraction below 10 µm of TRWPs, supposed to remain in the atmosphere after generation, was set to 2%. Recent literature reports values up to 10% [6]. While this represents a significant uncertainty for the atmosphere endpoint, it is a marginal uncertainty for the terrestrial and aquatic endpoints (at most a reduction of 8%) and was not further assessed in view of many larger uncertainties.

It is further noted that the applicability domain of the model excludes the estuarine and coastal compartments.

The use of the model was demonstrated by a scenario analysis targeting the mitigation potential of stormwater management infrastructure. One scenario aimed at exploiting combined sewer systems and existing WWTPs, while the other aimed at exploiting separated sewer systems and green infrastructure. The results indicated a high potential for the combined sewer optimization if the frequency of combined sewer overflow events (CSOs) can be reduced. The results were less promising for the separated sewers optimization in the very densely populated Yodo-Biwa watershed, mostly because of the presumed difficulty of implementing such systems in the city centres. Although effective in mitigation, the engineering of the capture system design, as well as many more aspects, such as the maintenance of such systems, should be carefully evaluated when considering their use as a management option [7].

4.4. Research Needs and Model Extensions

The results of our study suggest some priority research needs. Refined and contemporaneously representative TRWP generation emission factors would benefit the mass balances precision. These are expected to become available soon, as a result of a standardized tire test method [61]. Experimental studies to compile field balances for a given road section as a function of weather conditions are very rare. Speculating that sampling and analysis of TRWPs will become easier with increased capacity and harmonization, such studies are anticipated to appear. The current model proved sensitive to the assumed TRWP size distribution and settling velocity estimates. TRWP size distributions have been extensively investigated [5,10,48], whereas settling velocity has only more recently been studied for limited particle types [62]. Settling velocities were determined for TRWPs collected from tunnel dust pressure washed from the driving lane and sidewalk, as well as TRWPs generated using an interior drum road simulator with high efficiency vacuum [62]. Values substantially smaller than those used here were noted. As the representativeness of these velocities to TRWPs exposed to UV and other environmental weathering processes and hetero-aggregation is unknown, it remains a priority to collect laboratory data on settling velocities for particles representative of those suspended in the water column. Interestingly, simulated Seine 10–90 percentile water concentrations represented most of the field measurements collected in the 2021 well (Figure 5). This agreement suggests that deposition has not been overstated in the present simulations. With respect to the soils and aquatic sediments endpoints, there is an urgent need for studies to quantify the degradation of TRWPs in these compartments. A recent 2-year lab experiment on degradation of cryo-milled tire tread (CMTT) and TRWPs in water and in soil showed that "the half-life of TRWP and CMTT appears to be much longer than two years" [63]. This study did not lead to a quantified half-life in these compartments, however. Overall, the quantification of the fate and transport of tire wear particles remains challenging because of the spatial and temporal variability and the scarcity of field data, due to a lack of affordable and reliable standardized methods for the collection and analysis of environmental samples [7]. The introduction of TRWP analyses in regular surface water monitoring will require lower cost and simpler analysis methods than currently available.

5. Conclusions and Outlook

This work provides a TRWP terrestrial and aquatic mass balance modelling methodology appliable across the globe, building on datasets with global coverage. It consistently accounts for socio-economic, climatic, and geographic factors and stormwater management gradients. Its demonstrative applications systematically revealed for the first time the strong climate effect on the fate and transport of TRWPs. Indicative stormwater management scenarios demonstrated the sensitivity of TRWP fate and transport to stormwater management practices as well as the potential use of the model to explore management alternatives at the watershed scale.

The present results could be explored and extended in different ways. The spatial distribution method for releases of TRWPs could be coupled to atmospheric fate and transport models. The approach could be extended with transport and fate modelling of tire-associated chemicals to quantity riverine loads, sediment deposition and export to estuaries of such chemicals and their transformation products. This would complement existing local scale assessments [59] with watershed-, country- or even continental-scale assessments.

Supplementary Materials: The following supporting information can be downloaded at https://www.mdpi.com/article/10.3390/w18050562/s1, Sections SI-1 to SI-11 including tables, figures and additional references [64–73].

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Abbreviations

The following abbreviations are used in this manuscript:

CMTT	cryo-milled tire tread, a proxy for TRWP created in a laboratory
CS	combined sewer systems, a scenario aiming at such systems
CSO	combined sewer overflow event
EF	emission factor, referring to the abrasion of car tires per kilometre driven
OM10	particulate matter with a diameter below 10 µm, referring to material suspended in air
SI	supporting information
SS	separated stormwater collection systems, a scenario aiming at such systems
SPM	suspended particulate matter
TOC	total organic carbon
TRWP	tire and road wear particles
WWTP	wastewater treatment plant

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