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Modification of GLEAMS for modeling movement of organic contaminants from land-applied biosolids

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ABSTRACT

Municipal biosolids are commonly applied to agricultural lands as fertilizer, but this also poses potential risks to groundwater and surface water quality from constituents that may be mobilized during storm events. In the present study, an existing model, Groundwater Loading Effects of Agricultural Management Systems (GLEAMS), is modified to predict the fate and transport of organic contaminants from land-applied biosolids, primarily via addition of a labile biosolids organic carbon phase distinct from soil organic carbon. While capable of simulating contaminant transport in runoff and via percolation, only the runoff portion of the model was able to be calibrated using existing experimental data, and showed good agreement with field runoff data for acetaminophen, ibuprofen, triclosan, triclocarban, and estrone, but substantially under-predicted concentrations for carbamazepine, androstenedione, and progesterone. The model is applied to various scenarios using varied chemical properties, application date in the arid west, and application method (i.e., surface spreading vs. incorporation). Chemicals with longer half-lives and lower K_{OCs} exhibited higher losses in runoff than chemicals with shorter half-lives and higher K_{OCs} . For short half-life chemicals (i.e., ≤ 100 days), application at the beginning of the dry season resulted in the lowest losses. However, for long half-life chemicals (~ 1000 days) with high K_{OC} (10,000–100,000), application during the rainy season resulted in the lowest losses, because this caused organic carbon to be high during the period of highest runoff. While further work is necessary to calibrate the percolation and subsurface transport portion, the model can help predict environmental risk from land-application of biosolids, highlight gaps in our knowledge about how chemicals are mobilized and transported from biosolids, and help identify management practices that result in minimal impacts to water quality.

1. Introduction

In the United States, biosolids that are generated during municipal wastewater treatment are commonly used as soil amendment or fertilizer (Lu et al., 2012; U.S. Environmental Protection Agency Municipal and Industrial Solid Waste Division Office of Solid Waste, 1999). Biosolids contain nutrients and organic matter that can be a valuable resource to agricultural sites, but the beneficial reuse of biosolids must be evaluated relative to potential risks, which include impacts to water quality due to mobilization of trace organic compounds.

Several studies have measured the mobilization of trace organic compounds from land-applied municipal biosolids. These studies included analysis in tile drainage (Edwards et al., 2009; Lapen et al., 2008) and surface runoff (Giudice and Young, 2011; Sabourin et al., 2009; Topp et al., 2008), and of hormones in surface runoff (Yang et al.,

2012) following application. Concentrations found in tile drainage (Edwards et al., 2009) and surface runoff (Giudice and Young, 2011) were generally of the same magnitude or lower than those found in treated wastewater effluent, but some were still comparable or above the lowest known environmental endpoints, which were generally sublethal developmental or reproductive effects of chronic exposure to low levels (Edwards et al., 2009; Giudice and Young, 2011).

Some researchers have developed or applied models to examine losses due to subsurface transport from liquid municipal biosolids (Akhand et al., 2008; Larsbo et al., 2009). Models have also been developed for transport of nutrients from land-applied manure (Nelson and Parsons, 2006; Wang et al., 1996). A fugacity-based model has been developed for land-applied biosolids that does not include transport via rainfall runoff (Hughes et al., 2008). To our knowledge, no models have been developed that allow for prediction of chemical loss via both

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surface runoff and subsurface transport, and that also allow the user to examine effects of agricultural management practices (such as crop rotation, erosion control measures, tillage management, etc.) on chemical fate and transport from land-applied biosolids.

The Groundwater Loading Effects of Agricultural Management Systems (GLEAMS) model has been used previously to model pesticide losses from agricultural fields (Leonard et al., 1992, 1995). The model contains a pesticide fate and transport module that predicts movement of agriculturally applied pesticides in surface runoff and the subsurface. Research has indicated that mobilization of many (though not all) trace organic constituents from biosolids-amended soil is mechanistically similar to mobilization of pesticides from soil—specifically, that hydrophobic partitioning to organic carbon is the dominant sorption/desorption mechanism, and that it can be described via linear sorption isotherms and the organic carbon normalized partitioning coefficient K_{OC} (Lee et al., 2003; Massoudieh et al., 2005; Wauchope et al., 2002; Wu et al., 2009). With this recognition, in this study, GLEAMS is modified to allow the user to input biosolids application parameters, and then predicts the fate and transport of target chemicals from the field.

GLEAMS was developed in the 1980s and contains hydrology, erosion, pesticide, and nutrient submodels. The model was developed to evaluate complex interactions among soils, pesticide chemistry, climate, and management practices that affect chemical movement in runoff and through the root-zone (Leonard et al., 1987; Truman et al., 1998). The model can be used for plot or field sized units, in which soil, management, and areal precipitation are uniform. Soil properties vary with depth, and therefore, parameter values are required for each horizon. Computational layers are used to track and route water and chemicals. The surface layer is assumed to be a fixed thickness of 1 cm, even though it is known that factors such as tillage, time since last tillage, rainfall, soil texture, soil water content, and soil cover, among other things, affect infiltration control and interaction of runoff and chemical extraction (Leonard et al., 1987).

The model runs on a daily time-step, and daily climactic data are used to calculate the water balance. Runoff and infiltration due to precipitation are determined using the SCS curve-number method (Knisel, 1980). A storage-routing technique is used to simulate distribution of water and percolation in the subsurface (Knisel, 1993). Evapotranspiration is estimated using the Penman-Monteith method (Knisel, 1993). The erosion submodel uses a modified Universal Soil Loss Equation to simulate storm-by-storm rill and interrill erosion (Knisel, 1980, 1993). Sediment in runoff is affected by particle size, and thus sediment enrichment ratios are used in simulating adsorbed pesticide transport. In essence, coarser soil material is deposited or left in place, so the transported sediment has a higher per unit mass adsorptive capacity and adsorbed chemical concentration than that of the whole soil. The enrichment ratio is calculated in the erosion submodel based on the specific surface area of the sediment leaving the field and the specific surface area of the whole soil matrix (Knisel, 1980; Leonard et al., 1987).

The pesticide submodel tracks chemical movement in runoff and sediment, as well as in the subsurface. Degradation, extraction into runoff, and movement of chemical in the subsurface are described in more detail in the following section, which describes model modifications. Pesticide applications can be by surface application, incorporation, or injection. Complete descriptions of the model can be found in the model and related documentation (Knisel, 1980, 1993; Leonard et al., 1987).

The specific objectives of this study were to: (1) modify an existing chemical fate and transport model that incorporates agricultural management practices, and calibrate this model using existing studies in the literature, to enable simulation of chemical losses from land-applied municipal biosolids, and (2) apply the model to various hypothetical scenarios to investigate and make general observations about the effects of different factors and management decisions on chemical losses from

land-applied biosolids. As will be shown later on, while the model is capable of simulating both surface and subsurface transport, only the surface (runoff) portion of the model was able to be calibrated based on the limited data available from existing studies on subsurface transport of trace organic chemicals from biosolids. Nevertheless, results from this study can help predict environmental risk from land-application of municipal biosolids, highlight gaps in our knowledge about how chemicals are mobilized and transported from biosolids, and can help identify management practices that result in minimal impacts to water quality.

2. Materials and methods

2.1. Model development

Two modifications to GLEAMS were made: addition of a biosolids phase (to soil and water phases already present), and degradation of organic carbon in the biosolids phase. Instead of exclusively simulating pesticide fate and transport, the modified model simulates the fate and transport of any organic constituent present in the biosolids whose sorption can be described using linear partitioning to organic matter and whose degradation follows first order decay. Applications of the chemical to the system via surface application, incorporation, and injection can now occur as part of the application of the biosolids that contain the chemical. Fig. 1 includes a simplified diagram of surface and subsurface layers and phases as well as the various transport, transformation, and loss mechanisms included.

In the model, equilibrium between the three phases is established in each computational layer every time rainfall occurs. This means that there are times when the three phases are not at equilibrium (i.e., when conditions affecting equilibrium have changed but rainfall has not yet occurred). The most important factor in the modified model that causes this is the organic carbon degradation in the biosolids. There are therefore parameters, such as soil and biosolids concentrations of chemical, which the user can view as output on a daily time-step, but which may not be at equilibrium. However, since the primary objective of the model is to estimate losses due to rainfall, establishment of equilibrium only on days when rainfall occurs is considered an appropriate simplification.

2.1.1. Addition of biosolids phase

Degradation of chemical in soil and biosolids is assumed to follow a first-order relationship, and is defined in terms of the half-life,

$$C_{s(t+\Delta t)} = C_{s(t)} \cdot \exp(-0.693 \cdot \Delta t / S_{1/2s}) \quad (1)$$

$$C_{b(t+\Delta t)} = C_{b(t)} \cdot \exp(-0.693 \cdot \Delta t / S_{1/2b}) \quad (2)$$

where $C_{s(t+\Delta t)}$ = concentration in soil at time $t + \Delta t$ (mg/kg); $C_{s(t)}$ = concentration in soil at time t (mg/kg); Δt = time interval between computation (d); $S_{1/2s}$ = half-life in soil phase (d); $S_{1/2b}$ = half-life in biosolids phase (d); $C_{b(t+\Delta t)}$ = concentration in biosolids at time $t + \Delta t$ (mg/kg); and $C_{b(t)}$ = concentration in biosolids at time t (mg/kg).

Chemical distribution between the solution phase and the soil phase, and between the solution phase and the biosolids phase, is described as a simple linear sorption isotherm,

$$K_{ds} = C_s / C_w \quad (3)$$

$$K_{db} = C_b / C_w \quad (4)$$

where, at equilibrium, K_{ds} = soil-water distribution coefficient (L/kg); C_s = concentration in soil (mg/kg); C_w = concentration in water (mg/L); K_{db} = biosolids-water distribution coefficient (L/kg); and C_b = concentration in biosolids (mg/kg). Because the distribution coefficients are dependent on the organic carbon content of the soil or biosolids at a given time, they are defined in terms of the organic carbon normalized distribution coefficients,

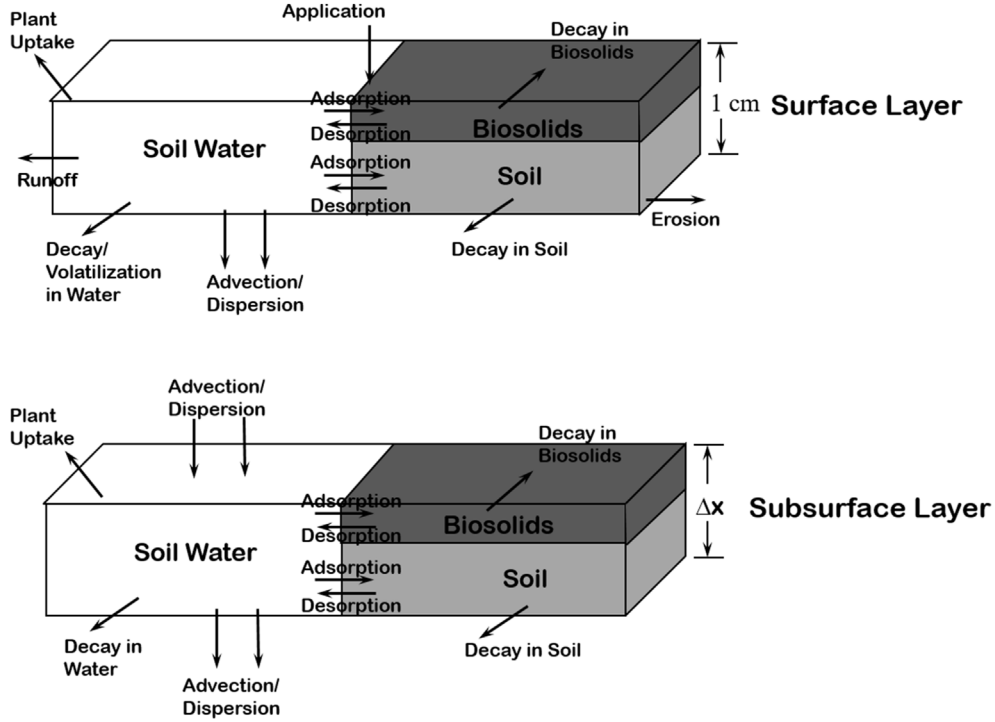


Fig. 1. Schematic showing surface layer and subsurface layer phases, transport, transformation, and loss mechanisms, and equilibrium relationships simulated in the developed model.

$$K_{ds} = K_{OCs} \cdot OC_s / 100 \quad (5)$$

$$K_{db} = K_{OCb} \cdot OC_b / 100 \quad (6)$$

where K_{OCs} = organic carbon normalized soil-water partitioning coefficient (L/kg); OC_s = % organic carbon in soil; K_{OCb} = organic carbon normalized biosolids-water partitioning coefficient (L/kg); and OC_b = % organic carbon in biosolids. Studies have shown that K_{OCs} and K_{OCb} are not always equal to each other—either because soil or biosolids components other than organic carbon participate significantly in the sorption process or because the different composition of the two phases cause variation in their sorptive efficiency (Agyin-Birikorang et al., 2010; Ogunyoku, 2011; Stumpe and Marschner, 2007; Wu et al., 2009). The model allows for users to input different values for the K_{OCs} and K_{OCb} .

At saturation, the volume of water per unit volume of the soil-biosolids matrix is

$$V_{fw} = POR \quad (7)$$

where V_{fw} = volume of water per unit volume of saturated soil-biosolids mixture (L) and POR = porosity. We can then define the volumes of soil and biosolids per unit volume of saturated soil-biosolids mixture (V_{fs} and V_{fb} , respectively, in L) as:

$$V_{fs} = 1 - POR - M_b / \rho_b \quad (8)$$

$$V_{fb} = 1 - POR - M_s / \rho_s \quad (9)$$

where ρ_s = density of soil (kg/L); and ρ_b = density of biosolids (kg/L). The mass of biosolids in each layer is determined according to the application rate and method of application (surface spreading, incorporation, or injection). The mass of soil in each layer is determined according to the bulk density of the soil. The approach described above assumes that porosity is unaffected by biosolids addition.

2.1.2. Chemical losses and movement during rainfall

In this section, three processes are described. First, loss of chemical from the surface layer due to infiltration into the layer beneath is determined. Second, loss of chemical due to extraction from the remaining

mass of chemical in the surface layer into overland flow is determined. Last, routing of chemical through subsurface layers is described.

At saturation, the chemical mass in the surface layer per unit volume of the soil-biosolids mixture (z , in mg) is

$$z = C_w \cdot V_{fw} + C_s \cdot V_{fs} \cdot \rho_s + C_b \cdot V_{fb} \cdot \rho_b \quad (10)$$

Now, combining Eq. (10) with Eqs. (3), (4) and (7),

$$z = C_w \cdot POR + K_{ds} \cdot C_w \cdot V_{fs} \cdot \rho_s + K_{db} \cdot C_w \cdot V_{fb} \cdot \rho_b \quad (11)$$

The rate of change of chemical mass in the surface layer due to infiltration during a storm is

$$-dz = C_w \cdot f \cdot dT \quad (12)$$

where f = water flux (L/h); and T = time (storm duration) (h). Rearranging Eq. (11) gives

$$C_w = z / (POR + K_{ds} \cdot V_{fs} \cdot \rho_s + K_{db} \cdot V_{fb} \cdot \rho_b) \quad (13)$$

Combining Eqs. (12) and (13), we then integrate from z_0 to z and from T_0 to T :

$$\int_{z_0}^z -dz = \int_{T_0}^T \frac{z \cdot f \cdot dT}{POR + K_{ds} \cdot V_{fs} \cdot \rho_s + K_{db} \cdot V_{fb} \cdot \rho_b} \quad (14)$$

where z_0 = concentration of chemical in surface layer at the beginning of the storm per unit volume of soil-biosolids mixture in the surface layer (mg). This yields:

$$z = z_0 \cdot \exp\left(\frac{-f \cdot T}{POR + K_{ds} \cdot V_{fs} \cdot \rho_s + K_{db} \cdot V_{fb} \cdot \rho_b}\right) \quad (15)$$

The infiltration flux through the top layer of soil (f) is

$$f = (P - Q - AWS) / T \quad (16)$$

where P = rainfall depth (cm); Q = surface runoff depth (cm); and AWS = soil water storage capacity to saturation (initial abstraction) (cm). Combining Eqs. (15) and (16) gives:

$$z = z_0 \cdot \exp\left(\frac{-(P - Q - AWS)}{POR + K_{ds} \cdot V_{fs} \cdot \rho_s + K_{db} \cdot V_{fb} \cdot \rho_b}\right) \quad (17)$$

The parameter z_0 , the mass of chemical in the surface layer at the beginning of the storm per unit volume of soil-biosolids mixture in the surface layer, is calculated as:

$$z_0 = \frac{C_s(t) \cdot M_s + C_b(t) \cdot M_b}{M_s + M_b} \cdot (V_{fs} \cdot \rho_s + V_{fb} \cdot \rho_b) \quad (18)$$

Thus, the total mass of chemical lost via infiltration from the surface layer (PERCM₁, in mg) is:

$$PERCM_1 = z - z_0 \quad (19)$$

In the model, at the time runoff occurs, the surface layer of soil contains the chemical mass determined after losses due to infiltration (vertical translocation), which are described above. The concentration of chemical available to runoff in this model is defined as the mass of chemical in the surface layer per unit volume of soil-biosolids mixture in the surface layer divided by the weighted average bulk density of the mixture,

$$C_{av(mix)} = z / (V_{fs} \cdot \rho_s + V_{fb} \cdot \rho_b) \quad (20)$$

where $C_{av(mix)}$ = concentration of chemical in surface layer of soil-biosolids mixture available to runoff (mg/kg). However, at the interface between the soil/biosolids matrix and the overland flow, only some portion of the soil-biosolids mixture is effective in supplying chemical to the flow. Thus, we introduce the term $B_{av(mix)}$, the soil-biosolids mixture mass available to supplying chemical to the overland flow per unit volume of overland flow (extraction coefficient; kg). The mass of chemical in the soil-biosolids mixture available to the overland flow is assumed to equilibrate instantly between the soil/biosolids mixture and the overland flow, so

$$C_{av(mix)} \cdot B_{av(mix)} = C_w \cdot V + C_s \cdot B_s + C_b \cdot B_b \quad (21)$$

where V = volume of water per unit volume of runoff interface (L), and B_s and B_b are the soil and biosolids (respectively) masses available to supplying chemical to the overland flow per unit volume of overland flow (kg). B_s and B_b represent a portion of (and thus must be less than) the total mass of soil and biosolids, respectively.

In Eq. (21), we can disregard the volume of the runoff interface occupied by soil compared to the much larger volume of water, so that $V = 1$. Combining Eqs. (3) and (4) with Eq. (21), we derive the expressions for the equilibrium concentrations in the overland flow, soil, and biosolids:

$$C_w = \frac{B_{av(mix)} \cdot C_{av(mix)}}{1 + B_s \cdot K_{ds} + B_b \cdot K_{db}} \quad (22)$$

$$C_s = \frac{B_{av(mix)} \cdot C_{av(mix)}}{(1 + K_{db} \cdot B_b) / K_{ds} + B_s} \quad (23)$$

$$C_b = \frac{B_{av(mix)} \cdot C_{av(mix)}}{(1 + K_{ds} \cdot B_s) / K_{db} + B_b} \quad (24)$$

The total mass of aqueous chemical lost via runoff from the surface layer (QM, in mg) is:

$$QM = Q \cdot C_w \quad (25)$$

where Q = surface runoff (L). The total mass of chemical sorbed to sediment lost via runoff from the surface layer (SM, in mg) is:

$$SM = S \cdot E \cdot C_s \quad (26)$$

where S = sediment loss (kg, calculated in the erosion component); and E = enrichment ratio (calculated in the erosion component, as described previously).

As noted in (Leonard et al., 1987), a functional relationship developed in the original GLEAMS relates B_s to K_{ds} as follows:

$$B_s = \begin{cases} 0.5, & K_{ds} \leq 1.0 \\ 0.7 - 0.2 \cdot K_{ds}, & 1.0 < K_{ds} \leq 3.0 \\ 0.1, & K_{ds} > 3.0 \end{cases} \quad (27)$$

Little is presented in (Leonard et al., 1987) regarding the development of these rules. In the present work, the rules for B_s were modified based on the calibration performed, which is discussed in a subsequent section. Although no previous work has been done to determine the value of B_b , it will be shown later that acceptable (i.e., within a factor of 2–5) results are obtained by equating the value of B_b to the entire mass of biosolids per unit volume of overland flow (i.e., all of the biosolids phase present in the 1 cm surface layer is available to supplying chemical to the overland flow), and thus $B_b = M_b$. The value of $B_{av(mix)}$ is calculated as the average of B_s and B_b , weighted by the K_d of each compartment,

$$B_{av(mix)} = \frac{K_{ds} \cdot B_s + K_{db} \cdot B_b}{K_{ds} + K_{db}} \quad (28)$$

The mass of chemical that infiltrates from the surface layer into the layer below was defined in Eq. (17). The mass of chemical that infiltrates from a given subsurface layer into the layer below it is:

$$PERCM_i = PERC_i \cdot C_{wi} \quad (29)$$

where $PERCM_i$ = mass of chemical in percolate from subsurface layer i (mg); $PERC_i$ = volume of water percolated from subsurface layer i (L); and C_{wi} = concentration of chemical in water in subsurface layer i (mg/L). This mass is added to any existing mass in the layer below, and the total mass of chemical in each layer is computed in this fashion. The total mass of chemical in any subsurface layer, I (PMS _{i} , in mg), is divided between the three phases:

$$PMS_i = C_{bi} \cdot M_{bi} + C_{si} \cdot M_{si} + C_{wi} \cdot V_{wi} \quad (30)$$

where C_{bi} = concentration of chemical in biosolids in subsurface layer i (mg/kg); M_{bi} = mass of biosolids in subsurface layer i (kg); C_{si} = concentration of chemical in soil in subsurface layer i (mg/kg); M_{si} = mass of soil in subsurface layer i (kg); C_{wi} = concentration of chemical in water in subsurface layer i (mg/L); and V_{wi} = volume of water in subsurface layer i (L). Assuming equilibrium, substituting Eqs. (4) and (5) into Eq. (30) gives

$$PMS_i = K_{db} / K_{ds} \cdot C_{si} \cdot M_{bi} + C_{si} \cdot M_{si} + C_{si} / K_{ds} \cdot V_{wi} \quad (31)$$

$$C_{si} = \frac{PMS_i}{K_{db} / K_{ds} \cdot M_{bi} + M_{si} + V_{wi} / K_{ds}} \quad (32)$$

And, again, at equilibrium,

$$C_{bi} = K_{db} / K_{ds} \cdot C_{si} \quad (33)$$

$$C_{wi} = C_{si} / K_{ds} \quad (34)$$

2.1.3. Degradation of organic carbon in biosolids phase

Mineralization of organic carbon in biosolids is treated in the model as a first order process acting on two compartments: fast-degrading organic carbon and slow-degrading organic carbon. A third compartment, recalcitrant organic carbon, does not degrade. The biosolids-borne organic carbon remaining at time t is thus the sum of these three components, so,

$$MOC_b(t) = MOC_{b,f}(t) + MOC_{b,s}(t) + MOC_{b,r}(t) \quad (35)$$

$$1 = fOC_{b,f}(t) + fOC_{b,s}(t) + fOC_{b,r}(t) \quad (36)$$

where $MOC_b(t)$ = mass of organic carbon in biosolids at time t (kg); $MOC_{b,f}(t)$ = mass of fast-degrading organic carbon in biosolids at time t (kg); $MOC_{b,s}(t)$ = mass of slow-degrading organic carbon in biosolids at time t (kg); $MOC_{b,r}(t)$ = mass of recalcitrant organic carbon in biosolids at time t (kg); $fOC_{b,f}(t)$ = fraction of organic carbon that is fast-degrading at time t ; $fOC_{b,s}(t)$ = fraction of organic carbon that is slow-degrading at time t ; and $fOC_{b,r}(t)$ = fraction of organic carbon that is

recalcitrant at time t . The equations for calculating mass in each fraction at any time are,

$$MOC_{b,f}(t + \Delta t) = MOC_{b,f}(t) \cdot \exp(-k_{b,f} \cdot \Delta t) \quad (37)$$

$$MOC_{b,s}(t + \Delta t) = MOC_{b,s}(t) \cdot \exp(-k_{b,s} \cdot \Delta t) \quad (38)$$

$$MOC_{b,r}(t) = MOC_{b,r}(0) \quad (39)$$

where $k_{b,f}$ = rate constant for degradation of fast-degrading organic carbon (d^{-1}); and $k_{b,s}$ = rate constant for degradation of slow-degrading organic carbon (d^{-1}).

Since biosolids are tracked as a distinct phase, to keep the mass of biosolids in each layer from growing unrealistically large in perennial application scenarios, immediately prior to the second and subsequent perennial biosolids applications, the mass of biosolids is set to 0, and the masses of organic carbon and of all chemicals in the remaining biosolids phase are added to the soil phase. This assumption is meant to approximate the mineralization of biosolids that occurs over time, in which biosolids as a unique phase separate from the soil cease to be recognizable due to natural phenomena. Over both short-term and long-term simulations with a single or with perennial applications, such as those employed in the present study, these assumptions are reasonable approximations. For scenarios that involve multiple applications per year, more sophisticated approaches may be more appropriate.

2.2. Calibration

Three case studies were selected to use for calibrating the runoff portion of the model (Giudice and Young, 2011; Sabourin et al., 2009; Yang et al., 2012). All three studies applied dewatered municipal biosolids to small (2–6 m^2) plots and used rainfall simulators to simulate intense (> 60 mm/h) rainfall events periodically over the course of 24–36 days following biosolids application. From the chemicals analyzed in these studies, a subset of chemicals were selected to use for calibrating the model. The chemicals selected met three criteria: 1) they were quantified in biosolids and runoff from rainfall simulations, 2) their sorption could be described by linear isotherms to organic carbon, and 3) data was available in the literature regarding their expected half-life and K_{OC} . These requirements were the minimum requirements necessary to run the model and compare the results to experimentally measured runoff concentrations. The chosen chemicals were acetaminophen, carbamazepine, ibuprofen, triclocarban, triclosan, androstenedione, estrone, and progesterone. Supplemental Table S1 summarizes chemicals analyzed in the studies, whether they were included in the present study for calibration, and the reasons for exclusion. Supplemental Tables S2–5 summarize important parameters used in the initial model runs to replicate the scenarios described in these studies. For parameters that were not measured or reported in the studies, estimates or assumptions were made, as necessary, or other data sources were consulted.

There were two phases to the calibration process. The first phase was to develop a set of relationships for B_s that would result in reasonable agreement (i.e., within an order of magnitude) between predicted and measured runoff concentrations for as many chemicals as possible. As explained further in the results section, part of the goal in this phase was to follow the approach from the original GLEAMS as much as possible, and modify that approach only as necessary to account for differences in mobilization from the biosolids-soil mixture vs. just soil.

The second phase was to modify chemical and scenario-specific parameters within justifiable ranges to obtain acceptable (i.e., within a factor of 2–5) agreement between model results and experimental data. Because we assume that all measurements made in the experimental studies, as well as the model itself, are to a certain extent in error, the goal of the calibration process was not to find an optimal and unique parameter set for the model application. In fact, many model parameter

combinations show essentially equivalent agreement with measured data. Instead, the goal of this and any calibration process is to find a parameter set that gives acceptable model results (Beven and Binley, 1992). This was done via a trial-and-error process, using a combination of literature-derived values for parameters of interest and using judgment as to what parameter estimates were realistic given our understanding of the physical experimental setups. Changing K_{OC} and half-life resulted in chemical-specific changes, while changing other parameters affected all chemicals simultaneously (biosolids OC %, POR, rate constants for fast and slow degrading biosolids). Also, changing certain parameters affected results for all storm simulations (K_{OC} , biosolids OC %, POR) while others primarily affected later storms ($t_{1/2}$, rate constants for fast and slow degrading biosolids). Again, within this step, the goal was to achieve acceptable agreement when using values of K_{OC} and half-life either found in the literature or justified based on the range of possible values found in the literature, and not necessarily to optimize the model fit by computationally selecting the best possible value for each parameter.

Runoff volume and erosion mass were not calibrated for the simulations. Previous work has shown that GLEAMS produces adequate predictions of long-term runoff and erosion, though short-term (< 1 year, or by storm) estimates may be quite variable (Knisel et al., 1991). It was for this reason that runoff concentrations, which were relatively insensitive to runoff volume, were used for calibration, and not mass of chemical. However, application of the model on a long-term basis and evaluation of chemical mass losses should still be valid, based on this and previous work.

Sediment concentrations were not formally calibrated, as insufficient data was available in the literature to determine accurate runoff sediment concentrations. However, what information was available was examined to determine whether model estimates of sediment-bound losses were reasonable.

Concentration in rainfall that infiltrated (percolate) were also not able to be calibrated due to limited data from existing literature studies. From the three studies used in runoff concentration calibration, subsurface concentrations were only measured in (Giudice and Young, 2011), and only detected for a single chemical. Moreover, measurements in (Giudice and Young, 2011) may have been affected by edge effects and soil cracking which cannot be accounted for in the model.

2.3. Application

Several theoretical scenarios were modeled to examine the effects of K_{OC} and half-life, to examine the effect of application date for an annual application scenario in the arid west, and to examine differences in chemical loss under surface spreading vs. incorporation application methods on long-term chemical losses. Twelve generic chemicals, each with a different combination of half-life (10, 100, and 1000 days) and Log K_{OC} (2, 3, 4, and 5) were simulated. Their names in this report are denoted by HL[*half-life, in days*]LKOC[*Log KOC*], where [*half-life*] and [*Log KOC*] are the generic chemical's half-life and Log K_{OC} . All scenarios used the soil and erosion parameters that were used in the Giudice and Young (2011) calibration model runs (Supplemental Tables 2–3). Precipitation data was obtained from the California Irrigation Management Information System database (CIMIS, (California Department of Water Resources, 2012)) for the 25-year period January 1, 1987 through December 31, 2011. Annual biosolids application was either on January 2, May 1, or September 1. All chemicals were at equivalent concentrations of 10 mg/kg in the applied biosolids. Biosolids were applied at a rate of 10,000 kg/ha, and either incorporated to a depth of 11.4 cm, or surface-applied.

3. Results and discussion

3.1. Calibration

As mentioned above, the first phase of calibration was to develop relationships for the parameter B_s that would result in generally reasonable agreement (i.e., within an order of magnitude) with runoff concentration data for as many chemicals as possible. It is known that transfer of chemical from the soil/soil-pore water in the upper layer of soil to overland flow is dependent on chemical properties such as K_d and molecular diffusivity, and rainfall/runoff characteristics such as rainfall intensity and bottom shear stress, both of which affect turbulent mixing (Massoudieh et al., 2005; Wallach and Vangenuchten, 1990; Yan and Kahawita, 2000). However, the original version of GLEAMS employs a functional simplification of this process (see Eq. (27)). In the present study, the desire was to use as similar an approach to the original version of GLEAMS as possible. For chemicals that are weakly sorbed (i.e., $K_{ds} \leq 3.0$), it was anticipated that the same relationship used in the original GLEAMS would be sufficient, since the differences in sorption between soil and biosolids-amended soil would be minimal for these chemicals. For chemicals that are more strongly sorbed (i.e., $K_{ds} > 3.0$), a new relationship had to be developed in order to obtain reasonable agreement with the experimental results. Fig. 2 shows the equations used to estimate B_s based on the value of K_{ds} in the calibrated model. For the two instances in which $1.0 \leq K_{ds} \leq 3.0$, the relationship from the original GLEAMS was indeed found to be sufficient and thus was maintained. For the instances in which $K_{ds} > 3.0$, a power law relationship was developed that resulted in reasonable agreement with experimental results. Thus, the value of B_s in the model is calculated as follows:

$$B_s = \begin{cases} 0.5, & K_{ds} \leq 1.0 \\ 0.7 - 0.2 \cdot K_{ds}, & 1.0 < K_{ds} \leq 3.0 \\ 0.0914 \times K_{ds}^{-0.274}, & K_{ds} > 3.0 \end{cases} \quad (40)$$

In the original GLEAMS, the value of B_s for $K_{ds} > 3.0$ is set to a constant value of 0.1 (see Eq. (27)), but it makes intuitive sense that as K_{ds} continues to increase, the value of B_s would also continue to decrease. Therefore, both a linear relationship between B_s and K_{ds} and a power law relationship were investigated, and the power law shown in Eq. (40) was found to give superior results.

The value of B_b was always set equal to the entire mass of biosolids per unit volume of overland flow (i.e., all of the biosolids phase is

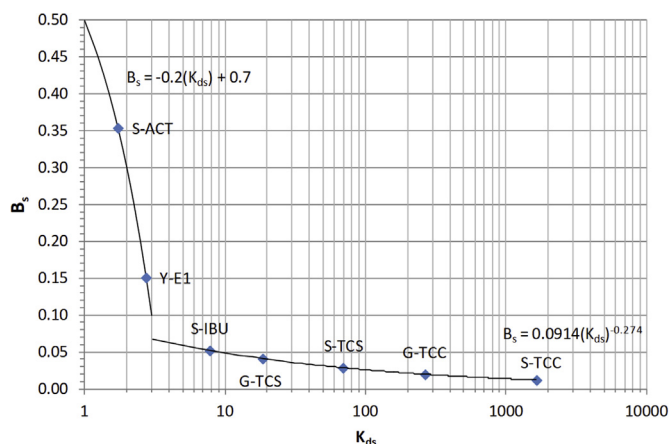


Fig. 2. Relationship developed between B_s (soil mass available to supplying chemical to the overland flow per unit volume of overland flow) and K_{ds} (soil-water distribution coefficient). Data labels denote the calibration scenario (S = Sabourin et al., 2009; G = Giudice and Young, 2011; Y = Yang et al., 2012) and the chemical (ACT = acetaminophen, IBU = ibuprofen, TCC = triclocarban, TCS = triclosan, E1 = estrone.).

available to supplying chemical to the overland flow). Further work is necessary to determine whether this assumption always provides reasonable results, but in the present study, no lesser value of B_b provided better agreement with experimental values.

Results of the calibration for all scenarios are shown in Figs. 3 and 4. After calibration, seven of the ten chemicals showed reasonable agreement with experimental runoff concentrations (Fig. 3), and the remaining three did not (Fig. 4). A linear regression was developed between modeled and measured concentrations in runoff for the seven chemicals for which reasonable agreement could be obtained. This relationship is shown in Fig. 5, and the resulting coefficient of determination (R^2) was 0.9697. A total of 21 of 28 modeled concentrations were within a factor of 2 of the corresponding measured concentrations, and 27 of 28 were within a factor of 5.

Results of the initial simulation for the Sabourin et al. (2009) (Sabourin et al., 2009) experiments showed generally acceptable agreement with experimental results for triclocarban. Adjusting the ibuprofen half-life from 32 days to 50 days and the K_{OC} from 66 (Yamamoto et al., 2009) to 338 (as in (Styszko et al., 2010)) improved agreement for ibuprofen. Adjusting the acetaminophen K_{OC} from 263 (Yamamoto et al., 2009) to 75 improved agreement for acetaminophen (literature values are as low as 18 were found (Martinez-Hernandez et al., 2015)). The improved agreement due to the change in K_{OC} for acetaminophen was due to 2 factors: the direct impact on K_{ds} and K_{db} of a lower K_{OC} in Eqs. (5) and (6), and thus on concentrations in Eqs. (22)–(24); and the impact of a decrease in K_{ds} to less than 3.0 in the surface layer, which increased B_s substantially. For triclosan, the K_{OC} in soil and biosolids had to be adjusted down from its initial estimate of 7946 to 3000, and the half-life from 187 to 15 days, to obtain acceptable agreement. For carbamazepine, results of the initial simulation were far below experimental values, and no reasonable adjustments to model parameters (including B_s) resulted in acceptable agreement.

The primary reason for the discrepancy for carbamazepine is related to the mass of carbamazepine available to runoff in the model. In (Sabourin et al., 2009), it was reported that over the course of the 5 rainfall simulations, approximately 20% of the carbamazepine applied was lost via runoff. Since biosolids were incorporated to a depth of 15 cm, and the surface runoff zone of the model is only the top 1 cm of the soil/biosolids, a maximum of 6.7% of the applied mass of carbamazepine is available for runoff in the model. Therefore, it would be impossible for the model to approach the concentrations in (Sabourin et al., 2009) without either increasing the depth of the computational layer of overland flow or increasing the initial mass of carbamazepine in the applied biosolids. Carbamazepine may also experience colloid facilitated transport (Maskaoui and Zhou, 2010) that limits its retardation in a field setting, resulting in higher concentrations than the model would predict.

Results of the initial simulation for the Giudice and Young (2011) experiments (Giudice and Young, 2011) showed generally acceptable agreement with experimental runoff concentrations for triclocarban and triclosan. In order to achieve better agreement, the triclosan K_{OC} in soil and biosolids was lowered from 7946 to 5000. Additionally, fractions of fast degrading and slow degrading organic carbon in the biosolids were adjusted, and the half-life of fast-degrading organic carbon increased, and these changes improved agreement for both triclocarban and triclosan. A reduction in the literature derived K_{OC} for triclosan in both the Giudice and Young (2011), and the Sabourin et al. (2009) scenarios is justified, since soil and runoff pH in the former was 8.0–8.1 (approximately the pK_a of triclosan) and biosolids pH in the latter was 8.0, while literature K_{OC} s have been derived under more acidic conditions. Wu et al. found that the K_d of the anionic form of triclosan was 0.5–0.66 the K_d of the neutral form in soils amended with biosolids, making the overall K_d at pH 8 between 0.7 and 0.8 the K_d at pH 5 or 6, for which the K_{OC} for triclosan was derived (Wu et al., 2009).

Results of the base simulation for the Yang et al. (2012) experiments (Yang et al., 2012) showed acceptable agreement between modeled

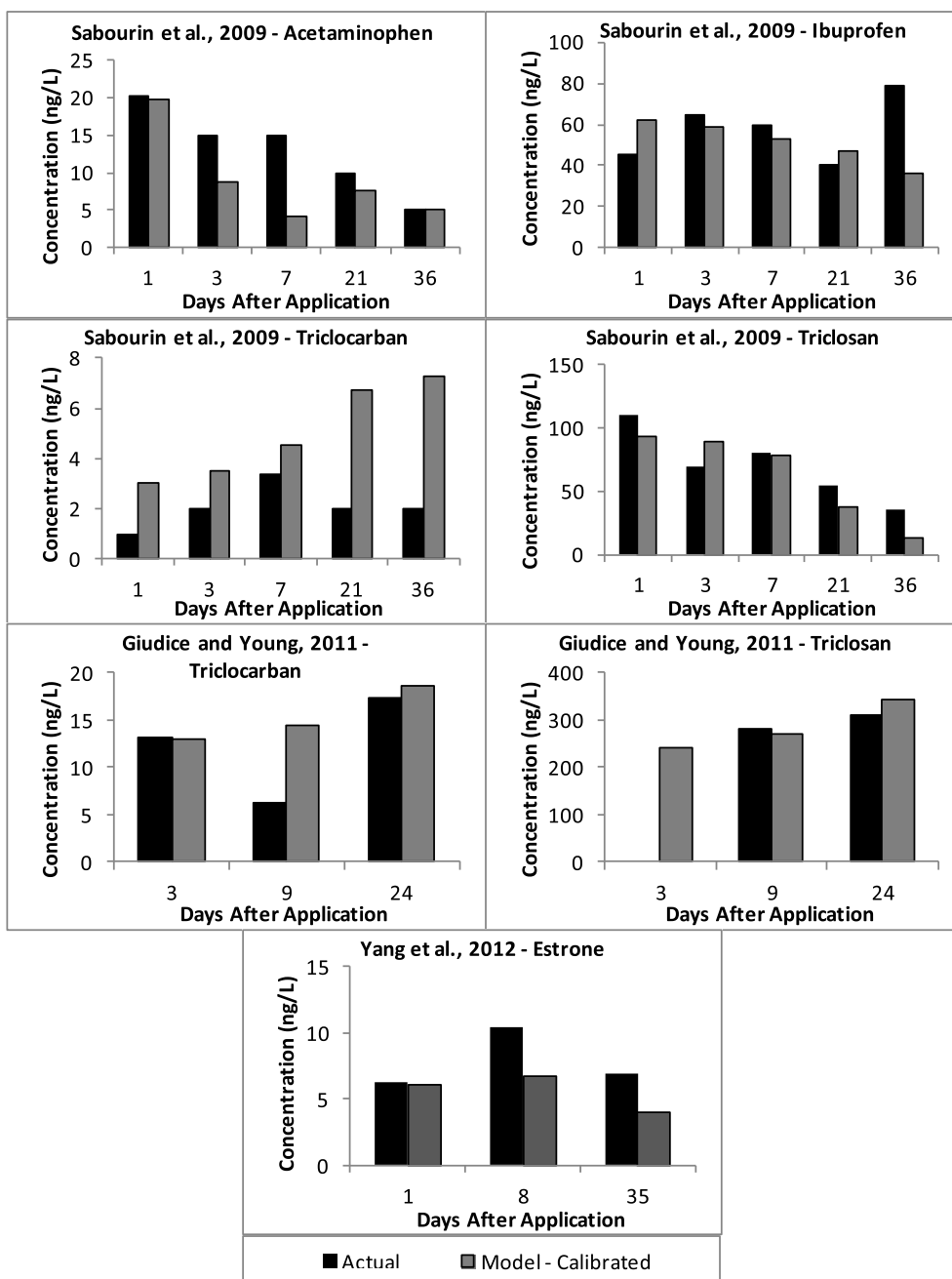


Fig. 3. Results of experiments and calibrated simulations for chemicals for which acceptable agreement was obtained: Sabourin et al. (2009) (acetaminophen, ibuprofen, triclocarban, and triclosan) Giudice and Young (2011) (triclocarban and triclosan), and Yang et al. (2012) (estrone).

results and experimental results for estrone, but modeled concentrations for androstenedione and progesterone were far below measured values. Increasing the half-life for estrone from 2.7 to 20 days improved agreement for the latter storms. No reasonable adjustments of model parameters (including B_s) resulted in acceptable agreement for androstenedione and progesterone. There are several potential reasons for the lack of agreement. First, hormone concentrations in the soil and biosolids may change rapidly on a day-to-day basis as hormones are conjugated or deconjugated, or due to interconversion of hormones (Yang et al., 2012). For example, studies have shown increases in androstenedione and progesterone in manure and manure-soil mixtures as they aged (Mansell et al., 2011; Zheng et al., 2008). Colloid facilitated transport and leaching via preferential subsurface pathways have also been shown to be major transport pathways for hormones (Steiner et al., 2010; Stumpe and Marschner, 2007). Taken together, it is clear

that the assumptions contained within the modified GLEAMS model described herein make simulation of the fate and transport of some hormones inappropriate, and more sophisticated approaches are necessary for these compounds.

While sediment concentrations were not formally calibrated, as insufficient data was available in the literature to determine accurate runoff sediment concentrations, available information was examined to determine whether model estimates of sediment-bound losses were reasonable. Because only mass losses, and not concentrations, were reported in the literature, the mass losses reported were compared to the mass losses modeled. This comparison assumes erosion to be modeled accurately for these experimental sites, but as mentioned previously it is uncertain whether this is the case in short-term model applications such as these. The mass of triclosan and triclocarban lost via the sorbed phase was reported in (Sabourin et al., 2009), and was

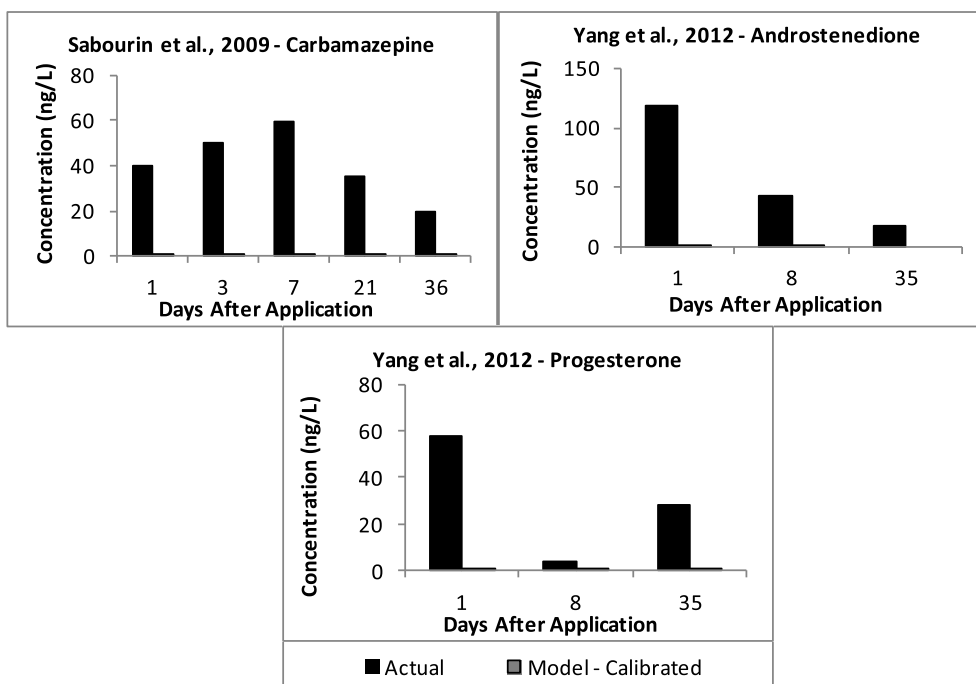


Fig. 4. Results of experiments and calibrated simulations for chemicals for which no acceptable agreement could be obtained: Sabourin et al. (2009) (carbamazepine), and Yang et al. (2012) (androstenedione, progesterone).

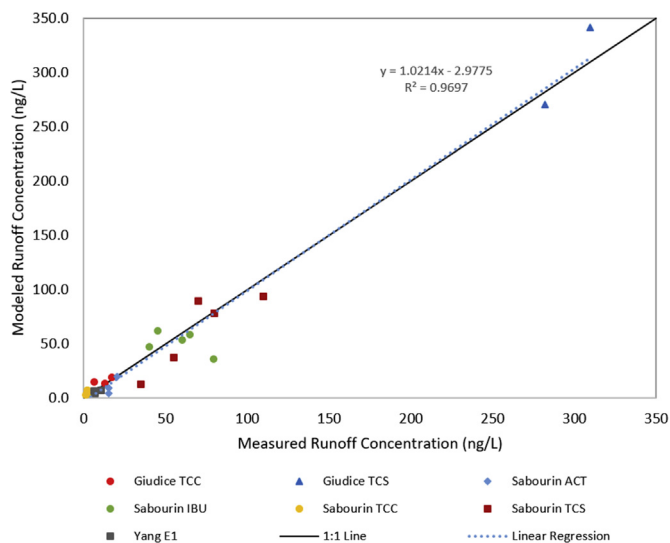


Fig. 5. Results of experiments and calibrated simulations for chemicals for which reasonable agreement could be obtained: Sabourin et al. (2009) (acetaminophen, ibuprofen, triclocarban, and triclosan) Giudice and Young (2011) (triclocarban and triclosan), and Yang et al. (2012) (estrone).

found to be generally 10–20% of the mass lost in the aqueous phase, though it was only slightly less than lost in the aqueous phase for triclocarban at 36 days post application. The results for TCS agree with the model, which predicted mass lost in the sorbed phase to be 13–24% of the mass lost in the aqueous phase. However, the model predicted for TCC that 3–5 times as much would be lost in the sorbed phase than the aqueous phase. The mass of hormones lost via the sorbed phase was investigated in (Yang et al., 2012), and approximately 30–40% of the total mass of estrone lost was via the sorbed phase. The model predicted that estrone loss in the sorbed phase would be only 1–10% of that lost in the aqueous phase, but this is likely due to very low modeled estimates of erosion for this experiment that may not reflect actual conditions.

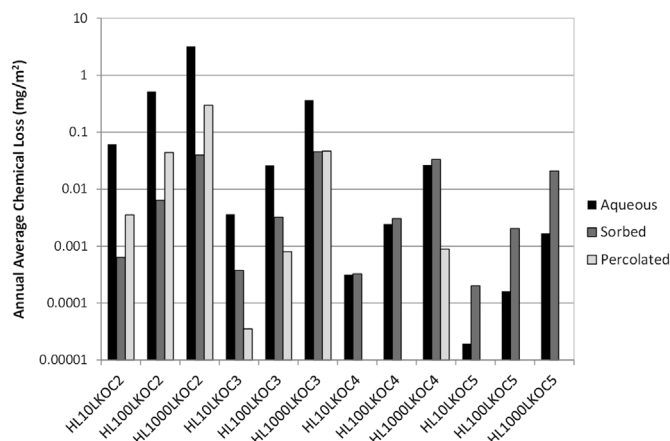


Fig. 6. Average annual mass loss of generic chemicals for aqueous and sorbed phases in runoff and in percolated rainfall of the 12 generic chemicals for biosolids applied annually on January 2. Annual application rate of each chemical was 10 mg/m².

As mentioned previously, further work is necessary to calibrate the subsurface transport component of the model.

3.2. Application

Fig. 6 shows the annual average mass loss in the sorbed and aqueous phases in runoff and in percolated rainfall of the 12 generic chemicals for biosolids applied annually on January 2. As expected, the chemical with the highest K_{OC} and shortest half-life (HL10LKO5) exhibited the smallest losses, while the chemical with the lowest K_{OC} and longest half-life (HL1000LKO2) exhibited the greatest losses. For each factor of 10 K_{OC} decrease (e.g., Log K_{OC} 5 to Log K_{OC} 4) and half-life increase (e.g., 10 days–100 days), annual average chemical loss (total, and within each phase) increased by approximately a factor of 10. Total annual chemical loss in runoff is plotted as a function of the recurrence interval for biosolids applied annually on January 2 in Supplemental Fig. S1. The figure shows that for the chemical with the highest losses,

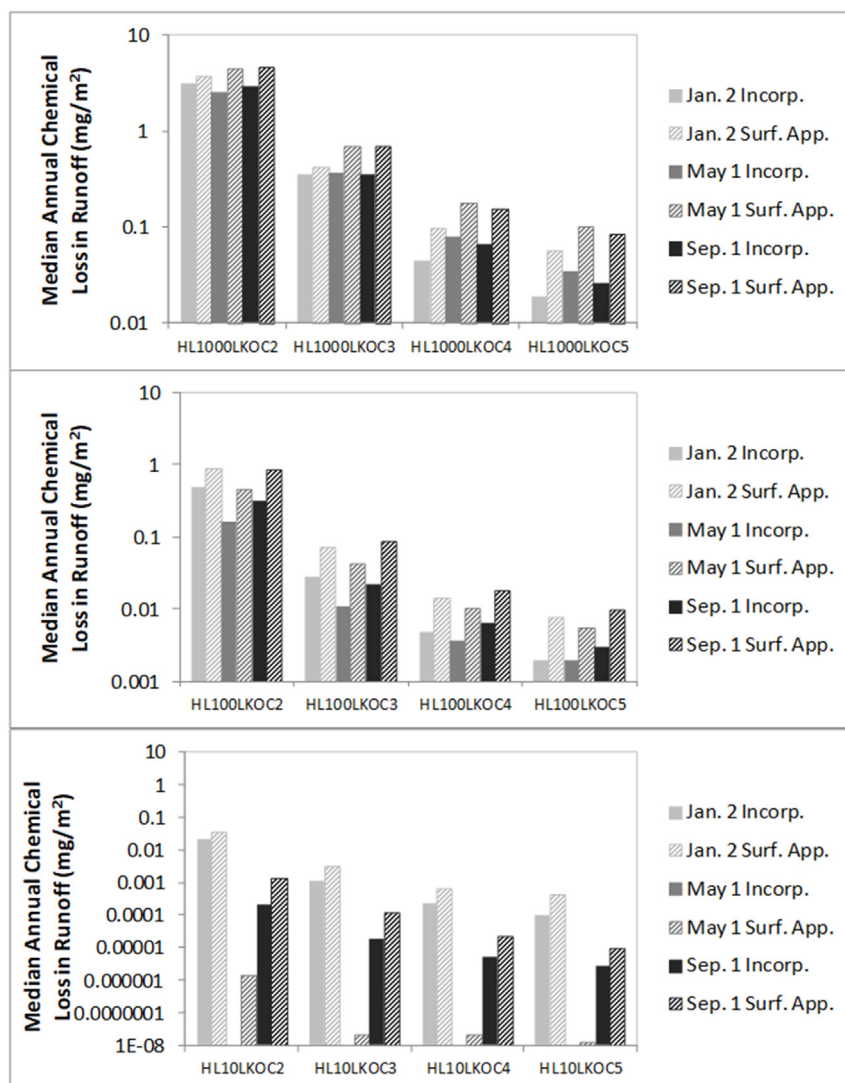


Fig. 7. Median annual mass loss of generic chemicals in runoff (aqueous plus sorbed) given annual biosolids applications of January 2, May 1, and September 1 for surface application and incorporation into the top 11.4 cm of soil.

HL1000LKOC2, over the course of the simulation, in many years the total amount of chemical lost was on the same order of the amount applied.

Fig. 6 also shows the relative contributions of sorbed and aqueous phases in runoff and of percolated rainfall of the 12 generic chemicals to their average annual mass loss. The runoff aqueous phase makes up the majority of the mass loss for chemicals with $\text{Log } K_{OC} < 4$, while for those with $\text{Log } K_{OC} > 4$, the sorbed phase in runoff makes up the majority of the loss. Loss via percolate is correlated with loss in the aqueous phase of runoff, though is always less, and is essentially zero for chemicals with $\text{Log } K_{OC} > 4$.

The effect of the annual application day is shown in Fig. 7. For chemicals with short half-lives (i.e., half-lives of 10 and 100 days; bottom two panels), annual application on May 1 results in the least chemical mobilized. This is because virtually all of the precipitation at this site occurs between November and April. Therefore, after annual application on May 1, in almost all years there is ample time for chemicals to degrade prior to any runoff-inducing rainfall. However, for chemicals with long half-lives, on the order of 1 year or longer (i.e., the 1000 day chemicals; top panel), and relatively high K_{OC} , annual application on January 2 results in the least chemical mobilized. This is because for these chemicals, organic carbon added to the plots as part of the biosolids matrix degrades much more rapidly than the chemicals

themselves. The chemicals accumulate in the soil over a period of years, and the primary driver for temporal variation in mobility is not chemical concentration, but soil/biosolids organic carbon content. The highest organic carbon content in the biosolids phase occurs in the weeks and months immediately following application, and for a January 2 application date, this coincides with the majority of the rainfall.

This phenomenon of increased mobility of biosolids-associated solutes as organic matter in the biosolids degrades has been previously documented and explored in depth for metals (Chang et al., 1997; McBride, 1995; Stacey et al., 2001), but very little work has been done with trace organic contaminants like those for which the model was developed. Further research is necessary to understand and determine whether and under what conditions the model results for long-term applications reflect actual organic carbon degradation and trace organic contaminant mobility.

Fig. 7 also shows the effect of application type. In all cases, surface spreading results in greater, and many times considerably greater, median annual losses in runoff than when incorporated. This is because in the case of surface spreading, all of the mass of chemical that is applied is done so into the top 1 cm of soil/biosolids, which is the layer that is available to surface runoff. In this case, since incorporation is to a depth of 11.4 cm, incorporation results in only about 9% of the chemical applied into the top layer (i.e., 1 cm/11.4 cm).

4. Conclusion

The model presented herein simulated the fate and transport in runoff of trace organic chemicals in land-applied biosolids. Seven out of ten chemical-scenarios for which experimental data was available in the literature (across three independent studies) showed good agreement between model predictions and experimental concentrations. Based on the chemicals for which the model was successful and unsuccessful, the model is useful for predicting the concentration in runoff of target chemicals for which concentrations are unlikely to increase due to degradation/interconversion of parent/related chemicals, and whose transport in soil and biosolids can be modeled via linear partitioning to organic matter. Increasing concentrations of chemicals after biosolids application (for example, transformation of one hormone to another), and colloid-facilitated transport are beyond the scope of this model, but could be integrated in the future once the mechanisms are well-understood. Applications of the model show the utility in helping to identify management practices that result in lesser impacts to water quality.

Declaration of interest

None.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2019.01.011>.

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