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Review

Fugacity modelling of the fate of micropollutants in aqueous systems — Uncertainty and sensitivity issues

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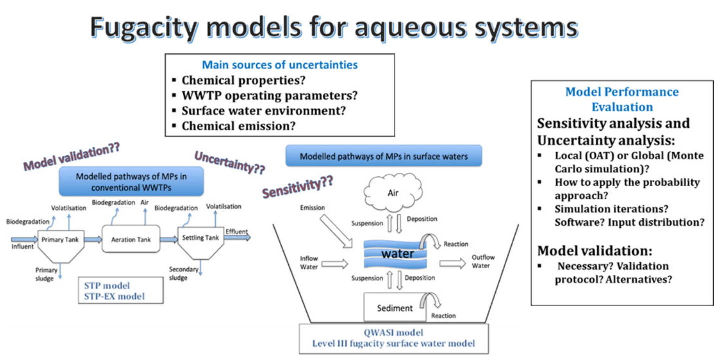
HIGHLIGHTS

- Critical review of 22 fugacity model applications with a focus on uncertainties
- Input data uncertainties tend to be a major issue in this modelling approach
- Clearer descriptions of sources and methodologies for model inputs are essential
- Both uncertainty and sensitivity analyses should be performed in fugacity modelling
- A procedure protocol for fugacity modelling strategies is useful for future work

GRAPHICAL ABSTRACT



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ABSTRACT

The application of multimedia fugacity models is useful to facilitate understanding of the behaviour of emerging contaminants during wastewater treatment, as well as after their release to the environment. In this paper, twenty-two fugacity modelling applications (reported over 1995–2019) describing the distribution of organic micropollutants in wastewater treatment plants and surface water bodies were analysed in terms of model application and modelling strategy. Disparities and similarities in strategies including selection of micropollutants, data sources for internal and external model inputs, sensitivity and uncertainty analysis, as well as model validation were discussed. This review confirmed that fugacity modelling is very applicable for providing qualitative predictions of the fate and removal of organic micropollutants in the various aqueous systems. However, it was also noted that there are issues related to the uncertainties and sensitivities of fugacity models such as the sources of model inputs and selection of default settings. The issues associated with the uncertainties in the investigated fugacity models are pointed out. Recommendations are given regarding the selection of the sources of model inputs, sensitivity analysis strategies and model validation methods. This review presents the challenges and opportunities for improving multimedia fugacity models, and so paves the way for future research in this field.

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Contents

1. Introduction

Over the past two decades, the increasing detection and reporting of micropollutants (MPs) in the aquatic environment has raised great concern. MPs such as pharmaceutical and personal care products (PPCPs), endocrine disrupting chemicals (EDCs) and pesticides are frequently detected globally in surface water and have been linked to adverse ecological effects (Luo et al., 2014). There is evidence showing that effluent discharge from wastewater treatment plants (WWTPs) is a major pathway for the introduction of MPs to the aquatic environment (Blair et al., 2013). Hence, a better understanding of the fate and removal of MPs in WWTPs and the environment would facilitate the management of these emerging contaminants. MPs are commonly present in waters at trace levels ranging from ng L^{-1} to μg L^{-1}, which makes their detection and quantification a challenge (Luo et al., 2014). Modelling is therefore regarded as a useful approach to predict the fate and removal of MPs during wastewater treatment and in the receiving environment (Pomiès et al., 2013). Data on the occurrence and removal of MPs in different water matrices were summarised in a review paper by Luo et al. (2014). Others have reviewed the removal of MPs during different types of wastewater treatment (Bolong et al., 2009; Deblonde et al., 2011) such as biological treatment systems including activated sludge (Onesios et al., 2009) and membrane bioreactors (MBRs) (Verlicchi et al., 2012). The application of concentration-based models for the biological wastewater treatment of MPs has been evaluated (Pomiès et al., 2013) and Su et al. (2019) recently reviewed the application of different multi-media models for the prediction of chemicals in various environmental systems.

Concentration-based models have been developed to simulate micropollutant removal mechanisms during wastewater treatment (Pomiès et al., 2013), as well as for multimedia environmental systems (Brandes and Den Hollander, 1996; Beyer and Matthies, 2002). Chemical concentrations for each removal pathway are required for the development of the model. The advantage of concentration-based models is that they enable the simulation of the detailed removal mechanisms of MPs during treatment processes or within a certain environment. Two well established concentration-based models are the ASMx model for the prediction of the fate of MPs during activated sludge treatment (Plósz et al., 2012; Plósz et al., 2013; Polesel et al., 2016), and the EOPLS model to support decision-making and assumed travel distance of MPs within the global environment (Beyer and Matthies, 2002; Wania and Dugani, 2003; Fenner et al., 2004; Zarfl et al., 2012). Other concentration-based models have been developed to better understand the behavior of MPs during various treatment processes over the past two decades (Lee et al., 1998; Byrns, 2001; Urase and Kikuta, 2005; Limousin et al., 2007; Plósz et al., 2009; Vasiliadou et al., 2013; Fernandez-Fontaina et al., 2014; Bürger et al., 2016).

A fugacity-based multimedia fate model is an environmental chemistry model which describes the processes controlling chemical behavior in environmental media by developing and applying mathematical statements of chemical fate (Mackay, 2001). These models could comprehensively illustrate the distribution of chemical concentrations in various compartments within a system (Mackay and Paterson, 1991). Quantitative analysis can be provided with accounts of the emission sources, transportation and transfer routes, as well as sinks of MPs (MacLeod et al., 2010), even on a national temporal level (Zhang et al., 2015).

One advantage of the fugacity approach is that the use of fugacity, instead of concentration, makes it easy to use in mass-balance calculations to describe the behavior of chemicals released into the environment and to simulate how chemicals are directionally transported between several compartments (Mackay and Paterson, 1991; Su et al., 2019).

Fugacity models have been used frequently for the prediction of the fate of MPs in aqueous systems. One of the limitations of this type of model is the uncertainty in model outputs. Buser et al. (2012) summarised six general principles of good modelling practice guidelines for applying multimedia model in a decision-making context. The present paper reviews recently published fugacity-based fate models applied to MPs in wastewater and surface water systems. The inherent sources and causes of uncertainties in aqueous fugacity models were investigated as an update to the recommendations given by Buser et al. (2012). Model features, applicabilty and modelling strategies are critically reviewed, with a focus on exploring the sources of uncertainties in the modelling applications, as well as the different approaches employed for uncertainty and sensitivity analysis. Future perspectives and recommendations for the application this type of model to different systems are discussed.

2. Fugacity models

A critical overview of the models covering the period from 1995 to 2019, sourced from two major scientific databases (Scopus and ScienceDirect) describing the fate of organic MPs during wastewater treatment and in surface water bodies, is presented here. For each model, the original literature was found, and models with higher citations were selected to review. Three main fugacity models, including the STP model, the QWASI model and similar static Level III fugacity model, and the dynamic fugacity model are reviewed. Twenty-two fugacity model applications (eight for wastewater treatment plants and fourteen for surface water bodies) are compared and critically discussed.

Fugacity models have also been used to predict the fate of metals (primarily mercury) in lakes and reservoirs (Diamond et al., 2000; Ethier et al., 2008; Liu et al., 2017). Some models have been developed to assess the fate of MPs in multi-media environments other than aqueous systems, such as the BETR model for global scale application (MacLeod et al., 2001) and the MUM model for urban system application (Diamond et al., 2001). Due to the differences in the compounds and systems under investigation, these model applications are not covered in this review.

2.1. Fugacity concepts and approach

Fugacity (denoted as f and measured in Pa) which describes chemical escaping tendency, is regarded as an equilibrium criterion. Therefore, when a compound moves between two compartments, equal fugacity in both phases is expected, and the escaping tendency, or pressure of this compound in both phases, is considered equal. Based on a linear correlation, chemical concentration (C, mol m^{-3}) is expressed as in Eq. (1).

$$C = Z \times f \tag{1}$$

where Z (mol m^{-3} Pa^{-1}) is the fugacity capacity. The value of Z is specific to a chemical, and depends on the phase in which it resides, and temperature. A compartment with a higher fugacity capacity is able to accept a higher concentration of a given micropollutant. Then fugacity can be deduced from a known concentration of a solute chemical in one phase, or vice versa (Mackay, 2001). The flows of the chemical in water, biomass solids, and air, for both degradation and surface volatilisation, constitute transport and transformation processes. The rates of these processes are known as the D values (mol h^{-1} Pa^{-1}) and also need to be determined to fully develop a fugacity-based mass balance model. To calculate the D values, the Z values, as well as information related to the compounds (removal mechanism coefficients/rate constants) and wastewater treatment plant under consideration (volume of sewage tank and mass flows) are required.

Levels I, II and III multimedia fugacity models are commonly used for the prediction of the fate and distribution of organic compounds in the environment (Mackay and Paterson, 1991). In level I models, steady-state and equilibrium conditions are assumed for the transport of a compound between all the environmental phases involved (air, water, soil, sediment and biota). In level II models, only transformations and advection of a compound are assumed as being in steady-state and equilibrium conditions. In level III models inter-media transport processes are included; the compound is discharged at a constant rate into the chosen environmental media and achieves a steady-state, but non-equilibrium condition at which input and output rates are equal (Mackay, 2001). Level III models are the most widely used approach and provide the most valuable insights into the fate of chemicals within an environmental system. There is another type known as a level IV model which gives predictions assuming unsteady state, non-equilibrium conditions. The level IV fugacity model best represents a system but is the most complex and has more data requirements (Kilic and Aral, 2009). It has been used for dynamic systems such as rivers (Wang et al., 2012b). Other models, such as SimpleTreat and SimpleBox models, were also developed based on a fugacity approach to describe the interface transfer between compartments. However, the mass balance equations were built based on compound concentration rather than fugacity. These models are regarded as concentration-based rather than fugacity-based models and so were not included in the scope of this review paper.

2.2. Fugacity-based fate models for MPs in aqueous systems

2.2.1. The STP model

The sewage treatment plant (STP) model was developed by Clark et al. (1995) to understand the fate of MPs in a conventional activated sludge treatment plant. In this model, Clark et al. (1995) set up mass balance equations for each stage of the wastewater treatment process to correlate and predict the steady-state phase concentrations, process stream fluxes and the fate of MPs in a sewage treatment plant. One limitation of this STP model was its limited capacity for handling ionised compounds. Seth et al. (2008) upgraded the STP model to the STP-EX model by updating the Z values of the ionising chemicals (multiplying by the ionic/neutral factor). The ionic/neutral factor was derived based on the pK_a of the compound under investigation, as well as the pH of the wastewater using the equation:factor $I = {}^{\mathrm{Ionic/Neutral}} = 10^{(\mathrm{pH}-\mathrm{p}K_a)}$)). Applications of STP and STP-EX models to conventional WWTPs have been reported worldwide to assess the treatability of selected MPs during treatment or to use it as a screening level risk assessment (Wang et al., 2007; Bock et al., 2010; Thompson et al., 2011; Wang et al., 2015a). The STP and the STP-EX models were applied by Tan et al. (2007) and Wang et al. (2015b), respectively, to WWTPs with unconventional designs and they confirmed that the fugacity approach can be successfully applied to these systems to predict the fate of MPs.

2.2.2. The QWASI model and the static level III fugacity model

The STP model is not suitable for simulating the fate and behaviour of MPs in surface waters. The Quantitative Water Air Sediment Interaction (QWASI) model was developed to study the fate of MPs in lakes and rivers. The QWASI model is a level III multi-media fate and transport model based on fugacity concepts (Mackay et al., 1983). The model was used to investigate the fate of a range of chemicals in lakes and its predictions have been demonstrated to match values measured onsite (Mackay and Diamond, 1989). Since its development, the QWASI model and its modified versions have been widely applied to simulate the concentrations, distributions, transfer fluxes, and bioaccumulation fluxes of chemicals in lake and river systems (Mackay and Diamond, 1989; Mackay and Hickie, 2000; Warren et al., 2002; Whelan, 2013; Xu et al., 2013; Mackay et al., 2014; Guo et al., 2019). Apart from the QWASI model, other Mackay-type static fugacity models have been developed to describe the fate of MPs in a river (Duan et al., 2013), a river basin (Zhang et al., 2013) and reservoirs (Cao et al., 2010; Hawker et al., 2011).

2.2.3. Dynamic fugacity model

One drawback of static fugacity models when being applied to rivers was that they did not account for the continuous spatial variability of hydrological characteristics in the corresponding input parameters, due to the lack of spatial profile in the model structure (Wang et al., 2012c). Therefore, dynamic and continuous fugacity models have been developed to assess the fate and distribution of MPs in rivers in a number of studies (Kilic and Aral, 2009; Zhang et al., 2011; Wang et al., 2012b). These coupled the static fugacity model with hydraulic dynamic water flows to simulate the river network, and the fugacity approach was applied to describe the dynamic interactions between all phases in the physical domain.

3. Modelling strategy

On reviewing the application of fugacity models, it is clear that the modelling approaches and/or model evaluation strategies varied between studies. This section compares several modelling aspects including the use of model inputs, approach to sensitivity and uncertainty analysis, as well as validation strategy.

Table 1
Summary of chemical selection and sources of physicochemical properties in the fugacity modelling applications reviewed (EPI: Estimation Programs Interface, QSAR: quantitative structural activity relationship).

MP category	No. of MPs	Source of chemical related inputs	Reference
Application to WWTPs			
Wide range selection	12	Literature Estimated k_{bio}	Clark et al. (1995)
EDCs	8	Literature	Tan et al. (2007)
PAHs	7	Literature	Wang et al. (2007)
Wide range selection	20	Literature Estimated k_{bio} (similar to STP default setting)	Seth et al. (2008)
EDC-Triclosan	1	Literature	Bock et al. (2010)
EDCs	6	Literature EPI Suite estimation Temperature corrected k_{bio}	Thompson et al. (2011)
Cyclic VMSs	3	Literature; Onsite measurement calculation (k_d)	Wang et al. (2015a)
Cyclic and linear VMSs	3	Literature (unclear)	Wang et al. (2015b)
Application to surface water systems (static models)			
PCB	1	Literature Assumption (degradation rate)	Mackay and Diamond (1989)
PAHs	7	Literature Temperature correction	Mackay and Hickie (2000)
Lindane, Benzo(a) pyrene	2	Literature	Warren et al. (2002)
EDCs	3	Literature EPI Suite estimation (H, Melting point) Measurement (half-life in water) Assumption	Cao et al. (2010)
Wide range selection	263	EPI Suite estimation	Hawker et al. (2011)
Pharmaceuticals	5	EPI Suite estimation Onsite measurement (k_{OC})	Duan et al. (2013)
Cyclic VMSs	3	Literature with adjustments	Whelan (2013)
PAHs	15	Literature	Xu et al. (2013)
EDCs	2	Literature	Zhang et al. (2013)
D5 & PCB-180	2	Literature	Mackay et al. (2014)
Cyclic VMSs	3	Literature Calculated/adjusted based on literature	Guo et al. (2019)
Application to surface water systems (dynamic models)			
PCB and atrazine	2	Literature	Kilic and Aral (2009)
EDCs	3	Literature Calculation Onsite measurement (k_{OC})	Zhang et al. (2011)
PAHs	8	Literature	Wang et al. (2012b)

3.1. Model inputs

There are two types of model inputs, those which are chemical-related (internal) and those which are the plant/environment-related (external). Tables 1, 2 and 3 summarise the sources of various sets of model inputs. Note that the study on the original development of the QWASI model (Mackay et al., 1983) is not included as this paper simply described the general features of the models and the mathematical concepts behind the establishment process, without describing specific inputs.

3.1.1. Chemical-related model inputs and selection of MPs

Table 1 summarises modelling strategies associated with the selection of target MPs as well as the source of the MP physicochemical properties used in the model applications in this review. The MPs studied cover a wide range of substance categories and physicochemical properties. It appeared most of the researchers tended to establish a model for a small number of chemicals within one category. However, Hawker et al. (2011) reported the fate of 15 chemicals with various functionality and physicochemical properties. Clark et al. (1995) developed the STP model for twelve chemicals from several categories including polyaromatic hydrocarbons (PAHs), pesticides and volatile organics. A similar selection of MPs was used in the advancement of the STP model performed by Seth et al. (2008) who used 20 MPs with a range of properties. The most frequently modelled category of MPs was EDCs, with three applications each in wastewater and surface water systems, and the most frequently studied EDCs were bisphenol A and triclosan, with three or more references. The other most commonly studied chemical was decamethylcyclopentasiloxane (D5), with five model applications investigating its fate in both surface water and wastewater systems. It can be concluded that the selection of target MPs was mostly based on: i) occurrence of certain MPs in the water matrix of interest or ii) coverage of widely varying physicochemical properties. However, for many of the models reviewed, the basis for selection of the MPs was not clearly explained.

Chemical-related inputs are the physicochemical properties of the target MPs. These parameters play a role in their removal mechanisms and can determine the fate of the MPs under investigation (Luo et al., 2014). The fate of MPs in WWTPs include volatilisation, biodegradation and sorption, with the Henry's law constant (H), biodegradation rate

Table 2
Summary of sources of WWTP-related input parameters in the fugacity model applications.

Model type	WWTPs related inputs	References
STP	Typical illustrative operating conditions Assumption from literature	Clark et al. (1995)
STP	Assumption from literature Onsite measured concentrations	Tan et al. (2007)
STP	Onsite record (operating conditions & concentrations)	Wang et al. (2007)
STP-EX	Literature Default STP setting Reported concentrations	Seth et al. (2008)
STP-EX	Default settings of STP Literature reported monitoring data Onsite measured concentrations	Bock et al. (2010)
STP	Adjustment of default STP settings Assumption from literature Reported concentrations	Thompson et al. (2011)
STP	Onsite operating conditions Onsite measured concentrations	Wang et al. (2015a)
STP	Onsite operating conditions Onsite measured concentration	Wang et al. (2015b)

Table 3
Summary of sources of surface water system-related input parameters in the fugacity model applications – static models.

Model type	Surface water system related inputs	Reference
QWASI	Actual measurement Literature	Mackay and Diamond (1989)
QWASI	Local environment agency report characteristics Reported emission	Mackay and Hickie (2000)
QWASI	Local sampling and monitoring Reported or estimated emission	Warren et al. (2002)
Level III	Literature Historical monitoring data	Cao et al. (2010)
Level III	Literature default Local authority data Maximum emission estimated from measurement	Hawker et al. (2011)
QWASI	Default values from literature Local sampling and monitoring Relevant literature measurement Local authority water discharge estimation	Xu et al. (2013)
Level III	Default values from literature Local hydrographic offices Measured emission	Duan et al. (2013)
QWASI	Literature Estimated emission based on usage	Whelan (2013)
Level III	Literature Calculation Reported emission	Zhang et al. (2013)
Updated QWASI	Default setting of QWASI model Literature Estimated emission	Mackay et al. (2014)
QWASI	Literature Temperature adjusted values Estimated emission based on usage	Guo et al. (2019)

constant (k_{bio}) and water-octanol coefficient (K_{ow}) being the fate determining properties, respectively (Clark et al., 1995). The fate of MPs in surface waters is a little more complicated as it can be attributed to various pathways including volatilisation, transformation in water and sediment, photolysis and sorption to sediment and particles in water. In addition to the Henry's law constant and the sediment sorption coefficient, photodegradation rate, transformation rate in water and sediment compartments are all important physicochemical properties which play significant roles.

The majority of the internal input parameters for the models were obtained from literature data or via conversion of literature data. In ten of the twenty-two applications, properties reported in the literature were the only source of chemical-related model inputs. However, the limited kinetic data available in the literature is a potential source of uncertainty in model inputs, as noted by Pomiès et al. (2013). Furthermore, the experimentally obtained data in the literature may have been determined under different conditions for the water matrices under study, leading to inaccurate model simulations. Although Seth et al. (2008) proposed a scheme for obtaining appropriate biodegradation half-lives from the literature for MPs in different water bodies, most researchers did not provide the rationale for the choice of data from the literature.

For both wastewater and surface water studies, the properties can be estimated by using computer programs such as USEPA EPI Suite (Cao et al., 2010; Hawker et al., 2011; Thompson et al., 2011; Duan et al., 2013). One of the limitations of this procedure is that the biodegradation rate constants estimated by the software used for this purpose (BIOWIN) has certain limitations: i) it only provides accurate predictions for MPs the molecular properties of which are fully described in the BIOWIN model database (Dayan and Kromidas, 2011), ii) it may underestimate the half-lives of persistent organic contaminants (Aronson et al., 2006), and iii) it does not provide temperature-dependent predictions as its outputs are generated for the given temperature of 25 °C (Thompson et al., 2011).

Model inputs were measured experimentally under plant/environmentally relevant conditions in only a limited number of studies as a means of avoiding generating more uncertainties in model predictions. Sorption-related parameters were frequently measured experimentally when fugacity modelling was utilised. Onsite measurement of the sediment sorption coefficient (K_{oc}) was performed by Duan et al. (2013) and Zhang et al. (2011). Similarly, Wang et al. (2015a) performed onsite measurements of the sludge sorption coefficient (K_d). Degradation (activated sludge biodegradation, photodegradation and degradation of chemicals in surface water or sediment) rate constants are another type of important input parameter. For surface water studies, only Cao et al. (2010) measured the degradation rates of MPs in the reservoir water.

Wang et al. (2007) did not provide a clear description of the source of their chemical-related model inputs. Since the degradation rates of MPs in various phases, especially the sediment and air phases, are not readily available, estimations were used for the determination of these model inputs for the surface water models (Cao et al., 2010; Mackay et al., 2014), which could generate uncertainties in the model outputs. This could have been further addressed using sensitivity and uncertainty analysis, which is further discussed in Section 3.2.

Adjustment of the chemical-related model inputs can enhance the accuracy of fugacity models. The performance of the STP model was evaluated by Wang et al. (2007) to compare the model predictions with actual data collected by sampling at various stages of a full-scale activated sludge process. Their results showed that the predicted removal efficiencies of PAHs were in agreement with the measured data after the input biodegradation half-lives were adjusted. They found that the reported aqueous biodegradation half-lives divided by scaling factors of 50 and 150 could result in more accurate model estimations of the concentrations of the target compounds. Improvement of the accuracy of the biodegradation parameters was also achieved by Thompson et al. (2011) who incorporated the effect of temperature on the biodegradation half-lives of the target compounds into the STP model to study the fate of four EDCs, phenol and tetrachloroethylene (PCE) in a conventional three stage WWTP. They concluded that with the temperature calibrated biodegradation half-lives, the STP model could provide useful operational considerations regarding removal of the target MPs in different seasons. Wang et al. (2015a) investigated the fate of three cyclic volatile methylsiloxanes (VMSs) in a conventional WWTP. Adjustments including negligible removal of VMSs via biodegradation and temperature corrected air-water coefficient (K_{aw}) values for the target compounds were made to make the model suitable for application in this study. The predicted and measured values were fairly consistent, with volatilisation being the primary removal pathway for octamethylcyclotetrasiloxane (D4) and sorption to the biomass the most important removal mechanism for D5 and dodecamethylcyclohexasiloxane (D6). This is reasonable since the sorption of D5 to particulate or dissolved organic matter has been observed by other researchers (David et al., 2000; Whelan et al., 2010).

3.1.2. Treatment plant related model inputs

WWTP-related parameters include operating parameters (plant design dimensions, operating conditions), and MP concentrations in the input water and sludge flows. As shown in Table 2, the WWTP operating parameters used in model applications mainly had three sources: i) onsite data obtained from either the plant management/operator; ii) operating data obtained from or assumed based on literature reported values for plants with similar configurations, and iii) default settings in the STP model. MP levels in the plants could be obtained from onsite measurements (current or records) and reported concentrations in the literature. Onsite plant operating conditions and MP measurements are more plant specific, and more relevant model outputs could be expected. However, the information collected can be rather limited, especially when grab samples are used (Wang et al., 2007). Values from many similar WWTPs reported in the literature could provide a larger input database (Bock et al., 2010), covering the best and worst scenarios as much as possible, which is a useful approach in terms of modelling and simulation. The use of a combination of onsite monitoring data and literature reported values could serve both purposes, to be site-specific and provide larger input databases (Seth et al., 2008; Bock et al., 2010). Moreover, updates of the default WWTP setting could be important to reflect the current plant operating conditions due to technical development since the original model was established.

It should also be noted that some of the WWTP fugacity model applications were generalised investigations of typical STPs (Clark et al., 1995; Seth et al., 2008; Bock et al., 2010; Thompson et al., 2011), rather than describing an existing specific plant (Tan et al., 2007; Wang et al., 2007; Wang et al., 2015a; Wang et al., 2015b). It is then reasonable to find that for the four generalised model applications default STP settings and concentrations reported in the literature were used. On the other hand, onsite operational parameters (except for Tan et al. (2007)) and measured concentrations were used for the other four model applications developed for existing WWTPs.

3.1.3. Surface water system-related model inputs — static models

Surface water system-related model inputs included system characteristics and chemical emission to the aqueous systems. The parameters used embrace the system dimensions, sediment characteristics (density, solid content etc.), water flow conditions and sediment movement rates.

As shown in Table 3, surface water system-related inputs used in the model applications mainly had three sources. Information on the system dimensions or flow rates is generally obtained as onsite data from local authorities (Mackay and Hickie, 2000; Hawker et al., 2011; Duan et al., 2013), local historical monitoring data (Cao et al., 2010) and actual measurement (Mackay and Diamond, 1989; Warren et al., 2002). Some studies only used data reported in the literature for other surface water systems, without explaining the relevance or similarity between the systems under study and the reported ones (Whelan, 2013; Zhang et al., 2013; Mackay et al., 2014). Information on the sediment conditions and movement was usually obtained from literature, default model settings or default data from literature, since this information was generally not readily available for the systems under study. Chemical emission to the surface waters was another important model input, onsite measurement of MP emission was performed by only one modelling group (Zhang et al., 2011; Duan et al., 2013). Most applications used estimated emission (Hawker et al., 2011; Whelan, 2013; Mackay et al., 2014; Guo et al., 2019) or reported values (Mackay and Hickie, 2000; Xu et al., 2013; Zhang et al., 2013), or a mixture of both (Warren et al., 2002). Mackay and Diamond (1989) and Cao et al. (2010) did not mention the MP emissions in their studies.

3.1.4. Surface water system-related model inputs — dynamic models

As well as the model inputs for static models, dynamic fugacity models also utilise hydrodynamic estimations. Kilic and Aral (2009) and Zhang et al. (2011) utilised modified one-dimensional advection-dispersion Saint Venant equations while Wang et al. (2012b) used a one-dimensional kinematic wave equation.

As noted in Table 4, Zhang et al. (2011) and Wang et al. (2012b) used onsite measurements as well as the chemical emission rates to the surface water system as model inputs, however, Kilic and Aral (2009) did not take these into consideration. This was probably because the aim of their study was to build the hydrodynamic model for the river network, with the evaluation of the fate of chemicals as a minor purpose.

Like the internal input parameters, data obtained from the literature, default settings or via estimations for external parameters could generate variability in model predictions for WWTPs and surface waters. These values contain degrees of uncertainty which should be addressed in the modelling process. Some parameters may have little impact on the model outputs and so their values can be estimated with little additional effort. For those to which the models are very sensitive, precise values should be obtained. This can be analysed using sensitivity and uncertainty analysis, which will be discussed in the next section.

3.2. Model sensitivity and uncertainty analysis

Uncertainty analysis is essential for describing the lack of knowledge inherent in a model and its parameters (Uusitalo et al., 2015). This is particularly useful for fugacity models, for which predictions are only an approximation of the actual fate of MPs and uncertainty is a major limiting factor that needs to be addressed (Cacuci et al., 2003; Mackay et al., 2014). Different forms of uncertainty exist and include the following: parameter uncertainty, model uncertainty, dependency uncertainty (Burgman, 2005). This section summarises the different strategies involved in the assessment of uncertainty and variability of model output.

3.2.1. Assessment strategies

Two commonly used approaches include uncertainty and sensitivity analyses. Two strategies are usually adopted to assess the uncertainty and sensitivity of a model. One strategy is known as the "one-at-a-time" (OAT) method in which each parameter under investigation is varied systematically over a range whilst keeping other factors constant (a local sensitivity/uncertainty analysis) (Cacuci et al., 2003). This method is generally used for sensitivity analysis (Cao et al., 2010; Zhang et al., 2011; Hu et al., 2017; Liu et al., 2017) and can be used to provide some insights into the causes of the model uncertainties (Whelan, 2013; Mackay et al., 2014; Guo et al., 2019).

Another approach is to perform random sampling and Monte Carlo simulation using commercial software (Bock et al., 2010; Mackay et al., 2014; Wang et al., 2015a; Wang et al., 2015b), also known as a global sensitivity/uncertainty analysis (Cacuci et al., 2003). In this method, a probabilistic approach via Monte Carlo simulation is used to quantify uncertainty and variability in the model predictions. This technique is based on a repeated random sampling of the probability distributions of each assumed parameter (Cao et al., 2004). Probability density functions are developed for input parameters to cover the expected variability using distribution fitting. The model runs many times (generally 500–20,000) so that an output distribution of concentrations and transfer fluxes in different environmental media can be constructed.

Among the 22 fugacity applications reviewed in this paper, Zhang et al. (2013), Xu et al. (2013) and Cao et al. (2010) used a mix of both strategies in their studies, including a global probabilistic approach for uncertainty analysis and an OAT local inspection for sensitivity analysis.

Table 4
Summary of sources of surface water system-related input parameters in the dynamic fugacity model applications.

Model type	Surface water system related inputs	Hydrodynamic estimations	Reference
	Literature and calculation based on measurements; Emission rate ignored	One-dimensional advection-dispersion St. Venant equation	K Kilic and Aral (2009)
Level III	Onsite measured parameters and emission/discharge	One-dimensional advection-dispersion St. Venant equation	Zhang et al. (2011)
Level IV	Onsite measured parameters and emission	One-dimensional kinematic wave equation	Wang et al. (2012b)

Mackay et al. (2014) adopted both strategies in their sensitivity and uncertainty assessment and obtained comparable yet not identical results.

Both local and global sensitivity/uncertainty analyses have been widely applied in a variety of modelling applications. Local techniques concentrate on estimating the local impact of a parameter on the model output. This approach means that the analysis focuses on the impact of changes in a certain parameter value. Local analysis methods, such as the OAT approach, are very useful for hydrological and water quality models (Francos et al., 2001; Van Griensven et al., 2002), since they can analyse the sensitivity of several parameters with a low computational cost. This approach can also enable the immediate knowledge of which input factor is responsible for a model failure under OAT analysis (Saltelli and Annoni, 2010). However, local methods have certain limitations, since they only make evaluations at one point in the parameter hyperspace (van Griensven et al., 2006). On the other hand, global techniques analyse the whole parameter space at once (van Griensven et al., 2006). A global approach aims to show that even varying the input assumptions within some plausible ranges, some desired inferences hold (Saltelli and Annoni, 2010). In principle, local analyses cannot be used for demonstrating the robustness of model-based inference unless the model is proven to be linear or at least additive (Saltelli et al., 2006). When the property of the models is unknown, a global sensitivity/uncertainty analysis is preferred (van Griensven et al., 2006; Saltelli and Annoni, 2010; Sin et al., 2011).

3.2.2. Sensitivity analysis

A sensitivity analysis can be used to identify the key input parameters and evaluate the influence of individual parameters on the outcome variance of the multimedia fugacity model (Cao et al., 2004). Performing a sensitivity analysis can demonstrate the importance of the individual input variables on the model outputs. Where the model is shown to be sensitive to a parameter, then the optimisation of that parameter needs to be dealt with carefully (Whelan, 2013). It is recommended that the higher impact parameters be obtained via onsite measurement to ensure the accuracy of the model output (Liu et al., 2017). On the other hand, the parameters to which the model is insensitive can be estimated or cited from other sources with less precision (Whelan, 2013; Liu et al., 2017).

With both local and global strategies, model sensitivity is generally assessed by the coefficient of correlation between input and output: either i) the Spearman rank order correlation coefficient obtained by Monte Carlo simulation; or ii) manually calculated sensitivity coefficients (SCs) in the OAT method which are defined as the ratio of the relative change of model output to that of the input parameters as shown in Eq. (2) (Zhang et al., 2011; Wang et al., 2012b; Zhang et al., 2013; Hu et al., 2017):

$$\mathrm{SC} = \frac{\Delta \mathrm{Y_i}/\mathrm{Y_i}}{\Delta \mathrm{X_i}/\mathrm{X_i}} \tag{2}$$

where X and Y are input and output parameters in the model. Some modelling groups did not report the SCs in their OAT sensitivity analyses.

Of the twenty-two model applications reviewed, sensitivity analysis using an OAT method was applied to several input parameters in nine studies with (Cao et al., 2010; Whelan, 2013; Xu et al., 2013; Zhang et al., 2013; Mackay et al., 2014) or without (Thompson et al., 2011; Zhang et al., 2011; Wang et al., 2012b; Guo et al., 2019) uncertainty analysis. Some studies did not clearly present the SCs calculated. Thompson et al. (2011) and Cao et al. (2010) simply compared the change (in %) of model outputs along with the change of each specific model input. It is difficult for the users to determine the extent of the sensitivity of the model to each parameter if SC calculations are not given. Moreover, it is also inconvenient to do comparisons when many input parameters are involved in the analysis.

Only four model applications (Bock et al., 2010; Wang et al., 2015a; Wang et al., 2015b) adopted global sensitivity analysis, including Mackay et al. (2014) who used both OAT and probabilistic approaches. These analyses were coupled with uncertainty analyses using a Monte Carlo simulation to better evaluate the model uncertainty and performance.

3.2.3. Uncertainty analysis

Generally, sensitivity and uncertainty analyses are performed in tandem (Saltelli et al., 2006). Uncertainty analysis is an assessment of the various sources of uncertainty to the model output (Zhang et al., 2013). Due to lack of knowledge about the model inputs and/or errors and variability in the experimental and environmental conditions, the input parameters will have some degree of uncertainty which will contribute to the overall uncertainty associated with the model results (Hawker et al., 2011; Kim et al., 2013).

However, of the twenty-two model applications reviewed in this paper, global uncertainty analysis was performed in only seven studies. Four conducted both sensitivity and uncertainty analyses as mentioned above. Most of these were conducted using global uncertainty analysis. Another two applications included simple local uncertainty analysis by discussing the possible changes caused by selected model inputs (Hawker et al., 2011; Whelan, 2013).

3.2.4. Probabilistic approach — global sensitivity/uncertainty analysis

As mentioned previously, seven model applications reviewed in this paper conducted global uncertainty/sensitivity analysis using a probabilistic approach. Different software or simulation sampling methods were applied, which are discussed in this section. The probabilistic factors used include the distribution fitting of input parameters, the software used for simulation, the type of simulation used as well as the number of iterations of simulation, which are summarised in Table 5.

As shown in Table 5, Crystal Ball was the most commonly used software for simulation. Three modelling groups did not provide information regarding the version of the software/function used. All researchers used Monte Carlo random sampling simulation except Mackay et al. (2014) who considered that Latin Hypercube was more efficient than random sampling. Latin Hypercube has advanced sampling efficiency and has been regarded as a variance-reduction technique (McKay et al., 1979). However, some researchers have pointed out that further assessment is required for Latin Hypercube simulation (Joseph and Hung, 2008; Pronzato and Müller, 2012). Monte Carlo random sampling analysis is an important tool in the probabilistic analysis in building simulation. Monte Carlo simulation is advantageous since it is flexible, easy to use and does not suffer from multidimensionality or non-linearity (Zio and Pedroni, 2013). However, the computational costs of Monte Carlo can be a limiting factor. It was shown that a better sampling strategy and convergence assessment will improve applicability (Janssen, 2013). The number of iterations of simulation ranged from 500 to 20,000, with 10,000 runs used in most applications (Bock et al., 2010; Mackay et al., 2014; Wang et al., 2015a; Wang et al., 2015b). Cao et al. (2010) used only 500 simulations, and although they stated that this enabled reasonably accurate representation of the probability profile, they did not provide any statistical proof to confirm the validity of their model. A larger sample size is recommended to provide better accuracy of the simulation (Kim et al., 2013).

A single type of distribution was fitted for all the input parameters in four studies (Cao et al., 2010; Mackay et al., 2014; Wang et al., 2015a; Wang et al., 2015b) which may not be realistic considering the variability of the input data. Only Mackay et al. (2014) provided a reason for their selection and noted that they chose log-normal distribution because it samples only positive values. However, this is questionable since the choice of distribution should be based on the performance of the distribution fitting. Zhang et al. (2013) assumed normal or log-normal distributions for inputs around the mean or point values, and

Table 5
Summary of the probabilistic approach applied in the seven fugacity model applications.

Water matrix	Analysis type	Distribution of parameters	Simulation Software	Simulation sampling (No. of runs)	Reference
Wastewater	Sensitivity + Uncertainty	Normal Log normal Uniform	Crystal Ball	Monte Carlo random (10,000)	Bock et al. (2010)
Surface water	Uncertainty	Normal	Visual Basic Macro Function in Excel	Monte Carlo random (500)	Cao et al. (2010)
Surface water	Uncertainty	Normal Log normal	Built-in function "Randn" in Matlab	Monte Carlo random (3000)	Xu et al. (2013)
Surface water	Sensitivity + Uncertainty	Log normal	Crystal Ball 7.3.1	Latin Hypercube (10,000)	Mackay et al. (2014)
Surface water	Uncertainty	Normal or log-normal	Built-in function "Randn" in Matlab	Monte Carlo (20,000)	Zhang et al. (2013)
Wastewater	Sensitivity + Uncertainty	Uniform	Crystal Ball 7.3.1	Monte Carlo random (10,000)	Wang et al. (2015a)
Wastewater	Sensitivity + Uncertainty	Normal	Crystal Ball 7.3.1	Monte Carlo random (10,000)	Wang et al. (2015b)

in the other two studies, three types of distribution fitting were selected to best represent the variability of their data.

3.3. Model validation

Validation of a fugacity model includes a check of both its internal and external consistency. In order to ensure that model calculations are internally consistent, a mass balance can be performed for the overall process to ensure that the chemical balance closes. An internal consistency check would be sufficient to ensure that output data could correctly reflect all the inherent equations, values of input parameters and simplifying assumptions. The model could be used simply as an evaluative tool to provide a description of the principal fate and transport processes, then the model results could be used illustratively rather than reflecting the real situation and no external validation is required (Mackay et al., 2014). On the other hand, if the model is to be used as a predictive or simulation tool, it would be better to conduct a thorough validation by comparing the results predicted by the model with field data (Mackay et al., 2014). Hence, additional data on chemical concentrations in the inlet and outlet of a system or even in each compartment within an aqueous system would be needed.

Information on model validation was not mentioned in the development and description of two models (Clark et al., 1995; Mackay et al., 2014). Although proper model validation was not performed, Cao et al. (2010) compared their simulated results with the concentration data in a local water quality monitoring report to check the validity of their model. Among the other twelve WWTP applications, validation of the applied fugacity model was undertaken for six of them (Tan et al., 2007; Bock et al., 2010; Xu et al., 2013; Wang et al., 2015a; Wang et al., 2015b; Guo et al., 2019), who performed validation by using MP concentrations measured in both water and sludge phases. Of the fugacity model applications for MPs in surface waters, four of ten groups performed model validation (Mackay and Diamond, 1989; Mackay and Hickie, 2000; Zhang et al., 2011; Zhang et al., 2013), and the concentrations of MPs in both water and sediment phases were measured to assess the validity of the models.

4. Discussion

4.1. Chemical properties of MPs

The physicochemical properties of the target MPs can be a source of uncertainty in fugacity modelling. Although Hawker et al. (2011) noted that the physicochemical properties of MPs can be predicted with reasonable precision and have a small influence on the output of a fugacity model, sensitivity analyses for several models indicated that chemical properties, especially the degradation rate/half-life and sorption related coefficients, are strongly linked to the variability and accuracy of the model outputs.

Thompson et al. (2011) reported that the removal of target MPs was very sensitive to their activated sludge biodegradation half-lives. Khan and Ongerth (2004) stated that the uncertainty in the biodegradation rates led to inaccurate predictions for chemical distribution in the effluent of a WWTP. However, Bock et al. (2010) suggested that although the half-life of triclosan in activated sludge was uncertain, it made a negligible contribution to variation in the model output. As found for surface water bodies, the fate of MPs was sensitive to the degradation rate (Hawker et al., 2011; Zhang et al., 2013) or hydrolysis half-life (Mackay et al., 2014) in water and sediment. Several research groups (Whelan, 2013; Xu et al., 2013; Zhang et al., 2013; Mackay et al., 2014) determined that K_{oc}, a factor with considerable uncertainty, was the most influential model input affecting the predicted concentrations of MPs in sediment (Guo et al., 2019).

By reviewing all these fugacity model applications, it appeared that for the WWTP studies, k_{bio} was an important parameter which needed extra care in choosing the most appropriate input sources. It would be more appropriate to obtain k_{bio} values by performing bench scale biodegradation experiments which simulate actual plant operating conditions. For those without the resources for performing laboratory work, software estimated k_{bio} values could be useful alternatives, where appropriate adjustment (e.g., based on temperature) may be needed to enhance the accuracy of the model outputs. In both static and dynamic fugacity model development for surface water systems, K_{oc} was a significant factor among the model inputs and required onsite measurement or proper deduction using mathematical models (Whelan et al., 2010; Hyland et al., 2012) to achieve better model performance. However, it is strongly recommended to perform onsite measurement of k_{oc} with quantification of MPs in both solid and aqueous phases for each WWTP if possible. Alternatively, mathematical models can be used to estimate the k_{oc} values for MPs (Whelan et al., 2010; Hyland et al., 2012). Generally, reasonably accurate Henry's Law constant (H) values can be estimated using software (USEPA EPI Suite) or obtained from literature.

There is complementarity between fugacity-based and concentration-based models. As mentioned in the Introduction section, concentration-based models have been established to simulate the detailed removal mechanisms of MPs during wastewater treatment, including biodegradation (Plósz et al., 2009; Min et al., 2018), sorption (Urase and Kikuta, 2005; Polesel et al., 2015a) and volatilisation (Byrns, 2001). The kinetic parameters required as the fugacity model inputs can be predicted using concentration-based models. In this way, fugacity model uncertainties caused by inappropriate sources of model inputs could be reduced. This is particularly the case for the study of the fate of MPs during wastewater treatment processes, where the complementarity of the two types of model may result in a more holistic picture.

4.2. Chemical emission rate

Chemical emission rate (or discharge rate) is among the most important model inputs in the application of fugacity models to natural water systems (Zhang et al., 2015). For some MPs (e.g., pharmaceuticals), their human excretion rates contribute significantly to model output uncertainty. It has been reported that significant uncertainties were associated with the estimation of community drug emission/consumption through wastewater analyses (Castiglioni et al., 2013; Polesel et al., 2015b; Gracia-Lor et al., 2016). Apart from sampling and chemical analysis, other issues included stability of drug biomarkers in sewage, back-calculation of drug use and estimation of population size in a given system (Castiglioni et al., 2013; Ort et al., 2014; Gracia-Lor et al., 2016).

Only three of the fourteen natural water applications did not incorporate this factor in their modelling strategies (Mackay and Diamond, 1989; Kilic and Aral, 2009; Cao et al., 2010). In previously reported studies, chemical emission rates were generally estimated from the usage of products containing the target MPs (Whelan, 2013; Guo et al., 2019), or based on historical reported values (Mackay and Hickie, 2000). Wang et al. (2012b) found that the predicted concentrations of PAHs in the water column were most sensitive to their emission rate. The uncertainty of emission rate can impact the simulation of the environmental concentrations of MPs as varied emission rates could lead to different absolute concentrations predicted for each compound in each compartment of the receiving environment (Whelan, 2013).

A better estimation or measurement of MP emission is crucial to better understand their environmental behaviour in natural water systems. Zhang et al. (2015) performed a comprehensive study to investigate the national consumption and emissions of a range of frequently detected antibiotics in China by a market survey. However, such large-scale studies can be challenging and resource consuming. Moreover, under some circumstances, it is difficult to quantify the consumption and emissions of some chemicals, thus leading to significant uncertainty. In these cases, to address the significant uncertainty, an alternative to precise measurements of emission rates is to perform emission scenario studies (Zhang et al., 2011; Whelan, 2013). Model outputs can be analysed at different emission rates to find the boundary conditions of the systems under investigation. Similar analysis has been performed by other researchers studying the fate of MPs at global scale (Liu et al., 2015; Su et al., 2018). Overall, it is very important for the modeller to explain how the emission rates (or emission threshold) for the MPs were estimated in each fugacity model application.

4.3. WWTP related parameters

Plant operating parameters are common sources of uncertainty in fugacity models developed for WWTPs. The concentration of suspended solids can be a source of uncertainty. Bock et al. (2010) found that variation in volatile suspended solids (VSS) concentration accounted for approximately 60% of the variation in predicted effluent concentrations and 70% of the variation in the predicted biosolids concentration of triclosan. The concentration of total suspended solids (TSS) in effluent was reported to be a major influence on the emission of VMSs in the effluent due to the hydrophobic nature of these MPs (Wang et al., 2015a; Wang et al., 2015b). Therefore, uncertainties in the TSS concentration could greatly affect the modelled output of VMS emissions.

Another essential factor was sludge retention time (SRT), which greatly impacts the biodegradation of MPs (Bock et al., 2010; Wang et al., 2015b). These researchers found that the rate of solids wasting from the secondary sedimentation tank (related to SRT) was a determining factor controlling the biodegradation of MPs in the activated sludge process. This was reasonable since it has been shown that longer SRTs will promote bacterial growth and enrich microbial diversity, and so facilitate biodegradation and thus greater removal of MPs during the treatment (Clara et al., 2005; Langford et al., 2005). On the other hand, Samaras et al. (2013) showed that increased SRT (from 8 days to 18 days) in full scale WWTPs did not lead to improved biodegradation of all selected target MPs. This could be due to the different microbes with different degradation kinetics present in the activated sludge in the WWTPs studied by the different authors. Another explanation could be the nature of the different MPs investigated in these studies. The time required for acclimation may differ for different microbial flora, leading to the different influences of SRT on their biological removal. As Kruglova et al. (2016) explained, longer SRT conditions can lead to greater diversity of microbial species, particularly for those which grow slowly. Other than this factor, WWTP influent flow rate (Wang et al., 2015a; Wang et al., 2015b) and mixed liquor suspended solids (MLSS) concentration (Wang et al., 2015b) can also influence the mass distributions of MPs in the WWTP and lead to uncertain model predictions.

Overall, for MPs which are primarily adsorbed during wastewater treatment, the concentration of suspended solids is likely to influence their fate the most. Other WWTP related parameters can also play important roles in influencing the fate of MPs. To achieve the best removal of persistent MPs, it is recommended to perform sensitivity analysis for various operating parameters for each WWTP system during fugacity modelling. The sensitivity analysis results can provide the means for the operators to obtain optimum operating conditions.

4.4. Surface water system characteristics

The hydraulic characteristics of surface waters are considered major sources of uncertainty in fugacity models developed for these water bodies. Hawker et al. (2011) assumed a well-mixed lake model, however the target water system stratified during six months of the year; this could generate a degree of uncertainty in the model output. A similar situation applies to QWASI models, and Mackay and Hickie (2000) assumed a well-mixed water column for a lake without considering stratification.

Surface water systems can be more complex than WWTPs as the environmental conditions are not engineered or controlled which can generate more variables within the system. Therefore, more characteristics need to be taken into consideration. System parameters such as water volume, system surface area, water temperature, wind speed (Cao et al., 2010; Hawker et al., 2011; Wang et al., 2012b; Xu et al., 2013) and biomass concentration (Cao et al., 2010) are subject to spatial and temporal variability, which can generate uncertainty. Zhang et al. (2013) concluded that water system inflow rate was an important factor. Despite this, Zhang et al. (2011) reported that their model was insensitive to water depth and water surface area. This was probably because they were not investigating photodegradable MPs. Xu et al. (2013) reported that the predictions for the PAHs they investigated were highly sensitive to water temperature. Since temporal differences of water temperature also apply to natural water systems, it is recommended that fugacity models for natural water systems are developed taking this into account. Temperature sensitive model input parameters should be adjusted according to temperature variations using the mathematical models available (Guo et al., 2019).

Some estimated system parameters such as sediment burial, deposition and resuspension rates are also potential sources of uncertainty (Hawker et al., 2011; Guo et al., 2019). It was reported by Mackay et al. (2014) that the concentration of PCB-180 in lake water was very sensitive to either the sediment deposition rate or the sediment burial rate in the three lakes studied. A similar conclusion was made by Guo et al. (2019) for D4. Therefore, the precision of these rates related to sediment movement is essential to ensure accuracy of model output.

The application of fugacity models to MPs in surface water bodies differs from application in WWTPs as the diffusion and mass transfer coefficients play more important roles. Zhang et al. (2013) found that the air-soil diffusion coefficient was an influential factor for a surface water body. Uncertainty analysis showed that predictions of concentrations in sediment had the largest uncertainty and more information about this key process should be collected, such as the mass transfer coefficients between the sediment and other phases (air and water) (Zhang et al., 2013). Zhang et al. (2011) reported negligible impact of the water-side mass transfer coefficient, whereas the air-side mass transfer coefficient was found to be an important factor as the model predictions displayed variability in response to this parameter.

4.5. Model performance evaluation

A better understanding of the impact of input parameters during sensitivity and uncertainty analyses has practical implications. The information obtained from these analyses can facilitate optimisation of WWTP operation to reduce the discharge of MPs to the environment (Wang et al., 2015a, Wang et al., 2015b). Moreover, information associated with surface water characteristics may also be useful to predict the changes in the amounts of MPs in the system under different weather or recycling conditions (Cao et al., 2010). Buser et al. (2012) suggested that sensitivity analysis and possible uncertainty analysis should be performed in multi-media model applications. Nearly half of the fugacity model applications covered in this review performed at least one analysis (sensitivity or uncertainty), with seven of them undertaking both analyses. Uncertainty analysis was generally performed using a probabilistic approach via computer simulation. However, the simulation methodologies adopted in several of the applications varied. Furthermore, some studies did not provide the statistical data to support their methodology selection. It is recommended that statistical quality assurance data (e.g., error, standard deviations, etc.) should be included in future fugacity model performance evaluations. Also, the number of simulation iterations should be considered with caution. Generally, a large number provides greater accuracy in Monte Carlo simulation. However, specific model structure and computational effort required should also be taken into account. The number of runs should be sufficient to demonstrate the statistical relevance of the results. Also, once a number has been chosen, the number of simulation runs should exceed that number as a test. Statistical proof should be provided to show that the increased sample size does not significantly affect the output probability distribution.

Some authors simply analysed the potential uncertainties present in model outputs by briefly demonstrating input data variability and/or uncertainty without using the simulation approach (Hawker et al., 2011; Zhang et al., 2011; Whelan, 2013). The reliability of these analyses is questionable since no statistical support was provided. Sensitivity analysis was performed either by using computer simulation or an OAT inspection. With the OAT method, several modelling groups did not report the SC in the analysis. More in depth understanding could be obtained if these SC values were included since they can directly show the users the relative sensitivity of the model outputs to each parameter investigated. For example, detailed explanation and demonstration of the sensitivity analysis were provided by Xu et al. (2013). The sensitivity coefficients of each parameter for each MP obtained in sensitivity analysis were tabulated. The variance coefficients for each MP obtained in uncertainty analysis were visualised using column graphs for comparison. These simple and powerful visualisation tools allowed a clear and straightforward understanding of the study. Inclusion of a table or graph of the analysis outputs is necessary for the users to better understand the model performance in terms of robustness and output variances (Huang et al., 2019).

Validation is an important approach to assess the performance of an established model. Even though Mackay et al. (2014) stated that model validation is not compulsory if the results were used qualitatively, some researchers did not describe the clear use of modelling data. Model validation was performed for twelve of the twenty-two model applications reviewed in this paper. All of the model validations were performed by the determination of onsite measurements in both liquid and solid phases, indicating a fairly high reliability of the data sets. Only two of the twelve models were calibrated (Bock et al., 2010; Zhang et al., 2011). Calibration is a process to adjust certain model parameters to achieve the best performance of the model for specific locations and applications, whereas validation attempts to assess the closeness between the predictions and the observations (Dee, 1995). Certain adjustment of model parameterisation is necessary to ensure a better model performance (Refsgaard, 1997). However, it is crucial to ensure the accuracy of the data used for model calibration. Overall, it is reasonable to not calibrate those parameters which were determined to be less influential on model outputs to reduce the number of parameters to be calibrated (Jens 1997). Sensitivity and uncertainty analyses facilitate the identification of the model parameters required to be calibrated (Su et al., 2019).

For those with reported validation data, a majority of models (with or without calibration) produced predictions which were fairly consistent or comparable with measured concentrations in both wastewater and surface water matrices (Mackay and Hickie, 2000; Wang et al., 2007; Bock et al., 2010; Zhang et al., 2011; Xu et al., 2013; Zhang et al., 2013; Wang et al., 2015a; Wang et al., 2015b). Tan et al. (2007) found differences ranging from 0 to 40% between the modelled and measured concentration of the EDCs being studied, suggesting some model predictions could be inaccurate. For multimedia fugacity models, it is acceptable to have differences of less than 0.5 orders of magnitude between the modelled and observed values (Cao et al., 2004). Hence, the reported model effectiveness demonstrates the success of this type of model in the prediction of the fate and concentrations of MPs in WWTPs and environmentally relevant aqueous systems. Therefore, it is suggested that in the absence of appropriate data, model calibration is not a compulsory component of the fugacity modelling strategy. Jørgensen et al. (2014) pointed out that the aim of calibration in fugacity modelling is to avoid impossible model parameters instead of fitting the model results as closely as possible to the observations.

It is strongly recommended that model validation be performed and validity demonstrated prior to quantitative application to prediction of chemical concentrations. A clear description of the purpose of the modelled results should be provided if validation was not conducted. In particular, the use of onsite measurements in both liquid and solid phase samples is recommended since it gives greater reliability of validation results. This is especially the case for applications aiming to improve or adjust a current model (Seth et al., 2008; Thompson et al., 2011). Particular attention should be paid to fugacity models developed for natural water systems with unique environmental conditions, where comparable data are not easily found. It is also suggested to collect historically measured data for the target MPs in the associated region from literature sources, this can provide a comprehensive dataset for comparison (Liu et al., 2015). It is recommended to use quantitative statistical methods for model validation to better evaluate the performance of the established model. Three methods including Nash–Sutcliffe efficiency (NSE), percent bias (PBIAS) and ratio of the root-mean-square to the standard deviation of measured data (RSR) have been used for similar purposes (Wang et al., 2012a; Kim et al., 2017). However, none of the reviewed model applications clearly presented statistical analysis in their model validation.

Another problem related to fugacity model performance is over-parameterisation. During model development, it is difficult to judge the appropriate level of detail within modelling processes. Consequently, it is easy for models to become over-parameterised, potentially increasing uncertainty in predictions (Crout et al., 2009). This is particularly the case for Bayesian network as well as hydrological models (Gaume and Gosset, 2003; van Griensven et al., 2006). Similar situations exist for fugacity models due to model complexity, especially for the dynamic fugacity models used for the study of MPs in rivers which include hydrological features. However, by applying sensitivity analysis and uncertainty analysis, the uncertainty generated by this issue can be mitigated (van Griensven et al., 2006).

5. Conclusions

Fugacity models have been applied to both conventional and unconventional WWTPs with different operating conditions, as well as various natural water bodies including reservoirs, lakes and rivers to gain insights into the behaviour and fate of MPs in the various aqueous systems. Three main types of fugacity models were reviewed in this paper including the STP model, the QWASI model and similar Level III fugacity model, as well as dynamic fugacity model. Twenty-two fugacity model applications for both surface water and wastewater systems were reviewed here.

STP is a commercial computer program designed for WWTPs and has been widely applied to predict the fate of MPs in sewage treatment plants. However, this model only covers conventional wastewater treatment process (activated sludge) configurations, with tertiary processes such as chlorination, ozonation, and lagoon treatment not included. In terms of surface water fugacity models, most input data for these models were obtained from literature or default data. It is recommended that more effort should be put into improving the quality of input data to enhance their performance in the prediction of the fate and removal of MPs in surface waters. Another limitation of surface waters fugacity models is that the sources of the pollutants are usually regarded as a steady state system so that the transport of pollutants can be described by a one-dimensional advection-dispersion equation of steady state.

For the STP and STP-EX model applications, k_{bio} appeared to be the parameter which requires particular attention when choosing the most appropriate input sources. For surface water fugacity models, K_{oc} is an important factor. More accurate measurement or careful deduction of this parameter is desirable to give better model performance. Chemical emission/discharge rate is a crucial model input which can lead to significant uncertainty. Careful estimation of this parameter and a detailed estimation scheme in the modelling strategy are desirable to achieve better model performance. Wherever applicable, it is recommended to take all the possible emission routes into consideration, with a thorough background investigation (such as literature study) on the usage, consumption and population. Alternatively, development of emission scenarios is useful to enable the determination of the boundary conditions which can be used in emission estimation. For experimentally obtained model inputs, laboratory conditions and experimental approach should be described in detail. For model inputs obtained from literature or by estimation, reasons for the selection of the data sources should be given.

Uncertainty and sensitivity analysis of the system parameters as well as of the chemical-related parameters can provide useful and insightful information regarding the variability of the outputs of a (fugacity) model. The most important input parameters of the model as well as the influence of those parameters on the model estimations can be identified with this approach. It is highly recommended to perform both analyses using the appropriate methodologies. Also, statistical quality assurance data should be presented along with the Monte Carlo simulation strategies. It is recommended that the number of simulation iterations be chosen carefully. A balance between simulation quality and computational effort should be maintained. For those research groups without the resources to perform multiple simulations, the OAT strategy can be applied. However, it is important for modellers to report detailed analysis results in a clear manner (including SC in the results to show the extent of sensitivity). Model validation by comparing the predicted data with site measurements was conducted for only approximately half of the model applications. Hence uncertainties remain with regard to the real performance of those models that have not been validated. Moreover, it is strongly recommended to use quantitative statistical methods for model validation to enable quantitative evaluation of the model performance.

This review has discussed the causes and sources of fugacity model uncertainties when used to predict the fate of MPs in aqueous systems. The importance of the quantity and quality of data in developing useful fugacity models to predict the fate of MPs in both wastewater and surface water systems has been demonstrated. In future research, it would be useful to adopt a chemical moiety approach to enhance both quantity and quality of internal input data for this type of model. Also, the applications of fugacity models can be expanded to make it suitable for the description of the fate of MPs in different aqueous systems; for example, for wastewater lagoon treatment and natural water systems with large water surfaces in which photolysis can be an important pathway. By including this MP removal mechanism in the fugacity modelling process, uncertainty of the model output can also be reduced.

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