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# A new multimedia contaminant fate model for China: How important are environmental parameters in influencing chemical persistence and long-range transport potential?

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ABSTRACT

We present a new multimedia chemical fate model (SESAMe) which was developed to assess chemical fate and behaviour across China. We apply the model to quantify the influence of environmental parameters on chemical overall persistence ($P_{OV}$) and long-range transport potential (LRTP) in China, which has extreme diversity in environmental conditions. Sobol sensitivity analysis was used to identify the relative importance of input parameters. Physicochemical properties were identified as more influential than environmental parameters on model output. Interactive effects of environmental parameters on $P_{OV}$ and LRTP occur mainly in combination with chemical properties. Hypothetical chemicals and emission data were used to model $P_{OV}$ and LRTP for neutral and acidic chemicals with different $K_{OW}/D_{OW}$, vapour pressure and $pK_a$ under different precipitation, wind speed, temperature and soil organic carbon contents ($f_{OC}$). Generally for $P_{OV}$, precipitation was more influential than the other environmental parameters, whilst temperature and wind speed did not contribute significantly to $P_{OV}$ variation; for LRTP, wind speed was more influential than the other environmental parameters, whilst the effects of other environmental parameters relied on specific chemical properties. $f_{OC}$ had a slight effect on $P_{OV}$ and LRTP, and higher $f_{OC}$ always increased $P_{OV}$ and decreased LRTP. Example case studies were performed on real test chemicals using SESAMe to explore the spatial variability of model output and how environmental properties affect $P_{OV}$ and LRTP. Dibenzofuran released to multiple media had higher $P_{OV}$ in northwest of Xinjiang, part of Gansu, northeast of Inner Mongolia, Heilongjiang and Jilin. Benzo[a]pyrene released to the air had higher LRTP in south Xinjiang and west Inner Mongolia, whilst acenaphthene had higher LRTP in Tibet and west Inner Mongolia. TCS released into water had higher LRTP in Yellow River and Yangtze River catchments. The initial case studies demonstrated that SESAMe performed well on comparing $P_{OV}$ and LRTP of chemicals in different regions across China in order to potentially identify the most sensitive regions. This model should not only be used to estimate $P_{OV}$ and LRTP for screening and risk assessments of chemicals, but could potentially be used to help design chemical monitoring programmes across China in the future.



## 1. Introduction

With the rapid development of the Chinese economy, the production, manufacture and consumption of chemicals has increased sharply over recent years. This has inevitably resulted in the detection of residues of pesticides, polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyl (PCBs), polybrominated diphenyl ethers (PBDEs) and emerging chemicals like pharmaceuticals and personal and household care product ingredients (PPCPs) in the environment including water (Bai et al., 2008; Zhao et al., 2010), air (Wang et al., 2012), soil (Wang et al., 2009, 2010), biota (Yuan et al., 2014) and food (Wang et al., 2013). The fate and behaviour of these chemicals in the diverse Chinese environment has increasingly attracted scientific and political interest, with particular concerns arising from human exposure to these chemicals via the environment. The Ministry of Environmental Protection (MEP) of China published the amended version of Provisions on Environmental Administration of New Chemical Substance in 2010 (MEP, 2010), which is similar to the EU REACH guideline (European Commission, 2006), and provides a risk based approach to chemical management. It is well recognized that the potential for harmful effects

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of chemicals is not limited to areas of close proximity to their use but also to remote and vulnerable ecosystems (Matthies et al., 2009). Overall persistence ($P_{OV}$) and long-range transport potential (LRTP) are two metrics that are widely used to determine the chemical fate and potential environmental concern. They are determined by both chemical and environmental properties and are reliant on model predictions in a particular geographical region (OECD, 2004). Multimedia models have been developed as valuable tools to calculate $P_{OV}$ and LRTP of chemicals (Fenner et al., 2004; Klasmeier et al., 2006; OECD, 2004). Such models can be used to investigate $P_{OV}$ and LRTP characteristics for chemicals including those that are not yet introduced into an environment and to explore how the spatial variability in environmental conditions may impact. In general, it has been concluded that chemical properties largely determine the ranking of chemicals according to $P_{OV}$ and LRTP. The effect of chemicals properties has been carefully discussed using hypothetical chemicals by Fenner et al. (Fenner et al., 2005). Though, environmental factors, especially climate, have been shown to influence chemical behaviour (Wania and Mackay, 2000) this is likely to be especially important for a climatologically diverse country such as China.

A number of researchers have developed multimedia models to investigate the fate of chemicals across large geographical regions in Europe and North America, such as EVn-BETR (Prevedouros et al., 2004), IMPACT 2002 (Margni et al., 2004) and BETR North America (MacLeod et al., 2001). Multimedia models perform an important role in risk assessment and decision support in Europe, e.g. Simplebox is embedded in EUSES (European Union System for the Evaluation of Substances) (Vermeire et al., 1997); CoZMo-POP2 is linked with a human food chain bioaccumulation model ACC-HUMAN in model CoZMoMAN to study the human exposure and body burden of PCBs for Baltic Sea drainage basin (Breivik et al., 2010; Quinn et al., 2011).

To date, as far as the authors are aware, there have been no attempts to develop a multimedia model for the whole of China for a broad range of substances including ionisable chemicals. Some multimedia fate models have been designed and applied for China but only in a local or a regional scale for neutral organic chemicals. For example, Mackay level III and IV fugacity models have been used to study the fate of benzo[a]pyrene, phenanthrene and γ-HCH in Tianjin by Tao et al. (Cao et al., 2004; Tao et al., 2003, 2006; Wang et al., 2002). Similar studies were completed in many other local or regional scale areas across China (Cao et al., 2007; Lang et al., 2007; Li et al., 2006; Xu et al., 2013; Zhang et al., 2013b).

This paper presents a Sino Evaluative Simplebox-MAMI Model (SESAMe) operating on two scales that is applicable for a broad range of ionisable and neutral chemicals. The model construct is presented and it has been used to investigate the influence of chemical and environmental properties on chemical $P_{OV}$ and LRTP. It could be utilized further for human exposure estimation and environmental risk assessment for China. Simplebox 3.24a (Huijbregts et al., 2008) and a single box multimedia model MAMI III for ionisable chemicals (Franco and Trapp, 2010) have been referred to in constructing the model. Sobol sensitivity analysis method, instead of classic sensitivity analysis methods, has been used to quantify the importance of input variables on $P_{OV}$ and LRTP. This method has been widely used in sensitivity analysis for hydrology models (Cibin et al., 2010; Tang et al., 2007a; Zhang et al., 2013a). The advantages of this method are that it considers a range of the input variables (Yang, 2011), which has not been considered by previous classical approaches (MacLeod et al., 2002); and it has been found to have the advantage of offering the most detailed description of the effect of a single parameter and its interactions with other parameters on the output, resulting in a more robust sensitivity ranking among the available global sensitivity analysis methods (Tang et al., 2007b), which is suitable for exploring the environmental–chemical interactions and influence on $P_{OV}$ and LRTP. A series of neutral and acidic hypothetical chemicals have been used to study how representative environmental parameters influence $P_{OV}$ and LRTP. Lastly, case studies using real chemicals have been conducted to explore the spatial variability of model output and how environmental properties affect $P_{OV}$ and LRTP. These include benzo[a]pyrene (Bap), acenaphthene (ACE), triclosan (TCS) and dibenzofuran (DF) and enable a preliminary evaluation of the performance of the model.

## 2. Methods and materials

### 2.1. Model definition

SESAMe has been designed using the nested structure of Simplebox 3.24a model (Huijbregts et al., 2008) and the formulas developed in the MAMI III model (Franco and Trapp, 2010; Trapp et al., 2010) for modelling the concentration and fate of ionisable chemicals. The model has two scales; regional and continental, and considering that 200 km × 200 km has been the default size of regional scale for Europe in Simplebox, it was determined to be the size of the regional scale in this study. The continental scale was considered to be a 9,496,200 km$^2$ box covering the rest of the mainland China (Fig. S1 in Supplementary Information). The nested approach to spatial scale makes it possible to predict the concentration of chemicals both within the emission region and further afield and then to calculate $P_{OV}$ and LRTP. Each scale contains 8 compartments including air, freshwater, sediment, natural soil, agricultural soil, urban soil, natural vegetation and agricultural vegetation. The chemical was assumed only to be emitted to the regional scale and fixed at 1 million tonnes per year. As in MAMI III model, only neutral molecules were assumed to volatilize to the air from water or soil compartments; unlike MAMI III model, the ideal condition was assumed in the water phase (ionic strength is 0). Vegetation was assumed to play an important role in the fate of airborne chemicals through deposition, so the vegetation compartment has been added for ionisable chemicals (acid, base and amphoter) to make the model complete (the calculations can be found in Supplementary Information, Eqs. (1–8)). In this study only acidic and neutral chemicals were included.

### 2.2. Environmental parameters

The spatial distribution maps of 14 environmental parameters were either collected from existing databases or generated from raw data taken from databases (Nos. 11–24 in Table 1 and Supplementary Information). ArcGIS 10 was used to extract the values of these parameters in order to represent each regional grid (Supplementary Information). The values of the variables for each of 267 grids completely within the boundary of mainland China were used to generate the environmental parameter database for the following hypothetical study.

### 2.3. Chemical parameters

To study the effect of environmental parameters on the model output, hypothetical chemicals were used: however, 130 real organic chemicals covering a wide range of chemical properties were taken from the substance database in Simplebox 3.24a model as chemical database, so that it would be easier to apply the results to real chemicals at a later date. The values for degradation rates and enthalpy of vaporization were updated or supplemented from the handbook of physico-chemical properties (Mackay, 2005). The range of each parameter is contained in Table 1 (Nos. 1–10).

### 2.4. Persistence and LRTP

Chemical persistence is commonly represented by $P_{OV}$, and is the overall residence time (d) of the chemical in the environment (Iqbal et al., 2013). LRTP was calculated as the ratio of a chemical exported