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Urban and rural transport of semivolatile organic compounds at regional scale: A multimedia model approach

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ABSTRACT

Urban areas are generally regarded as major sources of some semivolatile organic compounds and other persistent organic pollutants (POPs) to the surrounding regions. Huge differences in contaminant emissions between urban and rural areas directly affect their fate in environmental media. Little is known about POPs behavior between urban and rural areas at a regional scale. A spatially resolved Berkeley-Trent-Urban-Rural Fate Model (BETR-UR) was designed by coupling land cover information to simulate the transport of POPs between urban and rural areas, and the Bohai Rim was used as a case study to estimate Polycyclic Aromatic Hydrocarbon (PAH) fate. The processes of contaminant fate including emission, inter-compartmental transfer, advection and degradation in urban and rural areas were simulated in the model. Simulated PAH concentrations in environmental media of urban and rural areas were very close to measured values. The model accuracy was highly improved, with the average absolute relative error for PAH concentrations reduced from 37% to 3% compared with unimproved model results. PAH concentrations in urban soil and air were considerably higher than those in rural areas. Sensitivity analysis showed temperature was the most influential parameter for Phen rather than for Bap, whose fate was more influenced by emission rate, compartment dimension, transport velocity and chemical persistence. Uncertainty analysis indicated modeled results in urban media had higher uncertainty than those in rural areas due to larger variations of emissions in urban areas. The differences in urban and rural areas provided us with valuable guidance on policy setting for urban–rural POP control.

Introduction

In recent decades, contamination by persistent organic pollutants (POPs) has brought great concern from international environmental organizations, governments, academia, and the public. Assessment of the state and impacts of POPs is very important for environmental management, especially for regional ecological and human health risk management of POPs (Diamond and Hodge, 2007; Lindstrom et al., 2011). However, the availability of large-scale monitoring data in all environmental compartments (soil, water, vegetation, and air) is spatially and temporally limited. Therefore, application of multimedia fate models, which can simulate the concentrations, distributions, and persistence of chemicals in the environment based on mass balance equivalents (MacLeod et al., 2001; Liu et al., 2011, 2014), provides a useful tool for simulating the environmental behavior of chemicals.

Urban areas are generally regarded as major sources of some semivolatile organic compounds (SVOCs) and other POPs to the surrounding regions. These pollutants originate from, for example, production and use of chemicals, vehicle

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exhaust, and building materials. (Csiszar et al., 2014). Some chemical pollutant emissions (*e.g.*, PAHs, PBDEs) in urban areas may be several times or even thousands of times higher than those in suburbs, which may lead to chemical concentrations in urban soil several times or even tens of times higher than in rural soil (Wang et al., 2010; Jiao, 2009). Hence, huge differences in contaminant emissions between urban and rural areas directly affect their fate in environmental media. However, little work has been related to the behavior of POPs between urban and rural areas at a regional scale (Csiszar et al., 2013). When the fate of chemicals has been simulated at global or continental scale, the urban and rural areas were usually regarded as a whole. For example, the Berkeley–Trent fate model (BETR model) has been applied to model the multimedia fate of chemicals, including the North America (MacLeod et al., 2001; Woodfine et al., 2001), Europe (Prevedouros et al., 2004a, 2004b), and global environment (Scheringer et al., 2000; Macleod et al., 2005; Armitage et al., 2009). However, if urban and rural areas are taken as a whole at a regional scale, the differences in chemical behaviors between urban and rural areas cannot be well illustrated, and simulated concentrations in urban areas might be excessively undervalued due to the relatively high emissions in urban areas (Melymuk et al., 2011). Chemical concentrations that are not differentiated between urban and rural areas cannot really reflect the regional pollution level and ecological risk, especially for urban regions. Hence, modeling the interactions of POPs between rural and urban areas is important for the purposes of POP ecological risk assessment and control.

Models that can be used to simulate POP interactions between rural and urban areas are scarce. The Multimedia Urban Model (MUM) and Spatially Oriented MUM (SO-MUM) were used to simulate the fate of POPs from urban to suburban areas in Canada (Diamond et al., 2001; Csiszar et al., 2013). However, these two models focused on the intra-urban scale and the effects of impervious surfaces in urban area. Also, the BETR model is a spatially segmented multimedia model (Mackay, 1979, 2001) based on a fugacity model approach, which has been used to model chemicals including Toxaphene, PCBs, PAHs, PBDEs, and HCHs (MacLeod et al., 2001; Prevedouros et al., 2004a, 2004b; Toose et al., 2004). The BETR modeled result was the spatial average concentration in one sub-region, which may represent the average concentrations in various land use types such as urban land and farm land. Actually, the emissions of PAHs were considerably different between urban and rural areas (Shen et al., 2013). The BETR model application in the Chinese Bohai coastal region showed that the modeled concentrations of Benzo[α]pyrene (BaP) in air, fresh water, soil, and sediment generally agreed with field observations except for the soil concentration of urban areas, and the measured concentration of soil obtained from Beijing, one of the most developed cities in China, was 7 times higher than the modeled value (Liu et al., 2014). Due to the huge differences in PAH emissions between urban and rural areas, the results in urban soils were actually several times higher than the concentrations in rural soils (Wang et al., 2010; Jiao, 2009). The factors affecting the fate of PAHs in urban and rural areas include emissions, vegetation cover, soil properties, and atmospheric aerosols. In particular, atmospheric aerosols in urban air were found to be a few times or even tens of times higher than in rural air (Mackay, 2001). So, it is necessary to consider the differences in the above parameters in the two areas.

Based on the steady-state BETR model, this study developed a spatially resolved multi-media BETR-Urban–Rural (BETR-UR) model, which took the effects of land uses into account, dividing the soil and lower air compartments into urban soil and rural soil, lower urban air and lower rural air, respectively. The urban areas mainly indicated those areas with high population density, including industrial land use, commercial land use, urban residential land use, municipal land for public facilities, and their buffers. In the rural areas, land uses include rural villages, agricultural land, grassland, forest land, rural residential land, unused land and so on (Wang et al., 2010). Moreover, the complex inter-media transport processes were optimized, distinguishing the emissions from urban and rural areas effectively. In this study, the coastal region of the Bohai Sea was selected as a case study for evaluating the model accuracy and adaptability, and for simulating the multimedia fate of PAHs (Benzo[α]pyrene and Phenanthrene, BaP and Phen) in environmental compartments.

1. Materials and methods

1.1. *Description of optimized model structure*

Within the original BETR model (MacLeod et al., 2001), a connected system of seven discrete and homogeneous compartments is considered as one segment (or grid). The seven environmental compartments contain upper air, lower air, vegetation, soil, sediment, fresh water, and coastal water. Full details for using the fugacity concept in contaminant fate models are available in the text by Mackay (2001). For a given region, the more segments are divided, the more complex the emission data and background information needed. Thus, under the original framework, a region is usually divided into a relatively small number of grids (*e.g.*, <100) and a larger range (*e.g.*, >10 km × 10 km) for describing national, provincial, or urban agglomeration scales. The simulated results of homogeneous soil and air in one cell were often unable to meet the needs of risk assessment, because huge variability between the urban and suburban areas was ignored.

Fig. 1 illustrates the nine compartments in the optimized BETR-UR model framework. In the BETR-UR model, the environment within each segment containing nine compartments is appropriate for distinguishing the urban and rural areas on a regional scale. The red arrows represent the added intermedia transfer processes (Fig. 1). Among those processes, the interactions between rural air and urban air are the most significant and need to be carefully considered. Although the processes of flow transfer and mass diffusion in segments or among segments may become complex, parameterization of urban areas is relatively easy. Specific environmental parameters include urban area, urban perimeter, urban–rural atmospheric mixing rate, vegetation coverage area, freshwater area of urban area, and particle fraction in urban air. Other parameters also could be edited if needed, *e.g.*, water and solid runoff mass transfer coefficient (MTC), leaching from soil rate MTC, soil solid organic carbon, and volume fractions of soil water, air and solid.