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污染地块 VOCs 源衰减对室内蒸气入侵风险的影响

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摘要: 我国挥发性有机物(VOCs)污染地块普遍采用 J&E 模型预测蒸气入侵风险, 该模型假定污染源含量在整个暴露周期内恒定, 与其在地块中的客观变化规律不符. 以某 VOCs 污染地块为例, 采用 J&E 恒定源模型、SD 衰减源模型及 RBCA 衰减源模型, 分别预测 VOCs 侵入建筑物室内的质量浓度及蒸气入侵风险. 结果显示, J&E 预测暴露期内的源含量及室内污染物质量浓度始终较高, SD 和 RBCA 衰减源模型预测显示两者均呈指数下降. RBCA 衰减源模型预测的源衰减更快, 但其室内污染物质量浓度预测结果小于 SD 衰减源模型预测结果. SD 模型中建筑物室内外压差是影响源衰减的关键参数, 压差增大, 污染物以对流方式侵入室内, 源衰减速率增加. 压差降低, 污染物以扩散方式侵入室内, 建筑物对源衰减的作用减弱, 预测结果与 J&E 模型差异不明显. J&E 模型预测的致癌风险和危害商最高, SD 次之, RBCA 最低. 因此, J&E 模型易高估暴露期内蒸气入侵风险, RBCA 衰减源模型未考虑建筑物对源衰减的阻滞而低估风险, SD 模型考虑了建筑物对源衰减的影响, 更适用于评估实际场地的室内蒸气入侵风险.

关键词: 挥发性有机物(VOCs); 蒸气入侵; 源衰减; 风险评估; SD 模型

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Effects of Source Depletion on Vapor Intrusion Risk Assessment

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Abstract: The current regulatory site investigation employs the J&E model to predict vapor intrusion risk. However, the J&E model assumes that the source concentration is constant for a given exposure period, which is not consistent with the actual site source under depletion. In this study, we compared the differences between the J&E model (constant source), SD source depletion model, and RBCA source depletion model for predicting indoor concentration variation as well as the risk levels during the exposure period with a case study in Beijing. The results showed that the source and indoor air concentrations predicted by the SD and RBCA models showed exponential decreases, whereas those predicted by the J&E model maintained high concentrations throughout the exposure period, which greatly overestimated the risk. The RBCA predicted source depletion at the fastest rate, but the predicted indoor air concentrations were still lower than those of the SD model, which was related to the fact that the RBCA did not consider the effect of buildings on source depletion and did not follow mass conservation. Further, the sensitivity analysis showed that the pressure difference (ΔP) had the greatest influence on the source concentration in the SD model. For the calculated carcinogenic risk and hazard quotients, the J&E constant source model, the SD source depletion model, and the RBCA source depletion model were ranked in descending order. The results indicated that in general the J&E model was too conservative, the RBCA model may have underestimated risk, and the SD model was more suitable for quantifying vapor intrusion risk in reality.

Key words: volatile organic compounds(VOCs); vapor intrusion; source depletion; risk assessment; SD model

土壤中的挥发性有机物(Volatile organic compounds, VOCs)可通过扩散或对流经建筑物地板裂隙等通道进入室内造成空气污染, 该过程被称为“蒸气入侵”, 是危害人群健康的重要途径^[1~5]. 我国2005~2019年公开的136处有机污染地块土壤和地下水中VOCs检出比例分别高达65%和92%^[6], 69个城市的地下水中三氯甲烷检出率20.23%^[7], 13个采油区494个土壤点位中石油烃等VOCs超标点位23.6%^[8], 美国超级基金地块中约78%存在VOCs污染^[9]. 20世纪90年代美国丹佛市Redfield地块中二氯乙烯和三氯乙烯垂向扩散迁移造成300多栋住宅室内空气超标, 引起了各国对VOCs污染地块健康风险的重视^[10~12].

Johnson & Ettinger (J&E) 模型^[13~15]是目前国

内外运用最为广泛的模型, 该模型假设: ①VOCs在土壤固-液-气相之间为线性分配; ②VOCs垂向迁移过程无生物降解; ③污染源含量不随时间变化^[16~18]. 上述假设与地块中VOCs的实际分配传输过程存在较大差异: ①VOCs在土壤中的界面分配属于非线性^[19]; ②传输过程存在挥发和生物降解等过程^[20,21]; ③整个暴露周期内污染源在逐步衰减^[22]. 针对假设①, Zhang等^[23,24]在双相非线性吸附理论的基础上建立了多相非线性分配模型; 针对

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假设②, DeVuall^[20]开发了一维生物降解模型(Bio vapor)以量化 VOCs 垂向迁移过程中的生物降解过程, Yao 等^[25,26]开发了二维生物降解模型(PVI2D)用于量化石油烃类污染物在包气带迁移过程的生物降解. 针对假设③, 美国材料与试验协会(American Society for Testing and Materials, ASTM)在 J&E 恒定源模型的基础上提出了基于风险的矫正行动(Risk-Based Corrective Action, RBCA)的衰减源模型^[27], 该模型假定地表空气中 VOCs 质量浓度为 0 来计算源衰减速率. 但当地面存在建筑物时, 建筑底板下土壤中 VOCs 质量浓度通常不等于 0, 污染源至建筑物地板的质量浓度梯度小于 RBCA 假设地表质量浓度为 0 时的梯度, 导致存在建筑情景下 VOCs 的衰减速率降低. 此外, 建筑物室内外压差直接影响源衰减速率^[28~30], 但 RBCA 源衰减模型未考虑建筑物参数的影响. 针对 RBCA 衰减源模型的局限, 笔者团队

在对蒸气入侵的 3 个典型物理过程(即污染物从污染源扩散至建筑底板下、通过对流/扩散由底板裂隙进入室内空间、与室内外空气混合交换)分别进行数学描述的基础上, 基于通量守恒原理开发了 SD 衰减源模型^[22]. 本研究以某 VOCs 污染地块为例, 进一步分析了不同模型对蒸气入侵风险评估结果的影响, 以期完善我国 VOCs 污染地块风险评估提供技术支撑.

1 材料与方法

1.1 案例地块概述

案例地块占地面积约 0.09 km², 1979~2014 年主要生产各种型号沥青混合物, 原辅材料包括沥青、重油、柴油、导热油、砂石料、矿粉、改性剂和乳化剂等. 在重点污染区共设置了 83 个土壤采样点, 按规范采集检测土壤样品 736 个, 具体采样点分布如图 1.

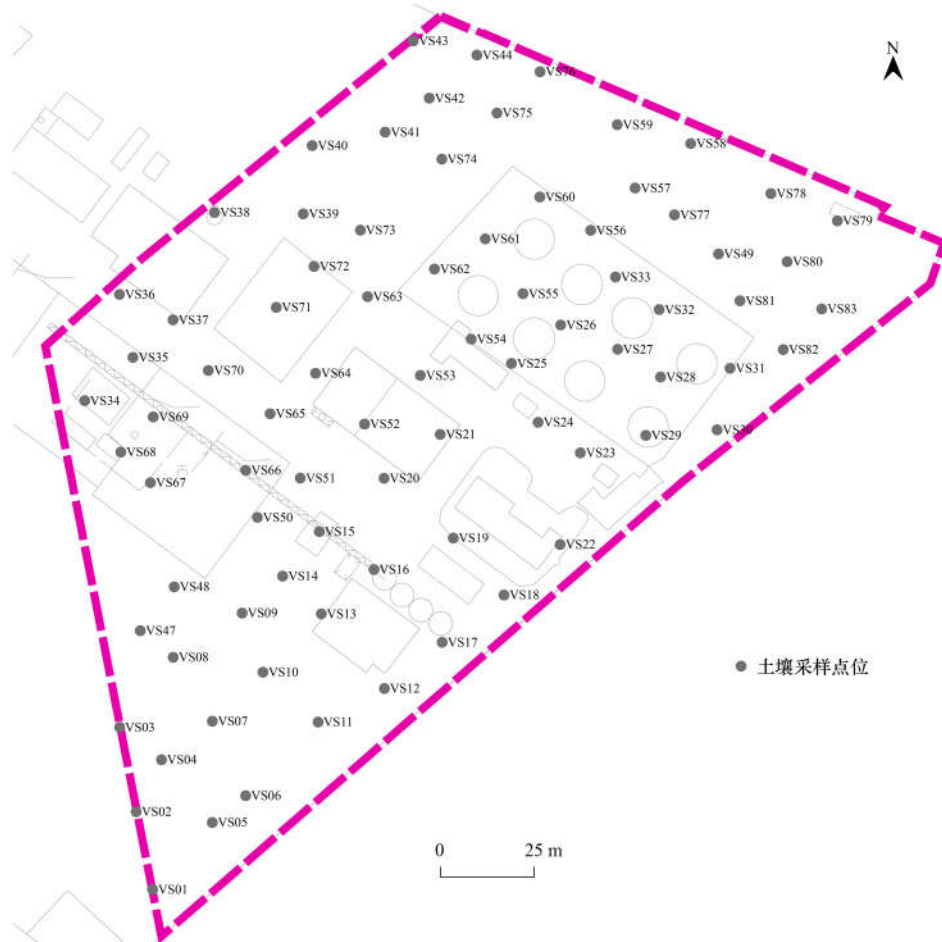


图 1 土壤采样点分布

Fig. 1 Distribution of soil sampling locations

1.2 风险评估

1.2.1 室内质量浓度预测

分别采用 J&E 恒定源模型^[13,31]、RBCA 衰减源模型^[27]和笔者团队开发的 SD 衰减源模型^[22]预测

VOCs 侵入建筑物室内的质量浓度 C_{in} , 计算公式如下.

J&E 源恒定模型:

$$C_{in} = \alpha \times C_{source} \quad (1)$$

RBCA 衰减源模型:

$$C_{in}(t) = \alpha \times C_{source}(t) \quad (2)$$

SD 衰减源模型:

$$C_{in}(t) = \alpha \times C_{source}(t) \quad (3)$$

以上模型中污染源处土壤气质量浓度 C_{source} 的计算公式分别为:

J&E 源恒定模型:

$$C_{source} = \beta \times C_0 \quad (4)$$

RBCA 衰减源模型:

$$C_{source}(t) = C_0 \times \exp\left[-\left(\frac{D_s^{eff} \times \beta}{h \times \rho \times L_T}\right) \times t\right] \quad (5)$$

SD 衰减源模型:

$$C_{source}(t) = C_0 \times \exp\left[-\left(\frac{\lambda \times \beta}{A_{source} \times h \times \rho}\right) \times t\right] \quad (6)$$

模型中涉及的过程参数计算公式为:

$$\alpha = \left[\left(\frac{D_s^{eff} \times A_b}{Q_{indoor} \times L_T} \right) \exp\left(\frac{Q_{soil} \times L_{crack}}{D_{crack} \times A_{crack}} \right) \right] \div$$

$$\left\{ \exp \frac{Q_{soil} \times L_{crack}}{D_{crack} \times A_{crack}} + \left(\frac{D_s^{eff} \times A_b}{Q_{indoor} \times L_T} \right) + \left(\frac{D_s^{eff} \times A_b}{Q_{soil} \times L_T} \right) \left[\exp\left(\frac{Q_{soil} \times L_{crack}}{D_{crack} \times A_{crack}} \right)^{-1} \right] \right\} \quad (7)$$

$$\lambda = \frac{Q_{soil} \times \exp(Pe)}{L_T \times Q_{soil} \times \exp(Pe) + \frac{Q_{soil}}{A_b \times D_s^{eff}} + \frac{Q_{soil}}{Q_{indoor}} + \exp(Pe) - 1} \quad (8)$$

$$\beta = \frac{H \times \rho}{\theta_{water} + H \times \theta_{air} + K_d \times \rho} \quad (9)$$

$$Pe = Q_{soil} \times L_{crack} / (D_{crack} \times A_{crack}) \quad (10)$$

$$D_s^{eff} = D_{air} \times \frac{\theta_{air}^{3.33}}{\theta^2} + D_{water} \times \frac{\theta_{water}^{3.33}}{\theta^2} \quad (11)$$

$$Q_{soil} = \frac{2 \times \pi \times K_v \times X_{crack} \times dP}{\mu_{air} \times \ln\left(\frac{2 \times L_{crack}}{R_{crack}}\right)} \quad (12)$$

$$Q_{indoor} = H_b \times A_b \times ER \times \frac{1}{3600} \quad (13)$$

上述 J&E 模型假定源含量恒定, C_{source} 不随时间变化, 见式(4). RBCA 衰减源模型假设地表无建筑物, VOCs 扩散进入地表大气被稀释是源衰减的主要驱动力. 同时, 假设大气快速扩散使地表 VOCs 质量浓度为 0. 在此基础上采用 Fick 第一定理计算挥发导致的 VOCs 源衰减, 建立随时间 t 变化的源含量 $C_{source}(t)$ 的计算公式, 见式(5). SD 衰减源模型结合实际场地蒸气入侵的物理过程, 假定 VOCs 通过建筑地板侵入室内后与室内外空气混合稀释是污染源衰减的主要原因, 基于通量守恒原理建立随时间 t

的源含量 $C_{source}(t)$ 的计算公式, 见式(6). 可见, SD 衰减源模型充分考虑了建筑物特征对源衰减的影响. 由于 RBCA 衰减源模型未考虑建筑物对源衰减过程的影响, 其预测的源衰减速率与存在建筑物情形下的源衰减速率不一致, 因此即使不考虑在土层中扩散过程的生物降解, 此模型实际上也违背了质量守恒原则.

SD 衰减源模型中各参数的定义及取值如表 1 所示, 其中建筑物和污染物性质等参数取自《建设用地土壤污染风险评估技术导则》(HJ 25.3-2019)^[31], 土壤理化参数取自地块实测值. 值得注意的是, J&E 恒定源模型计算建筑物室内污染物质量浓度时表中所有参数都被用到, RBCA 衰减源模型只用到了前 3 类参数, 未涉及建筑物参数, SD 衰减源模型用到了全部参数, 但与国内目前推荐的 J&E 恒定源模型相比并未增加新的参数.

1.2.2 风险计算

(1) 致癌风险

衰减源情景下, 室内污染物质量浓度 $C_{indoor}(t)$ 随时间发生变化, 根据致癌风险定义^[31], 可推导致癌风险 (Risk) 的计算公式:

$$\text{Risk} = \frac{SF}{BW_c \times AT_{ca}} \times \int_0^6 \text{DAIR}_c \times EF_c \times C_{in}(t) dt + \frac{SF}{BW_a \times AT_{ca}} \times \int_6^{30} \text{DAIR}_a \times EF_a \times C_{in}(t) dt \quad (14)$$

式中, $\int_0^6 \text{DAIR}_c \times EF_c \times C_{in}(t) dt$ 为给定暴露年限下儿童呼吸摄入污染物的总量, $\int_6^{30} \text{DAIR}_a \times EF_a \times C_{in}(t) dt$ 为给定暴露年限下成人呼吸摄入污染物的总量.

污染源恒定时, 上式可简化为:

$$\text{Risk} = C_{in} \times \left(\frac{\text{DAIR}_a \times EF_a \times ED_a}{BW_a \times AT_{ca}} + \frac{\text{DAIR}_c \times EF_c \times ED_c}{BW_c \times AT_{ca}} \right) \times SF \quad (15)$$

(2) 非致癌危害商

非致癌危害商 (HQ) 计算以儿童 (0~6 岁) 为敏感人群, 按照危害商的定义^[31], 衰减源情景下计算公式如下:

$$\text{HQ} = \left[\frac{1}{BW_c \times ED_c \times 365} \times \int_0^6 \text{DAIR}_c \times EF_c \times C_{in}(t) dt \right] \div \text{RfD} \quad (16)$$

污染源恒定时, 上式可简化为:

表 1 参数取值

Table 1 Values of model parameters

参数分类	参数符合	参数全称	单位	取值
苯	C_0	污染源土壤中污染物含量	$\text{mg}\cdot\text{kg}^{-1}$	30.5
	H	亨利常数	— ¹⁾	0.227
	D_{air}	在空气中扩散系数	$\text{cm}^2\cdot\text{s}^{-1}$	0.089 5
	D_{water}	在水中扩散系数	$\text{cm}^2\cdot\text{s}^{-1}$	1.03×10^{-5}
	K_{oc}	有机碳分配系数	$\text{L}\cdot\text{kg}^{-1}$	146
污染源	L_{T}	污染源深度	m	13
	A_{source}	污染源顶部面积	m^2	1 500
	h	污染源厚度	m	5
土壤	ρ	土壤容重	$\text{kg}\cdot\text{m}^{-3}$	1 700
	θ_{air}	土壤孔隙气体积比	—	0.33
	θ_{water}	土壤孔隙水体积比	—	0.08
	θ	土壤孔隙体积	—	0.41
	f_{oc}	土壤有机碳含量	—	0.01
	K_{v}	底板裂隙中蒸气渗透率	m^2	1×10^{-12}
	θ_{acrack}	底板裂隙土壤孔隙气体积比	—	0.33
	θ_{wcrack}	底板裂隙土壤孔隙水体积比	—	0.08
	L_{crack}	底板厚度	m	0.6
建筑物	A_{b}	底板面积	m^2	1 500
	η	裂隙率	—	0.000 5
	dP	室内外压差	Pa	5
	R_{crack}	裂隙宽度	m	0.005
	H_{b}	建筑物混合高度	m	2.44
	ER	室内外换气率	h^{-1}	0.25

1) “—”表示无量纲

$$\text{HQ} = C_{\text{in}} \times \frac{\text{DAIR}_{\text{c}} \times \text{EF}_{\text{c}} \times \text{ED}_{\text{c}}}{\text{BW}_{\text{c}} \times \text{ED}_{\text{c}} \times 365 \times \text{RfD}} \quad (17)$$

本研究可接受致癌风险水平设为 10^{-6} , 可接受危害商设为小于 1^[32-35]. 暴露参数取 HJ 25.3-2019^[31] 中第一类用地推荐值. 其中, 儿童期暴露周期 (ED_{c}) 为 6 a, 成人期 (ED_{a}) 为 24 a, 致癌风险作用时间 (AT_{ca}) 为 27 740 d, 非致癌作用时间 (AT_{nc}) 为 2 190 d, 儿童暴露频率 (EF_{c}) 和成人暴露频率 (EF_{a}) 均为 $250 \text{ d}\cdot\text{a}^{-1}$, 成人体重 (BW_{a}) 为 65 kg, 儿童平均体重 (BW_{c}) 为 19.2 kg, 成人空气呼吸率 (DAIR_{a}) 为 $14.5 \text{ m}^3\cdot\text{d}^{-1}$, 儿童 DAIR_{c} 为 $7.5 \text{ m}^3\cdot\text{d}^{-1}$. 毒性参数 SF 为 $5.5\times 10^{-2} (\text{kg}\cdot\text{d})\cdot\text{mg}^{-1}$, RfC 为 $0.03 \text{ mg}\cdot\text{m}^{-3}$.

1.3 敏感性分析

因 RBCA 衰减源模型未考虑建筑物对源衰减的影响, 为便于对比讨论, 本研究仅分析了 SD 衰减源模型中建筑物特征参数对源含量的影响. 敏感性分析采用 @ RISK 软件中的蒙特卡洛方法^[36,37], 建筑物特征参数分布函数见表 2. 以 30 a 为暴露周期, 对暴露周期内的每一年进行 100 000 次的模拟运算. 以斯皮尔曼秩相关系数作为各参数影响的比重^[38], 计算公式如式 (18) 所示:

$$P_i = \omega_i / \sum_{i=1}^n |\omega_i| \quad (18)$$

式中, ω_i 为参数 X_i 的比重, 即斯皮尔曼秩相关系数, 无量纲; P_i 为参数 X_i 对输出参数 Y 不确定性的贡献, 无量纲.

表 2 敏感性分析参数取值

Table 2 Parameter distributions for sensitive analysis

建筑物参数	概率分布函数	特征值 ¹⁾	文献
A_{b}	Uniform	min = 80, max = 2 000	[39]
η	Uniform	min = 0.0005, max = 0.01	[39]
dP	Uniform	min = 0, max = 20	[39]
R_{crack}	Power function	min = 0.000 5, max = 0.005	[39]
H_{b}	Uniform	min = 2.1, max = 3.0	[40]
L_{crack}	Uniform	min = 0.15, max = 0.8	[40]
ER	Uniform	min = 0.1, max = 1.5	[39,41,42]

1) min 为最小值, max 为最大值

2 结果与讨论

2.1 污染特征

案例地块土壤中苯含量 $0\sim 30.7 \text{ mg}\cdot\text{kg}^{-1}$, 与居住用地土壤苯筛选值 $1 \text{ mg}\cdot\text{kg}^{-1}$ ^[43] 进行对比发现, 样品超标率 14.02%, 最大超标 30 倍. 如图 2(a) 所示, 案例地块土壤苯为局部污染, 主要分布在加油站和配油车间所在区域. 如图 2(b) 所示, 随深度增加土壤中苯含量升高, 含量最高的样品位于在地面以下 15 m 附近的土壤中, 原因是该地块地面以下 13~18 m 深度存在一层厚度不一且不连续的低渗

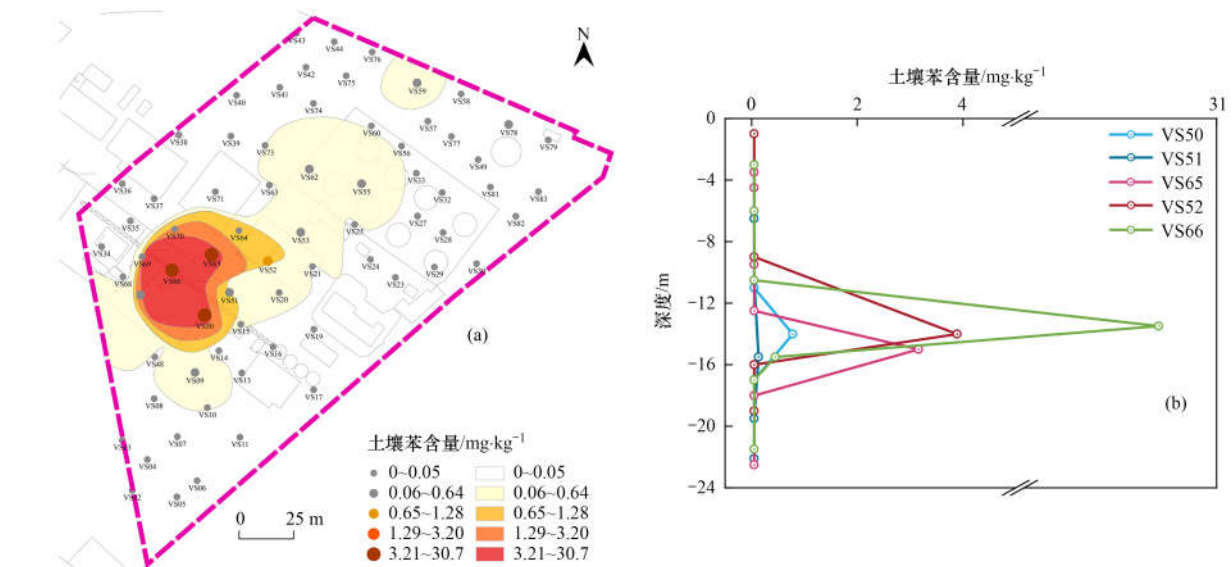


图2 土壤中苯空间分布

Fig. 2 Spatial distribution of benzene in soil

透土层,易吸附污染物,导致苯易在这一深度范围内的土壤中富集.

2.2 建筑物对源衰减的影响

案例地块污染区修复合格后拟建设一栋带有一层地下室的高层住宅. 由于苯检出的土样主要集中在地面以下 13~18 m 且在地下水位之上的包气带, 因此选定距地表 13~18 m 深度的污染土壤作为蒸气入侵的污染源,其顶部埋深设置为 13 m. 考虑到未来建筑地下室层高 3.1 m, 建筑物底板厚度 0.6 m, 则污染源顶部距未来建筑底板的距离为 9.3 m. 污染源长和宽分别为 50 m 和 30 m, 因此污染源面积为1 500 m². 由图 3 可知, 对 SD 衰减源模型源含量预测结果影响最大的建筑物参数为室内外压差, 其次是底板裂隙率和底板面积, 均与源含量呈负相关. 底板裂隙宽度对源含量也有较大影响, 但与源含量呈正相关. 其余参数对源含量的影响较小^[44]. 图

4 进一步显示了建筑物室内外压差对源衰减的影响. 压差越大,SD 衰减源模型预测的源衰减速率越快,预测结果与 RBCA 衰减源模型预测结果越接近. 相反,随着压差增大,SD 衰减源模型预测结果与 J&E 恒定源模型预测结果差异增加. 因此,室内外压差较大的情况下,现有导则中推荐的 J&E 恒定源模型计算结果更保守.

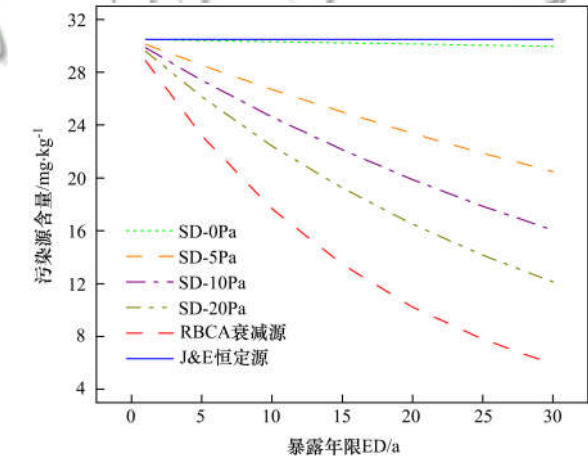


图4 不同模型预测的污染源处苯含量衰减速率

Fig. 4 Benzene depletion rates at source predicated by the different models

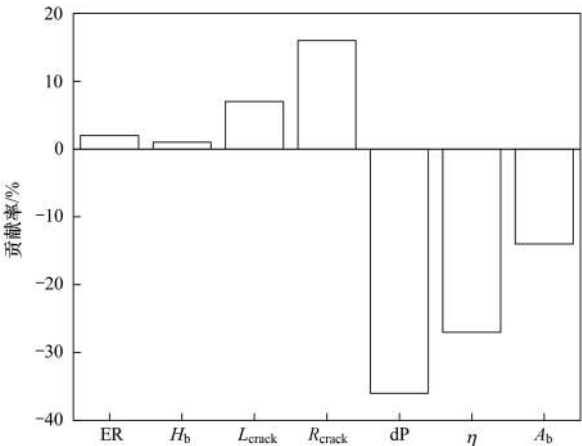


图3 建筑参数对 SD 模型计算得到的源含量不确定性的贡献率

Fig. 3 Contribution of building parameters to the uncertainty of source concentration calculated by the SD model

2.3 室内质量浓度及风险

不同模型预测的室内质量浓度变化如图 5 所示, J&E 恒定源模型计算的室内质量浓度在整个暴露周期内保持不变,SD 和 RBCA 衰减源模型计算的室内质量浓度呈指数形式下降,且建筑物室内外压差越大,SD 衰减源模型预测的室内质量浓度越高. 尽管 RBCA 衰减源模型预测的源衰减速率更快,即单位时间挥发的污染物的质量更多,但该模型预测的室内质量浓度却低于 SD 衰

减源模型预测结果,这是因为 RBCA 衰减源模型中污染源衰减的质量不等于进入室内的污染物的质量,即模型不遵循质量守恒^[22,27]. SD 和 RBCA 衰减源模型计算的污染物室内质量浓度下降趋势与污染源处污染物含量衰减规律一致,均

表现为质量浓度随时间呈指数下降,主要原因是两者都是采用污染源处土壤气质量浓度乘以衰减因子的方法预测污染物室内质量浓度^[13,22,27],且污染源处土壤气质量浓度衰减均为指数模型[见式(5)和式(6)].

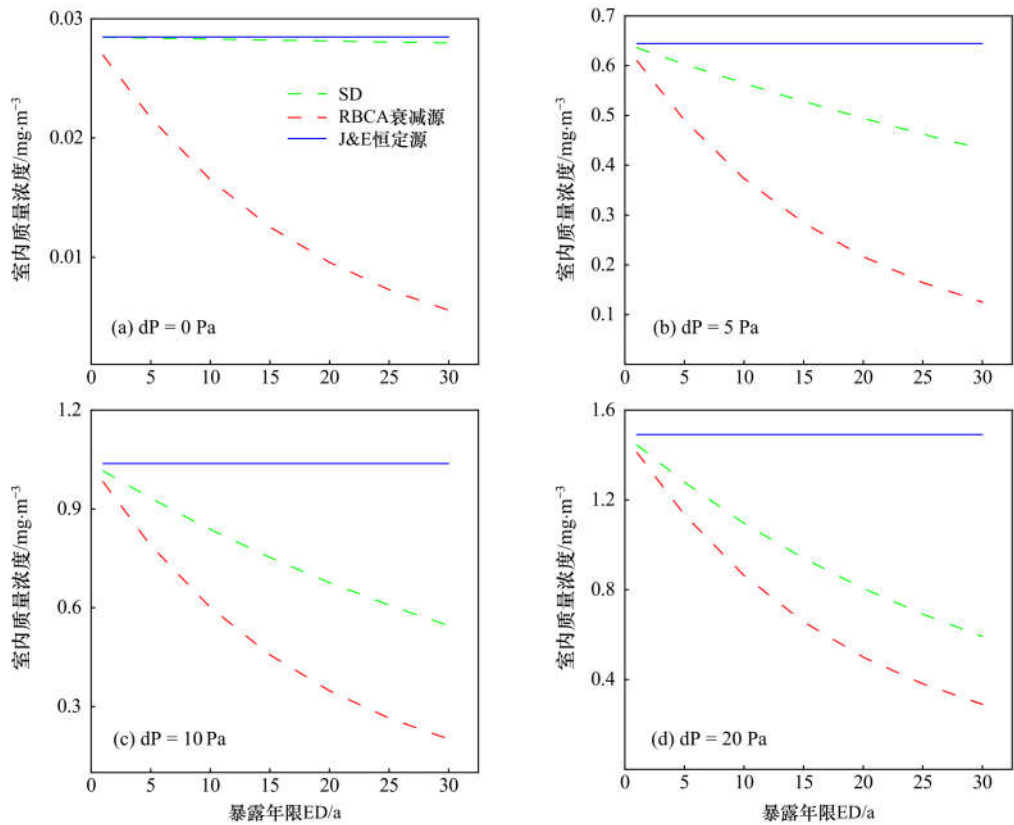


图5 不同压差下室内空气苯质量浓度预测结果

Fig. 5 Indoor concentration of benzene predicted by the models at different pressure differences

如表3所示,本研究中的3个模型预测的苯致癌风险均高于可接受水平 10^{-6} ,非致癌危害商也超过了可接受值1. J&E模型因假定源恒定^[13],预测的室内苯质量浓度始终维持初始的较高水平,健康风险计算结果最高,其次为SD衰减源模型,RBCA衰减源模型预测的健康风险最低.主要原因是RBCA模型不遵循质量守恒,尽管其预测污染源处

苯挥发损失的质量最大,但实际预测进入室内的污染物质量小于挥发损失的质量,导致源含量快速衰减但并没有同步增加室内苯质量浓度,可能低估风险.压差是影响室内蒸气入侵风险的最主要因素,压差增加,3个模型预测的室内风险均升高,主要原因是压差增加导致污染物进入室内空间的速率增加,在室内换气速率固定的情形下污染物在室内空气中

表3 蒸气入侵风险评估结果

Table 3 Results of vapor intrusion risk

室内室外气压差/Pa	项目	SD 衰减源模型	RBCA 衰减源模型	J&E 恒定源模型
0	致癌风险	1.56×10^{-4}	8.19×10^{-5}	1.58×10^{-4}
	危害商	1.5	1.3	1.5
	修复目标值	1.94×10^{-1}	3.71×10^{-1}	1.93×10^{-1}
5	致癌风险	2.98×10^{-3}	1.85×10^{-3}	3.57×10^{-3}
	危害商	33	29	34
	修复目标值	1.02×10^{-2}	1.64×10^{-2}	8.52×10^{-3}
10	致癌风险	4.63×10^{-3}	3.28×10^{-3}	6.31×10^{-3}
	危害商	56	50.4	60.7
	修复目标值	6.56×10^{-3}	9.26×10^{-3}	4.81×10^{-3}
20	致癌风险	5.56×10^{-3}	4.29×10^{-3}	8.25×10^{-3}
	危害商	71	66	79
	修复目标值	5.47×10^{-3}	7.08×10^{-3}	3.68×10^{-3}

聚集,导致室内浓度及健康风险升高。但当建筑底板下与室内的压差较小时,SD 衰减源模型预测的风险与 J&E 恒定源模型预测结果差异不明显,主要原因是压差降低污染物进入室内的速率降低,污染源衰减速率也随之下降,衰减作用不明显。随着压差增加,SD 衰减源模型预测的室内蒸气入侵风险更接近 RBCA 衰减源模型的预测结果。因此,目前国家导则 HJ 25.3-2019 中采用的 J&E 恒定源模型预测的 30 a 暴露周期内室内蒸气入侵总暴露量将大大高于衰减源模型预测结果。相比 SD 和 RBCA 衰减源模型,建筑物底板下和室内的压差越大,J&E 恒定源模型高估室内污染物质量浓度及风险越显著^[45],导致计算的土壤修复目标值更低,易造成土壤过度修复。

3 结论

(1) 室内外压差是影响 VOCs 源衰减速率的关键建筑物特征参数,压差越大,衰减越快,源含量越低,SD 衰减源模型计算结果与 J&E 恒定源模型差异越明显,但与 RBCA 衰减源模型计算结果越接近。

(2) RBCA 和 SD 衰减源模型均能反映污染源衰减对室内蒸气入侵风险的影响,一定程度上解决了 J&E 恒定源模型过于保守的问题。因考虑了建筑物对源衰减的影响,SD 衰减源模型更适于评估实际场地的蒸气入侵风险。

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