# COMPARISON OF GAUSSIAN AND LAGRANGIAN PUFF DISPERSION MODELS FOR THE RISK ASSESSMENT OF RECEPTORS NEARBY A CONTAMINATED SITE

MARCO RAVINA[1], IASON VERGINELLI[2], RENATO BACIOCCHI[2] & MARIACHIARA ZANETTI[1]
[1]Department of Environment, Land and Infrastructure Engineering, Turin Polytechnic, Italy.
[2]Department of Civil Engineering and Computer Science, University of Rome "Tor Vergata", Italy.

ABSTRACT

Human health risk assessment for off-site receptors located in the proximity of a contaminated site is based on the application of pollutant atmospheric dispersion models. In the standard ASTM Risk-Based Corrective Action (RBCA-ASTM) procedure, this evaluation is carried out by coupling a one-dimensional Gaussian dispersion model to a simple dilution box model. In this work, the accuracy of this approach is examined by comparing the output obtained by the standard Gaussian box model with the results obtained with the non-steady-state Lagrangian puff dispersion model CALPUFF. A case study was considered, assuming the emission of benzene from a contaminated area of 200 × 200 m on flat terrain. The comparison of concentration profiles as a function of the distance from the source showed that the standard procedure overestimated concentrations by more than one order of magnitude. Two possible refinements to the standard RBCA-ASTM procedure were suggested. The first is the introduction of an equivalent mixing height for the application of the box model, calculated on the basis of the atmospheric stability class, land use typology, and dimension of the source. The second is the consideration of the wind distribution of the area. The introduction of these modifications allowed to reduce the discrepancy between the Gaussian box model and CALPUFF. This study also showed that the use of advanced dispersion models integrated with the risk calculation methodologies, allowed a detailed characterization of the risk in the area under examination, highlighting the most critical areas and comparing them with the presence of any sensitive receptors.

*Keywords: atmospheric dispersion modelling, concentration exposure, contaminated site, human health risk assessment.*

## 1 INTRODUCTION

Macro- and micro-pollutants emitted into the lower levels of the atmosphere pose a serious threat, as they contribute in generating adverse consequences on human health and ecosystems [1]. Health impact assessment (HIA) methodologies have been developed to evaluate the impacts of pollution on human health. If urban air quality problems are studied, HIAs are often based on epidemiologic criteria, in which the concentration of different species (e.g. $NO_x$, CO, $SO_2$, and particulate matter) is associated to a response function [2]. Otherwise, if a contaminated site is considered, the impact of micro-pollutants (e.g. petroleum hydrocarbons, chlorinated solvents, PCBs) inhalation of nearby receptors is assessed by mean of sanitary health risk analysis (HRAs). The latter is based on toxicological studies, which define the limits of exposure concentrations for carcinogenic and toxic substances [3].

Both HIAs and HRAs are well-known topics among the scientific community. Decades of toxicological, clinical, and epidemiological research support the tools and methodologies currently applied [4, 5]. Several modelling tools have been developed in support of HIA and HRA studies. The great majority of these tools is based on the application of pollutant dispersion models.

This study is focused on HRAs. Currently, the most acknowledged technical and scientific reference for HRA methodology on polluted sites is the ASTM Risk-Based Corrective


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Action (RBCA) standards (E 2081-00) [6]. The RBCA methodology is based on a tiered approach, with increasing complexity in the definition of the site conceptual model and in the description of the physical and chemical phenomena underlying the fate and transport of contaminants. Usually, the risk analysis procedure is performed using the Tier 2 conditions that represent a reasonable compromise between the need for a detailed site assessment and the advantage of handling a rather simple and easy-to-use management tool [7]. For the evaluation of the atmospheric dispersion of vapors emitted from the subsurface, the ASTM method incorporates a Gaussian dispersion model to a simple "box model" where it is assumed that all the contaminant mass flow volatilized from the subsurface source is dispersed in atmosphere within a mixing height equal to 2 m (value representing the breathing zone height). This assumption is very conservative and thus can lead to an overestimation of the risks. Furthermore, the ASTM Gaussian model does not account for the spatial variability in the area considered thus adding a further approximation to the estimation of the risks for the receptors in proximity of the contaminated sites. In this paper, the accuracy of this approach was examined by comparing it with advanced dispersion models. The results provided by applying the standard Tier 2 risk analysis procedure were compared with those obtained using CALPUFF dispersion model. Subsequently, possible improvements of the standard procedure were examined.

## 2 METHODOLOGY

### 2.1 Pollutant dispersion models

In this study, the Gaussian box model applied in the RBCA-ASTM [6] procedure was compared with the CALPUFF dispersion model on the basis of the same emission conditions (i.e. same average emission rate from the contaminated soil).

CALPUFF is a multi-layer, multi-species, non-steady-state Lagrangian puff dispersion model that simulates the effects of time- and space-varying meteorological conditions on pollution transport, transformation, and removal [8]. The modelling system consists of three main components and a set of pre-processing and post-processing modules. The main components of the modelling system are CALMET (a diagnostic three-dimensional meteorological model), CALPUFF (an air quality dispersion model), and CALPOST (a post-processing package). The model includes algorithms for subgrid scale effects (such as terrain impingement), as well as longer range effects, such as pollutant removal due to wet scavenging and dry deposition, chemical transformation, and visibility effects of particulate matter concentrations. CALPUFF simulates puffs of material emitted from modelled sources, reproducing dispersion and transformation processes along the way. The primary output files from CALPUFF contain either concentrations or deposition fluxes evaluated at selected receptor locations. CALPOST is used to process these files, identifying the highest and second highest 3-h average concentrations at each receptor, for example.

In the RBCA-ASTM [6] procedure, the outdoor air concentration at the point of exposure ($C_{\text{outdoor}}$) due to volatilization from contaminated soil or groundwater is calculated using the following equation:

$$C_{\text{outdoor}} = \frac{J \cdot W}{U_{\text{air}} \cdot \delta_{\text{air}}} \cdot \tag{1}$$

Where $J$ is the emissive flux from the ground, $W$ is the extension of the source in the main wind direction, $\delta_{\text{air}}$ is the thickness of the mixing zone in air, and $U_{\text{air}}$ is the wind speed, assumed to be constant and not varying in the direction. The thickness of the mixing zone ($\delta_{\text{air}}$) is assumed to be constant and equal to 2 m as a value representative of the breathing zone.

The off-site concentration is calculated as follows:

$$C_{\text{offsite}} = C_{\text{outdoor}} \cdot ADF. \tag{2}$$

The term ADF represents the air dispersion factor based on a one-dimensional Gaussian model:

$$ADF = \frac{\delta_{\text{air}} \cdot S_w}{2\pi \cdot \sigma_y \cdot \sigma_z} \cdot \left[ 2 \cdot \exp\left( -\frac{1}{2} \frac{\delta_{\text{air}}^{\ 2}}{\sigma_z^{\ 2}} \right) \right]. \tag{3}$$

where $\sigma_y$ and $\sigma_z$ are the transverse and vertical dispersion coefficients that are calculated using Briggs' empirical formulae for urban soils, for atmospheric stability classes B, D, and F [9].

## 2.2 Risk calculation

The risk indices were calculated by dividing, for each pollutant, the simulated concentration value by the corresponding risk threshold value $RC$. The risk $R$ for a generic carcinogenic substance $i$ was calculated as follows:

$$R(i) = \frac{C_{\text{air}}(i)}{RC_{\text{canc}}(i)} \cdot 10^{-6}, \tag{4}$$

where $C_{\text{air}}$ is the average concentration in air, and $RC_{\text{canc}}$ is the reference concentration in air for carcinogenic effects:

$$RC_{\text{canc}} = \frac{TR}{IUR \cdot EC}, \tag{5}$$

where $TR$ is the target risk, $IUR$ is the inhalation unit risk for each contaminant of interest, and EC is the exposure factor for the inhalation pathway.

In the case of non-carcinogenic toxic substances, the hazard quotient for a generic non-carcinogenic substance $i$ is calculated as follows:

$$HQ(i) = \frac{C_{\text{air}}(i)}{RC_{\text{no.canc}}(i)} \cdot \tag{6}$$

The reference concentration in air for non-carcinogenic toxic effects $RC_{\text{no.canc}}$ can be estimated as follows:

$$RC_{\text{no.canc}} = \frac{THQ \cdot RfC}{EC}, \tag{7}$$

where *THQ* is the acceptable target hazard quotient, *RfC* is the reference concentration for each contaminant of interest, and EC is the exposure factor for the inhalation pathway.

The exposure factor for the inhalation pathway (EC) can be estimated with the following equation:

$$EC = \frac{EF_{go} \cdot EF \cdot ED}{AT \cdot 365 \ \frac{\text{days}}{\text{year}} \cdot 24 \frac{\text{h}}{\text{day}}}, \tag{8}$$

where *EF* is the exposure frequency (d/y), *ED* is the exposure duration (y), $EF_{go}$ is the daily outdoor frequency (h/d), and *AT* is the exposure time averaging (set at 70 years for carcinogens and at ED for non-carcinogens).

On the basis of the calculated risks for each contaminant, it is possible to calculate the cumulative risks and hazard ratios associated with the presence of *n* substances:

$$R_{\text{cum}} = \sum_{i}^{n} R(i) \cdot \tag{9}$$

$$HQ_{\text{cum}} = \sum_{i}^{n} HQ(i) \cdot \tag{10}$$

In this study, the reference concentrations for benzene were calculated with the Risk-net 3.1 software [10] for residential receptors (adults and children) assuming the following exposure parameters: *EF* = 350 d/y, $EF_{go}$ = 24 h/d, *ED* = 6 years (children), *ED* = 24 years (adults), and *AT* = 70 years. For the toxicological parameters, a RfC of 0.03 mg/m$^3$ and a IUR of $7.80 \times 10^{-6}$ (μg/m$^3$)$^{-1}$ were used [11].

## 2.3 Case study

In this study, the results provided by the standard Gaussian box model (Tier 2 assessment of the RBCA procedure) were compared with those obtained using the CALPUFF dispersion model. The illustrative case study concerned the emission to the atmosphere of benzene from a contaminated soil. The presence of a single contaminated area of 200 × 200 m on flat terrain was assumed (Fig. 1). Subsurface contamination was assumed to be at a depth of 1 m. The soil was assumed to be sandy, with an organic carbon content of 1%. The comparison between the two methods was carried out based on the same emission rate to the atmosphere (average emission of J = 1.31 g m$^{-2}$ d$^{-1}$). Emission rate per unit area was calculated with Fick's law from soil concentrations.

In the CALPUFF simulation, the source was discretized into 50 × 50 m areas in order to assess the effect of spatially non-uniform contamination. The unit emission rate of benzene for each sub-area ranged from 0.03 to 3 g m$^{-2}$ d$^{-1}$, with an average value for the whole source of 1.31 g m$^{-2}$ d$^{-1}$. The height of the sources was set in CALPUFF equal to 1 m and the temperature equal to the ambient temperature. The pollutant dispersion simulation was conducted over an area of 4 × 4 km, with a spatial resolution of 50 m and a temporal resolution of 1 h. The starting hourly meteorological data for the year 2019 were processed with the CALMET pre-processor.


Figure 1: Modelling domain and contamination source (in yellow). The simulations were compared on the basis of the same atmospheric emission rate.

In the standard Gaussian box model, the expected off-site outdoor air concentration was calculated considering a representative wind speed of 1.1 m $s^{-1}$ corresponding to the average value of the wind speeds used in CALPUFF for the simulated period. The extension of the source was considered to be 200 m regardless of the wind direction. For the application of the box model, the average emission rate of 1.31 g $m^{-2}$ $d^{-1}$ for the whole source was used. As discussed earlier, a mixing zone thickness in air ($\delta_{air}$) of 2 m was assumed. For the main wind directions, the distance-dependent concentration profiles calculated with the Gaussian model were reconciled with the values obtained with CALPUFF in the different wind directions.

The input parameters used in the two models are summarized in Table 1.

## 3 RESULTS AND DISCUSSION

The average wind distribution in the area is reported in Fig. 2. This figure shows that the prevailing wind direction is NW. Wind speed is frequently limited to values below 2 m $s^{-1}$. Wind calm frequency is around 60%. Figures 3 and 4 show the maps of carcinogenic risk and hazard quotient (HQ), respectively. These maps were obtained by applying eqns (4) and (6), respectively, to the concentration fields obtained by CALPUFF simulations. These figures show that the risk and HQ values around the source are affected by the uneven wind distribution. The same figures also show that, by representing the risk indices in form of a map, the impacts on nearby receptors can be evaluated immediately. In fact, in the entirely

Table 1: Input parameters.

| | CALPUFF | Gaussian box model | | |
|---|---|---|---|---|
| **Parameter** | **Figures 5–7** | **Figure 5** | **Figure 6** | **Figure 7** |
| Emission rates of benzene ($g/m^2/d$) | 0.03-3 (see Figure 1) | 1.31 | 1.31 | 1.31 |
| Wind speed (m/s) | See Figure 2 | 1.1 | 1.1 | 1.1 |
| Source dimension (m) | 200 × 200 | 200 | 200 | 200 |
| Mixing zone height (m) | Not applicable | 2 (all classes) | 11.7 (Class B) | 11.7 (Class B) |
| | | | 8.3 (Class D) | 8.3 (Class D) |
| | | | 5.6 (Class F) | 5.6 (Class F) |
| Wind direction frequency | See Figure 2 | Not considered | Not considered | 20% along NW |

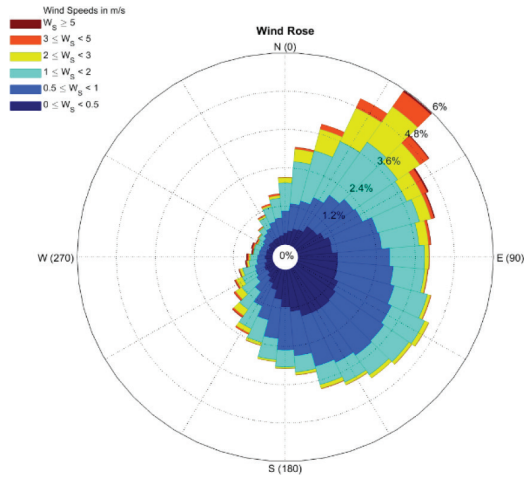


Figure 2: Wind distribution of the case study.

hypothetical case under consideration, non-negligible R values affect sensitive receptors (residential) located near the contaminated site (red areas in Fig. 3).

Figure 5 shows the concentration profile of benzene as function of the distance from the source obtained with the standard Gaussian box model and by CALPUFF in the prevailing wind direction (NW) for the different atmospheric stability classes.

The figure shows that the standard procedure overestimates the concentrations (and consequently the risk and the HQ) by more than one order of magnitude. The main reason of this overestimation is found in the structure of the ASTM box model, which assumes

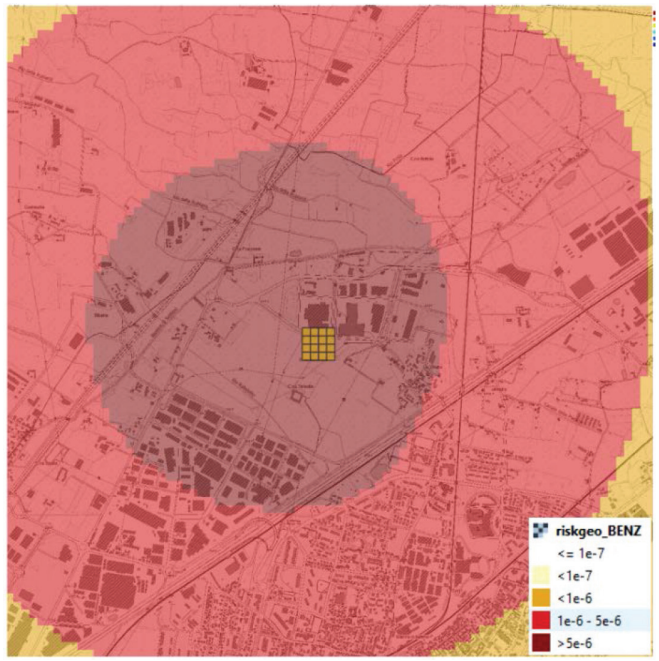


Figure 3: Carcinogenic risk map for benzene inhalation obtained with CALPUFF simulations.

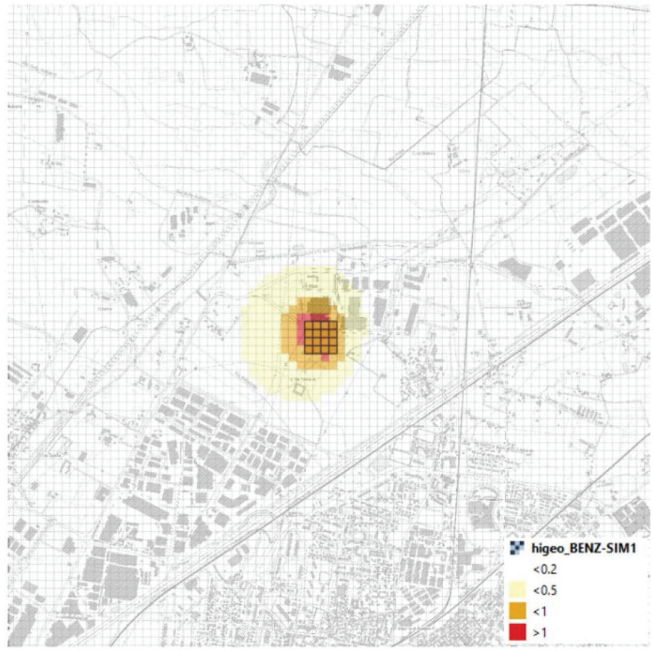


Figure 4: HQ map for benzene inhalation obtained with CALPUFF simulations.

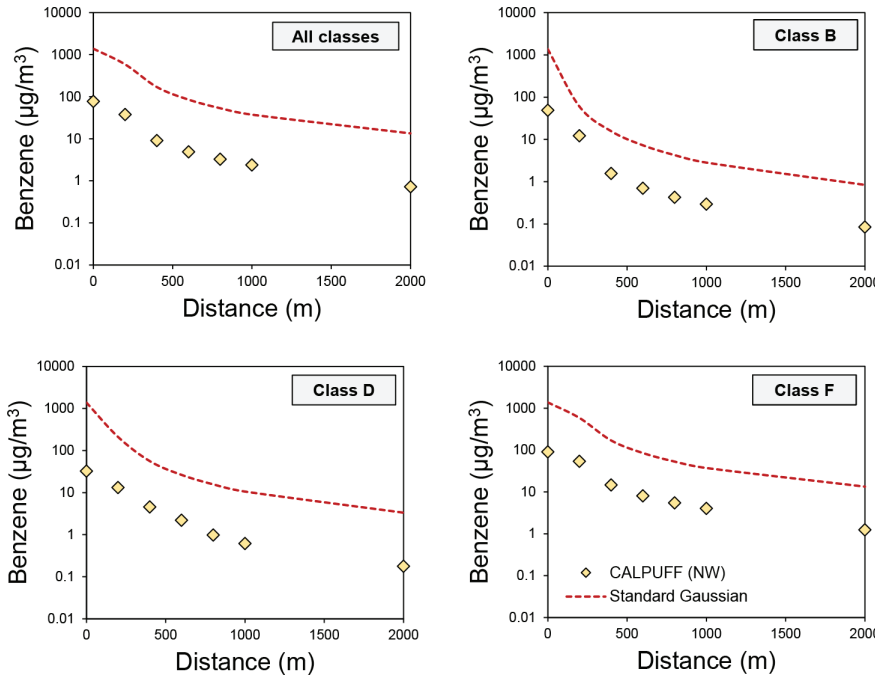


Figure 5: Concentration profiles as a function of the distance from source. Comparison between standard Gaussian model and CALPUFF in the prevailing wind direction (NW).

that all the contaminant mass flow volatilized from the subsurface source is dispersed in atmosphere within a mixing height equal to the breathing zone ($\delta_{air}$ = 2 m). This assumption can be considered satisfactory only if the mass flow of the contaminant is contained within this height. On the contrary, if the effective mixing height is higher than 2 m, as it would easily occur for unstable atmospheric conditions and high source lengths, the ASTM box model overestimates the concentration at the point of exposure. To overcome this limitation, Verginelli *et al.* (2017) [12] proposed a method to replace the mixing height of the box model with an "equivalent mixing height" for which a simple analytical solution was given. The equivalent mixing height, $\delta_{air,eq}$, accounts for the effective attenuation in air as a result of the vertical dispersion within the source zone. It is defined as follows:

$$\partial_{air,eq} = \frac{0.68 \cdot k_{lin} \cdot W}{1 + 0.68\sqrt{\frac{2}{\pi}} \cdot \ln\left(\frac{0.68 \cdot k_{lin} \cdot W}{\partial_{air}}\right)}, \tag{11}$$

where $W$ is the extension of the source in the main wind direction, and $k_{lin}$ is a parameter depending on the Pasquill-Gifford atmospheric stability class, the land use typology (rural or urban), and the distance from the source. For more information, refer to Verginelli *et al.* (2017) [12]. If $\delta_{air}$ of eqn (1) is replaced with $\delta_{air,eq}$, eqn (1) can be re-written and applied to the Gaussian box model:

$$C_{\text{outdoor}} = \frac{J \cdot W}{U_{\text{air}} \cdot \delta_{\text{air,eq}}} . \tag{12}$$

The comparison of concentration profiles obtained by introducing $\delta_{\text{air,eq}}$ (see Table 1) into the standard Gaussian model is reported in Fig. 6. This figure shows a minor discrepancy between the two approaches. Nevertheless, the application of the standard Gaussian model still leads to an overestimation of the concentrations.

A further improvement can be obtained by introducing in the Gaussian model the distribution frequency of wind. In this case, the concentration profile along a given direction, $C_{\text{outdoor}}$ (d), can be obtained as follows:

$$C_{\text{outdoor}}(d) = C_{\text{outdoor}} \cdot r(d), \tag{13}$$

where $C_{\text{outdoor}}$ is the outdoor concentration calculated with eqn (12), and $r(d)$ is the measured frequency of the wind along the direction $d$ (e.g. in this case for the direction NW, $r(d)$ is 20%). The comparison of the concentration profiles obtained by introducing $\delta_{\text{air,eq}}$ and the real wind frequency is reported in Fig. 7. The application of this method shows that the concentration profiles calculated by the modified Gaussian model approach the ones obtained with CALPUFF. The presented modifications could therefore be easily introduced in the Tier 2 ASTM-RBCA procedure.

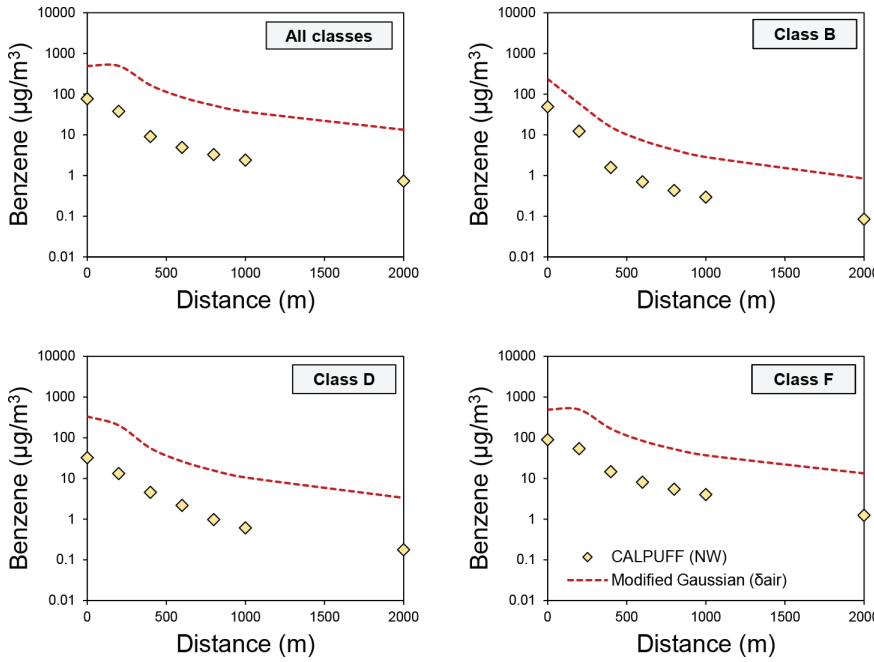


Figure 6: Concentration profiles as a function of the distance from source. Comparison between standard Gaussian model (modified with the equivalent mixing height) and CALPUFF in the prevailing wind direction (NW).

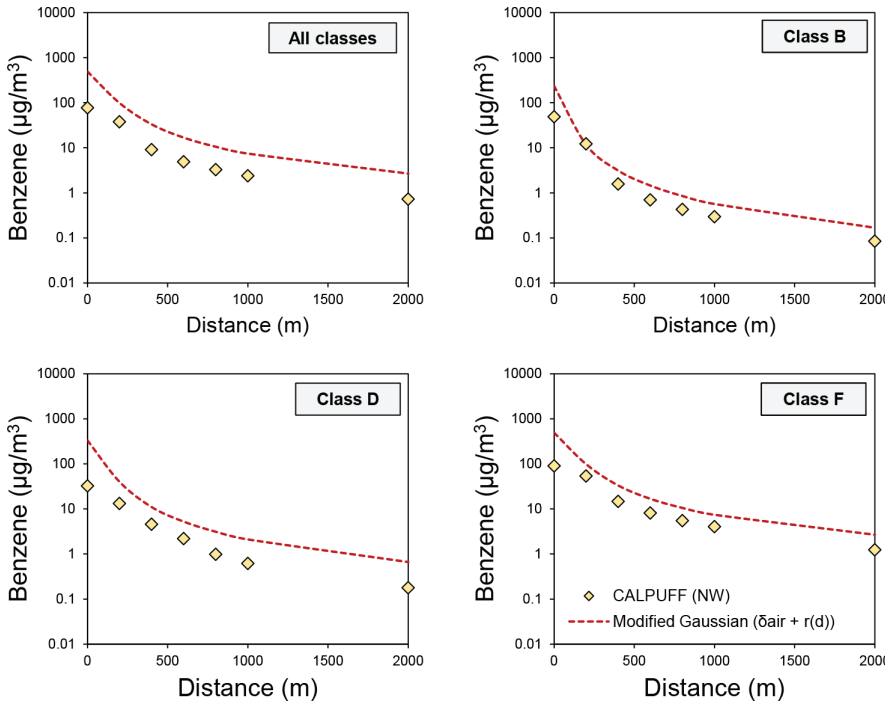


Figure 7: Concentration profiles as a function of the distance from source. Comparison between standard Gaussian model (modified with the equivalent mixing height and the wind distribution) and CALPUFF in the prevailing wind direction (NW).

## 4 CONCLUSIONS

The results of the proposed case study show that, for toxicology-based human health risk assessment, in the evaluation of inhalation risk for off-site receptors located in the vicinity of a contaminated site, the application of advanced tools for the assessment of spatial variability of risk is of primary importance. The comparison of the concentration profiles generated by two different modelling approaches (the standard Gaussian box model and CALPUFF) showed that, depending on the model, concentrations can vary up to more than one order of magnitude. Given the structure of the model and according to existing studies, the application of the standard Gaussian box model (described in the Tier 2 procedure of the ASTM-RBCA methodology) leads to an overestimation of exposure concentrations, in particular for unstable atmospheric conditions and high source lengths. Conversely, the results provided by CALPUFF seems more realistic, as this model is based on real meteorology and robust parameterization of the atmospheric boundary layer. In this study, two possible approaches for reducing the discrepancy between standard Gaussian model and CALPUFF were suggested. The first was the introduction of an equivalent mixing height, that is dependent on atmospheric stability class, land use typology, and the distance from the source. The second was the consideration of real wind distribution of the area. The introduction of these modifications could improve the performance of the standard Gaussian models thus reducing the overestimation of health risk indices.

Finally, this study showed that the use of advanced dispersion models integrated with the risk calculation methodologies, allowed a detailed characterization of the risk in the area under examination, highlighting the most critical areas and comparing them with the presence of any sensitive receptors.

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