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Comparison of plume lateral dispersion coefficients schemes: Effect of averaging time

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ABSTRACT

Dispersion modeling is an important decision tool for estimating the impact of human activities on the environment and its populations. However, it was proved by researchers that AERMOD and CALPUFF, the current regulatory models, do not account for the effect of averaging time. In consequence, these models do not have the ability to predict short-term time peak concentrations. This inability arises from the errors in the lateral and vertical dispersion estimates, which are reliable only to predict 10 min or longer average concentrations. In this paper, a novel evaluation based on Irwin (1983) was conducted to investigate the effect of averaging time on the lateral dispersion and maximum concentration estimates. The Pasquill-Gifford, Högström, Draxler (embedded in CALPUFF) and AERMOD lateral dispersion schemes were tested using the Round Hill II experiment, developed to investigate the effects of averaging time on atmospheric transport and diffusion. The observed lateral dispersion was derived from the lateral concentration profiles along 3 sampling arcs (50, 100 and 200 m), measured on 3 different averaging times (0.5; 3 and 10 min). The observed lateral dispersion was compared to those estimates. The results of the comparison show that AERMOD and Draxler correlate better with measured data than the PG and Högström methods. However, their estimates are biased and the magnitude of systematic errors tends to grow as the averaging time decreases. Moreover, AERMOD and Draxler, with Peak-to-Mean (P-M) adjustment, tend to overestimate the lateral dispersion farther from the source and underestimate at downwind distances less than 200 m. The analysis also highlights some concerns on the P-M ratio application due its subjectivity. The present investigation on the effect of short-term averaging times on atmospheric transport and diffusion may help to understand some issues related to the use of dispersion models in the case of flammability, malodor nuisance and toxicity

1. Introduction

Dispersion models have been frequently used in air pollution problems to determine the concentration of contaminants downwind from a continuous point source (Draxler, 1976). However, most of the Gaussian and Puff models including Industrial Source Complex 3 (ISC3), AERMOD and CALPUFF, the most used and recommended models by the US EPA, are unable to predict short-term peak concentrations. Several applications require estimates of concentrations averaged over shorter time periods that those estimated with models commonly used for regulatory applications, such as AERMOD and CALPUFF. For example, predicting odor concentrations requires converting AERMOD 1 h estimates to values that correspond to averaging times of a few seconds to few minutes (Venkatram, 2002). According to several researchers, a lack of agreement has been found between the estimated and observed downwind concentrations using these models on shorter averaging times (Beaman, 1988). In fact, those models were not designed to predict short-term peak concentrations.

The widely used Gaussian approximations were calibrated from historical tracer dispersion experiments, with averaging times of 10 min or longer (Irwin et al., 2007). Therefore, estimates are only reliable under these respective temporal scales. Common practice consists of converting model predicted estimates to shorter time periods using Peak-to-Mean (P-M) formula presented on Equation (1) (Dourado et al., 2012; Venkatram, 2002; Vieira de Melo et al., 2012; Wang et al., 2006).

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$$\frac{C_m}{C_p} = \left(\frac{t_m}{t_p}\right)^{-c} \tag{1}$$

This expression relates the maximum mean concentration (C_p) observed for a shorter averaging period (t_p) and the maximum mean concentration (C_m) observed for a longer averaging period (t_m), which is the mean concentration calculated by the model. The values of the exponent, c, in the literature range from 0.2 to 0.5 (Venkatram, 2002), depending on atmospheric stability (Schauberger et al., 2012).

For shorter averaging times, ISC3, AERMOD and CALPUFF require P-M conversion due to the sum of the effects of dispersion and change in the axis of the plume (meandering), which are considered as absolute dispersion in their estimates. These effects are caused by different turbulent scales, which are virtually indistinguishable, except that only the relative diffusion of the plume around its instantaneous centroid is responsible for the effective dilution of pollutants. Plume meandering is the slow lateral back-and-forth shifting of a plume in response to nondispersing lateral eddies that are larger than the plume (Cimorelli et al., 2005). The more the averaging time increases and the distance from the source to the receptor decreases, the more important the meandering influence on the lateral dispersion is. Meandering tends to disappear over longer averaging times and farther from the source, and the fluctuations are mainly internal (Mortarini et al., 2009). Generally odors are no longer perceived further than few kilometers from the source (Guo et al., 2004).

The reason why both meandering and relative dispersion effects are treated as absolute dispersion in the regulatory models, has the critical point in the estimate of the vertical and horizontal growth of the plume. This growth is usually expressed in terms of the standard deviation of the concentrations in the lateral and vertical directions (σ_y–σ_z) (Draxler, 1976). In practice, these terms are very difficult to quantify effectively and in problems of atmospheric diffusion, σ_y and σ_z are estimated by empirical and semi-empirical methods (Hay and Pasquill, 1957). According to Draxler (1976), several methods have been suggested to determine the dispersion coefficients. However, they all share a weakness: the inability to calculate short-term time averages, as in the case of flammability, malodor nuisance and, often, toxicity (Vieira de Melo et al., 2012; Dourado et al., 2014). In spite of this limitation, those methods have been extensively used to predict odor and toxic dispersion. In this sense, more discussion appears to be needed on the communication of the magnitude of errors to decision makers (Irwin et al., 2007).

In this respect, the present work aims to evaluate the lateral plume dispersion parameters compared to field tracer data collected in three different averaging times. Complementing Irwin's (1983) work, this novel evaluation was conducted to investigate the effect of averaging time on the lateral dispersion and arc maximum concentration estimates. The dispersion parameters schemes used in this analysis include Pasquill-Gifford using Turner's technique (Turner, 1997), Högström (1964) and those embedded in AERMOD (Cimorelli et al., 2005) and CALPUFF using Draxler's (1976) method. The performances of these methods are compared with observations of Round Hill II tracer data. The main focus of this work is to help understanding some problems that occur when employing dispersion models to predict short-term peak concentrations.

2. Background

Due the lack of understanding of turbulence, for atmospheric transport and dispersion, it is very difficult to reproduce exactly the observations of a plume at a given time and location. (Yee et al., 1994). Plume dispersion is caused by turbulent eddies of different sizes. While small turbulent eddies tend to spread the plume, large eddies tend to cause it to meander. As the plume becomes wider, larger eddies become effective in dispersing it and smaller eddies become increasingly ineffective (Gifford Jr., 1959; Seinfeld and Pandis, 2006). Therefore, eddies that are larger than the instantaneous plume width will waft around the plume as a whole without changing its internal structure, and contribute to the low-frequency motions of the dispersing material in the form of plume meandering, causing intermittency (periods of zero concentration). On the other hand, eddies of smaller size that are comparable to the size of the plume produce local distortions and convolutions that contribute to the in-plume fluctuations due to clean the air entrainment (Yee et al., 1994).

To mitigate the effects of fluctuations, the best that can be done is to predict the average characteristics of plume dispersion (Irwin et al., 2007). Nevertheless, there are some effects on averaging the plume properties. Figure 1 shows the real case of plume boundaries and concentration distributions of an instantaneous snapshot and exposures of a few minutes and several hours. The meandering behavior of the instantaneous plume can be seen, with the width of the plume gradually growing downwind of the source. As the averaging time increases, the plume assumes a more regular appearance and the concentrations have a smoother distribution (Seinfeld and Pandis, 2006). Due to the sum of the large and small eddies effects, it is typical of observed plumes that the lateral and vertical instantaneous dispersion are smaller than the averages and, consequently, the instantaneous concentrations are at least as large as the mean (Hanna, 1967). The plume meandering dominates the concentration fluctuations of time averaged plumes at short downwind distances (in the range of few kilometers), while the effects of in-plume fluctuation appears farther from the source.

Irwin et al. (2007) reported the influence of averaging time on atmospheric transport and diffusion. Analyzing data from the Round Hill II field experiment, the concentrations measured at 30 s are around 1.66 times higher than those measured at 10 min.

Most of the Gaussian models consider an average concentration for a time period ranging from 10 min to 1 h (De Melo Lisboa et al., 2006). According to Cimorelli et al. (2005), in the AERMOD the lateral dispersion expression was reformulated to better fit the data from the Prairie Grass Experiment. On the respective tracer database, samples were collected over 10 min averages, allowing the AERMOD to estimate lateral dispersion over this averaging time or longer. The limitations of CALPUFF and the Pasquill-Gifford curves are similar. One of the most reliable methods used to calculate the dispersion coefficients in CALPUFF is based on Draxler's (1976) formulation. The semi-empirical method developed by Draxler employed the major part of the data with averaging times of 30 min or longer. Pasquill-Gifford empirical curves were based on samples collected over 10 min averages of near-ground level releases.

According to Mortarini et al (2009) and Franzese (2003), Gifford's (1959) fluctuating plume model proved to be a simple and effective tool for predicting concentration moments of order higher than the mean for stationary releases of contaminant in idealized homogeneous turbulence. The Gifford's model, later adapted by Mussio et al. (2001), De Melo Lisboa et al. (2006) and Dourado et al. (2014), is a Gaussian model capable of providing the percentage of time during which concentration remains above or below a defined threshold. This characteristic turns the respective model a valuable tool for odorant compound dispersion modelling. This model is based on the idea that the plume can be decomposed into two independent parts: a meandering part and a relative-diffusion part (Mortarini et al., 2009). However, it is assumed that there are no fluctuations inside the instantaneous plume (Hanna,

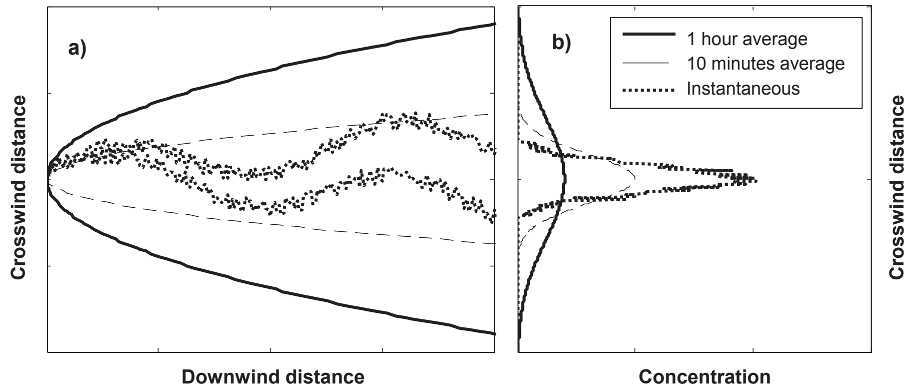

Fig. 1. Plume boundaries and concentration distributions of a plume at different averaging times.
Source: Adapted from Seinfeld and Pandis (2006).

1967). More sophisticated models as idealized by Thomson (1990), Franzese (2003) and Mortarini et al (2009) can deal with both meandering and in-plume fluctuations in complex situations.

The use of fluctuating plume model was hindered by a lack of good estimates of the relative or dispersion (Hanna, 1967). Högström (1964) developed one of the few works in this sense. The last author conducted a series of tests in which puffs of smoke tracer were released at 30 second intervals and photographically tracked downwind. From these experiments he extracted short-term averaging time horizontal and vertical diffusion parameters. However, even Högström, later in 1972, acknowledged in his paper that there is great deal of uncertainty in modeling the spread of the instantaneous plume, which determines the relevant concentrations (Högström, 1972).

According to Hanna (1967), the fluctuation plume model and the traditional Gaussian models such as AERMOD and CALPUFF are fundamentally the same. One of the differences between these models lies on the dispersion coefficients. While fluctuation plume models uses Högström's (1964) coefficients, the CALPUFF and ISC3 employ Draxler (1976) and Pasquill-Gifford curves, respectively. AERMOD uses its own dispersion coefficient. From this perspective, a comparison of plume lateral dispersion coefficients schemes employed on the most used models for odors and toxic pollutants regulations would be a valuable information for the model developers and users. This article intends to show some issues of these schemes when used to predict short-term times averages over short travel times and simple terrain.

3. Methodology

Four lateral dispersion methodologies were tested: Pasquill-Gifford, Högström, AERMOD and Draxler. A detailed description of the dispersion parameter schemes can be found at Seinfeld and Pandis (2006), Högström (1964), Cimorelli et al. (2005) and Draxler (1976). The methods performances were evaluated using the Round Hill II field tracer experiment, conducted in 1957 (Cramer and Record, 1957). Round Hill II has a unique set of 10 experiments having joint measurements of 0.5-min, 3-min and 10-min concentrations along arcs. An objective of the 10 Round Hill II releases was to investigate the effects of averaging time on atmospheric transport and diffusion. Sulfur dioxide concentrations were sampled along 3 arcs (50, 100, and 200 m), and the release height and sampling height was 1.5 m. Samples were taken for the first 30 s and the first 3 min of each 10 min sample. Temperature and wind speed were measured on a tower located at the experiment site (2 m altitude). The data were originally averaged over 10 min. Complementary meteorological data (relative humidity, atmospheric pressure, cloud cover, ceiling height, dew point) from the Taunton, Massachussetts[1] station (located around 50 km from the experiment site) were used, in order to meet the minimum model requirements. The last meteorological station was located in a grassy and flat terrain (17 m altitude) with low roughness, surrounded by small suburbs and trees. Convective mixing heights were characterized by upper air soundings from a meteorological station at Nantucket airport[2] (around 130 km from Round Hill). The Nantucket sounding station (4 m altitude) was located around 500 meters from the ocean, with vegetation and terrain similar to the experiment site. These supplementary data far away from the experiment site were used due to the unavailability of closer measurements.

For the set of 10 experiments, the observed lateral dispersion was derived from the lateral concentration profiles of dosage along sampling arcs with the help of the MATLAB® least squares curve fitting routine. The estimates of dispersion coefficients and maximum concentrations were the result of the best fit reached between the observed data and Gaussian curve. The procedures followed the instructions of the ASTM standard guide "ASTM D 6589: *Standard Guide for Statistical Evaluation of Atmospheric Dispersion Model Performance*".

Statistical comparisons of the estimated dispersion parameters with the observed values were computed. The following statistical indices, proposed by Chang and Hanna (2004), were used: Bias (Equation (2)), mean Fractional Error – *FE* (Equation (3)), Normalized Mean Square Error – *NMSE* (Equation (4)), Spearman correlation coefficient – ρ (Equation (5)) and Factor of two – *FACT2* (Equation (6)). The Mean Absolute Error – *MAE* (Equation (7)), suggested by Willmott and Matsuura (2005) was also employed.

$$\mathrm{BIAS} = \overline{\mathrm{C_p} - \mathrm{C_o}} \tag{2}$$

$$\mathrm{FE} = \frac{2.\left(\overline{\mathrm{C_p}} - \overline{\mathrm{C_o}}\right)}{\left(\overline{\mathrm{C_p}} + \overline{\mathrm{C_o}}\right)} \tag{3}$$

$$\mathrm{NMSE} = \frac{\overline{\left(\mathrm{C_p} - \mathrm{C_o}\right)^2}}{\overline{\mathrm{C_p}} \cdot \overline{\mathrm{C_o}}} \tag{4}$$

[1] http://www.ncdc.noaa.gov/most-popular-data#dsi-3505.
[2] http://www.esrl.noaa.gov/raobs/.

$$\rho = \frac{\sum\left(C_p - \overline{C_p}\right)\left(C_o - \overline{C_o}\right)}{\sqrt{\sum\left(C_p - \overline{C_p}\right)^2\left(C_o - \overline{C_o}\right)^2}} \quad (5)$$

$$\mathrm{FAC2} = 0.5 \le \frac{C_p}{C_0} \le 2 \quad (6)$$

$$\mathrm{MAE} = \frac{1}{n}\sum_{i=1}^{n}\left|C_p - C_o\right| \quad (7)$$

The estimates of maximum concentrations were compared with the observed data, using the Gaussian model of Equation (8):

$$C_{(x,y,z,H)} = \frac{Q_s}{2\pi\sigma_y\sigma_z u}\cdot exp\left(\frac{-y^2}{2\sigma_y{}^2}\right)\cdot\left[exp\left(\frac{-(z-H)^2}{2\sigma_z{}^2}\right) + exp\left(\frac{-(z+H)^2}{2\sigma_z{}^2}\right)\right] \quad (8)$$

$C_{(x,y,z)}$ is the concentration of the emission at x meters downwind of the source, y meters laterally from the centerline of the plume, and z meters above ground level. Q_s is the quantity or mass of the emission per unit of time, u is the wind speed, H is the height of the source above ground level and σ_y and σ_z are the standard deviations of a statistically normal plume in the lateral and vertical dimensions, respectively.

The AERMOD and Draxler methods provide lateral dispersion results for long averaging times of approximately 1 h (Vieira de Melo et al., 2012). In order to examine these techniques on shorter averaging times, the results of the methods need to be appropriately converted. To account for the effect of averaging time on AERMOD and Draxler, the Peak-to-Mean (P-M) presented in Equation (1) was used. The values of the exponent, c, in the literature range from 0.2 to 0.5 (Venkatram, 2002), depending on atmospheric stability (Schauberger et al., 2012). Due to neutral to slightly instable experimental condition, it was assumed that coefficient c was equal to 0.4. Peak-M values of 6.79 (10 min), 3.31 (3 min) and 2.04 (30 s) were employed. Analogously to Equation (1), a formula was used to relate larger and shorter averaging times to their respective dispersion coefficients (Equation (9)).

$$\frac{\sigma_m}{\sigma_p} = \left(\frac{t_m}{t_p}\right)^c \quad (9)$$

where σ_p and σ_m are the values of σ_y assumed for a shorter and a longer averaging period respectively. The Pasquill and Högström estimates are valid for 10 min and 30 second averages, so Equation (9) was not used for these cases.

Table 1 summarizes the average and standard deviation of lateral dispersion (σ_y) and maximum concentrations (C_{max}) along crosswind arcs at various distances downwind for each averaging time. As expected, C_{max} increases as the averaging time decreases and lateral dispersion increases as averaging times increase. As shown by the standard deviation (Std) in Table 1, the variability of C_{max} seems to decrease as the distance increases. According to Irwin (2007), because Std is a measure of the relative scatter about C_{max} and because C_{max} is seen to increase as the averaging time decreases, this suggests that the actual variability of C_{max} may increase as the averaging time decreases. The opposite happens to σ_y, since the Std of σ_y decreases with σ_y. Another important influencing factor in Std of σ_y are the different turbulent scales in the atmosphere, since the longer the averaging time is, the more significant the role played by eddies of different sizes in spreading the plume, increasing the variability of σ_y.

4. Results

4.1. Comparison of lateral dispersion estimates

Table 2 shows the lateral dispersion estimates performances against the Round Hill II tracer experiment. The results suggest that Högström and Pasquill-Gifford improve their estimates as the averaging time decreases, for downwind distances around 200 m. Högström reduces the Fractional Error (FE) from 90% to 50%, as the averaging time decreases from 10 min to 30 s. For PG estimates, there is an FE reduction of 50% from the 10 minute to 30 second averages. The AERMOD and Draxler results showed an opposite behavior, since these methods show inferior performances at shorter averaging times. Without P-M adjustment, AERMOD overestimates by a range of about 70–110%, and Draxler by a range of 80–120%.

Instead of overestimating, AERMOD and Draxler underestimate the observed lateral dispersion after using the P-M. Their performances were improved significantly. For instance, Bias and FE are reduced from 5.5 m to 70% to −1.3 m and 0%, for an averaging time of 10 min. P-M also has an effect on the optimization of NMSE indexes, decreasing from 0.40 to 0.01 in AERMOD and 0.65 to 0.01 in Draxler, both for 10 min averages. FACT2 was also enhanced to 53% on AERMOD and 67% on Draxler for 30 second averages. This reveals the predominance of systematic errors that are corrected after P-M implementation. Despite having worse performances at shorter averaging times, overall, after peak-to-mean scaling, AERMOD and Draxler reached the best index of bias, FE, NMSE and FACT2. However, for the smallest averaging time considered, PG had the least bias, FE, NMSE and 83% of its estimates were within a Factor of 2 (Table 2).

As shown by the correlation coefficients (ρ) in Table 2, stronger relationships between observed and predicted results were found for AERMOD and Draxler. Of all the methods, Högström presented the lowest values of ρ.

The results for the three downwind distances ranges were used to assess the variation in the methods' performances as a function of the distance traveled by the plume and also as a function of averaging time. The dependence of the distance and averaging time in the comparison results on the lateral dispersion parameter

Table 1
Geometric average and standard deviation of lateral dispersion (σ_y) and maximum concentration (C_{max}) segregated by distance and averaging time.

Parameter	Averaging time	Average			Standard deviation (Std)		
		10 min	3 min	0.5 min	10 min	3 min	0.5 min
σ_y (m)	50 m	9.5	6.8	6.3	3.5	2.3	2.5
	100 m	16.4	11.9	8.4	8.0	4.4	2.9
	200 m	28.4	20.2	15.8	19.4	9.8	8.4
C_{max} (mg m^{-3})	50 m	229.9	327.1	364.5	148.7	193.4	259.5
	100 m	99.1	125.1	166.6	108.9	114.3	162.8
	200 m	36.8	37.1	23.2	48.7	40.5	9.9

Table 2
Summary of statistical performances for Högström, AERMOD, Draxler and PG lateral dispersion schemes as a function of averaging time of 0.5, 3 and 10 min, using the Round Hill II data set. AERMOD and Draxler estimates without peak-to-mean (default) and with peak-to-mean (w/P-M).

Statistical index	Averaging time	Högström	AERMOD		Draxler		PG
			Default	W/P-M	Default	W/P-M	
BIAS (m)	10 min	−11.56	15.47	−1.27	21.30	1.59	−8.83
	3 min	−7.08	19.96	−2.96	25.78	−1.20	−4.35
	0.5 min	−3.35	23.69	−4.31	29.51	−3.45	−0.61
FE	10 min	−0.90	0.67	0.02	0.80	0.17	−0.58
	3 min	−0.71	0.88	−0.20	1.00	−0.06	−0.37
	0.5 min	−0.46	1.10	−0.60	1.20	−0.47	−0.09
NMSE	10 min	1.28	0.40	0.01	0.65	0.01	0.51
	3 min	0.67	0.94	0.07	1.33	0.01	0.17
	0.5 min	0.19	1.68	0.38	2.21	0.21	4.5×10^{-3}
ρ	10 min	0.56	0.69	0.69	0.75	0.75	0.63
	3 min	0.61	0.79	0.79	0.82	0.82	0.68
	0.5 min	0.63	0.78	0.78	0.76	0.76	0.75
FACT2	10 min	0.23	0.40	0.78	0.27	0.81	0.53
	3 min	0.37	0.20	0.81	0.13	0.93	0.73
	0.5 min	0.67	0.03	0.59	0.00	0.74	0.83
MAE	10 min	11.65	17.41	7.22	21.74	7.91	9.08
	3 min	7.30	19.96	4.67	25.78	3.95	5.07
	0.5 min	3.80	23.69	4.33	29.51	3.80	2.53

estimates is illustrated in Figure 2. The mean FE is one of the more useful statistics for characterizing the systematic errors. The illustration of the FE was used to characterize the precision of estimates, similar to those employed by Irwin (1983) to allow comparison between results. The mean FE and standard deviation of FE was used as measure of average bias and scatter of the estimates.

Draxler and AERMOD use a similar approach to parameterize the lateral dispersion, and the statistical index in Table 2 and the FE

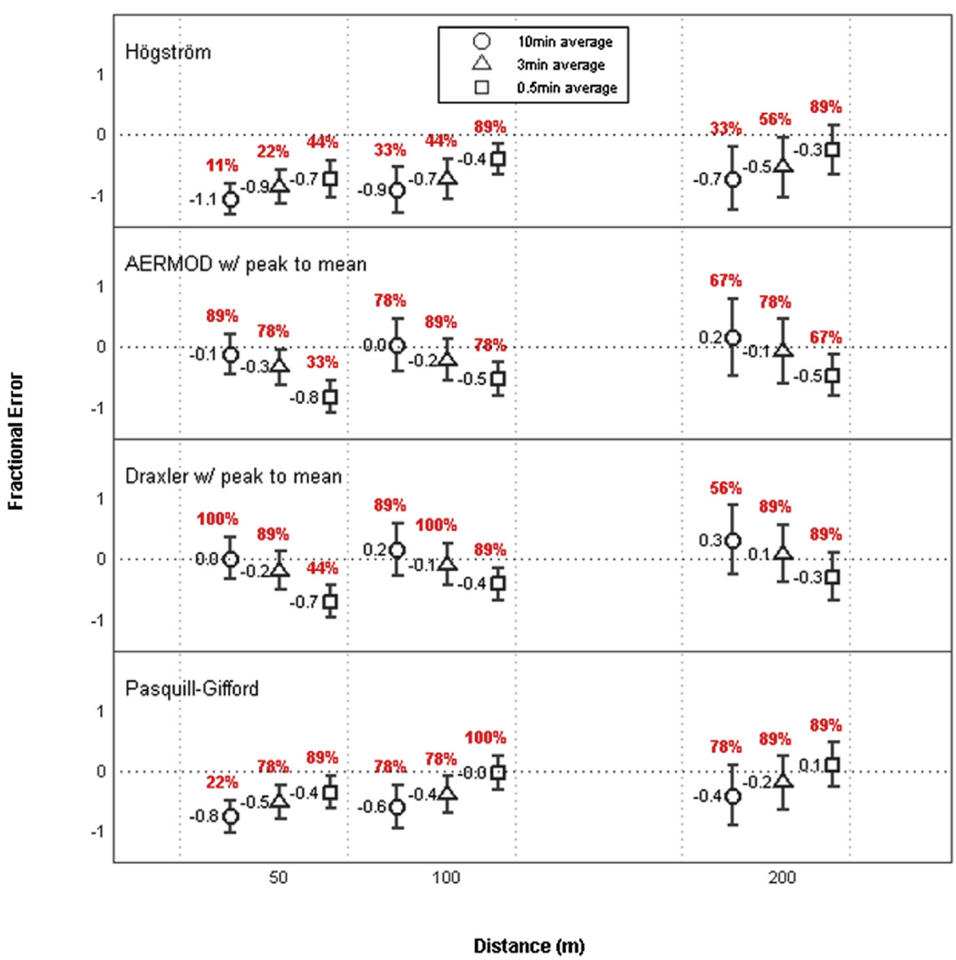

Fig. 2. Mean Fractional Error (FE) between measured and estimated lateral dispersion for the four schemes assessed, as a function of downwind distances of 50, 100 and 200 m and averaging times of 0.5, 3 and 10 min. The number above each symbol indicates the percent of the estimated values within a factor of 2. The mean fractional error is given to the left of each symbol. The bars depict the mean fractional error plus or minus one standard deviation. Results of AERMOD and Draxler are adjusted by the P-M factor for each averaging time.

profile in Figure 2 show this similarity. Using the P-M adjustment, there was a trend in the AERMOD and Draxler methods to overestimate the lateral dispersion at the farthest downwind distances and to overestimate it at closer distances. In general, Draxler reached better agreement with the observed lateral dispersion compared to AERMOD, as can be seen from the FE values and the fraction of data within the factor of 2 range in Figure 2. Both methods show improved performance at longer averaging times. Draxler (1976) and Irwin (1983) had already found that the Draxler scheme (without P-M adjustment) results in the smallest mean FE in the estimated dispersion parameter compared to PG and the other methods for longer averaging times (>10 min). There is a trend to find better results using AERMOD and Draxler in downwind distances greater than 200 m, for averaging times of 30 s or smaller with P-M adjustment. However, extrapolation of the results to greater downwind distances cannot be accomplished without some uncertainty.

Of the four methods compared, PG had the smallest FE in downwind distances of 50, 100 and 200 m for averaging times of 30 s. Högström underestimates the Round Hill observation of the lateral spread of the plumes for all the averaging times and distances evaluated. Similarly to PG, Högström's performance improved as the plume traveled away from the source and also for shorter averaging times. Högström developed his method to calculate the dispersion of a puff over 30 second averages. It is therefore expected that the Högström estimates find better results at shorter averaging times. Unlike AERMOD and Draxler, the Högström and PG estimates tend to reach better agreement with the measured lateral dispersion farther from the emission. However, there is a tendency of PG to overestimate rather than underestimate for distances greater than 200 m and it is possible that the Högström estimates have this same behavior.

The variation of FE is smaller at shorter distances and shorter averaging times, as also reported by Irwin (1983). This suggests that variation of the horizontal wind direction contributes significantly to the lateral dispersion at greater transport distances. According to Irwin (1983), the dispersion estimation schemes assume steady-state meteorological conditions during transport downwind. Therefore, it is possible that variation in the transport direction may result in unexpectedly large values of lateral dispersion. Additionally, the effect of fluctuation of the wind direction (large eddies) on plume spreading is stronger for longer distances and longer averaging times. Hence, the standard deviations of FE (represented by the bars in Figure 2) are higher in these conditions.

The correlations between model estimates and measured values segregated by averaging time and downwind distance and their respective statistical significance values (p) are presented in Table 3. The correlations were stronger for longer averaging times. This highlights a difficulty in explaining the data variability of short-term time concentrations. In general, worse agreement between the observed and predicted data was found for longer distances due to the effect of variation in the transport direction, not addressed by the methods. Overall, AERMOD and Draxler (with P-M adjustment) appear to correlate better with the measured data, compared to PG and Högström. These last two methods use a purely empirical approach to characterize the dispersion coefficients. On the other hand, both AERMOD and Draxler use a more robust approach to parameterize the turbulence and plume dispersion.

4.2. Comparison of maximum concentration estimates

A summary of the statistics of the model predictions compared with measurements of maximum concentrations along the arcs for the three averaging times considered is presented in Table 4. While Högström and PG overestimate the maximum concentrations, Draxler and AERMOD underestimate them, when used in a Gaussian model (Equation (8)). Draxler performed better than AERMOD, although both methods underestimated the maximum concentration even after P-M had been used. According to Irwin (1983), the tendency to underestimate the peak concentration is related to the tendency for the models to overestimate mostly the vertical dispersion parameter. For shorter averaging times (30 s), AERMOD and Draxler found the better statistical indexes and had more estimates within a factor 2 among the methods, after P-M implementation. Such improvement could arise from correction of systematic errors by P-M adjustment. However, this scaling factor did not reach the same agreement for the other time scales (10 and 3 min). It may prove difficult to set correct P-M values for different averaging times and atmospheric conditions. Some issues have already been warned of by researchers, such as Guo (2006), about the choice of an appropriate P-M ratio to adjust the modeled concentrations. The subjectivity of this scaling method may lead to discrepant results. Assuming slightly different values of c available in literature, Equations (1) and (9) may produce different maximum concentration values among users.

The statistics show the tendency of Högström to underestimate the lateral and vertical dispersion, resulting in unexpectedly elevated maximum concentration estimates. The PG scheme presents similar results with small bias, however.

Despite the high bias, the strength of the correlation coefficients in Table 4 shows that the model estimates have a strong relationship with the measured data. As expected, AERMOD and Draxler obtained higher ρ values.

Table 3
Correlation and statistical significance between observed and predicted lateral dispersion segregated by downwind distances of 50, 100 and 200 m and averaging time of 10, 3 and 0.5 min. Spearman correlation (ρ) between predictions and observations.

Método	Averaging time	ρ			p		
		50 m	100 m	200 m	50 m	100 m	200 m
Hogstrom	10 min	−0.12	0.02	0.08	0.78	0.98	0.84
	3 min	−0.17	−0.18	−0.27	0.68	0.64	0.49
	0.5 min	−0.27	0.35	−0.67	0.49	0.36	0.06
AERMOD	10 min	0.80	0.85	−0.35	0.01	0.01	0.36
	3 min	0.87	0.88	−0.13	0.00	0.00	0.74
	0.5 min	0.55	0.22	0.78	0.13	0.58	0.02
Draxler	10 min	0.63	0.80	0.63	0.08	0.01	0.08
	3 min	0.63	0.55	0.72	0.08	0.13	0.04
	0.5 min	0.62	0.38	0.17	0.09	0.31	0.68
P.G.	10 min	0.07	0.17	0.23	0.88	0.68	0.55
	3 min	−0.13	−0.10	0.12	0.74	0.81	0.78
	0.5 min	0.57	0.27	0.43	0.12	0.49	0.25

5. Conclusions

The results of the comparison of lateral plume dispersion coefficients schemes support the conclusions that AERMOD and Draxler performed better than the PG and Högström methods, after using the P-M ratio. The AERMOD and Draxler estimates are strongly correlated with the observed maximum concentrations and lateral dispersion. However, their estimates are biased and the magnitude of systematic errors tends to grow as the averaging time decreases. Despite the bias reduction, the application of the P-M ratio is subjective and can produce different performances as it depends on the modeler's experience, the atmospheric conditions and distance.

The distance dependence of the methods' performances was evaluated. The analysis revealed that AERMOD and Draxler, with P-M adjustment, tend to overestimate the lateral dispersion farther

Table 4
Performance of methods in estimating the maximum concentrations in the Round Hill experiment for times of 0.5, 3 and 10 min through a Gaussian equation.

Statistical index	Averaging time	Högström	AERMOD		Draxler		PG
			Default	W/P-M	Default	W/P-M	
BIAS (mg m^{-3})	10 min	669.9	−114.3	−100.5	−112.8	−97.5	184.5
	3 min	692.6	−147.3	−114.7	−145.7	−109.4	169.3
	0.5 min	658.0	−169.0	−88.5	−167.4	−77.6	141.9
FE	10 min	1.5	−1.5	−1.1	−1.5	−1.1	0.9
	3 min	1.4	−1.6	−0.9	−1.5	−0.8	0.7
	0.5 min	1.3	−1.6	−0.4	−1.5	−0.3	0.7
NMSE	10 min	4.6	8.1	3.1	7.1	2.6	0.9
	3 min	3.4	9.4	1.7	8.3	1.4	0.5
	0.5 min	2.8	11.1	0.4	9.8	0.3	0.3
ρ	10 min	0.86	0.86	0.9	0.87	0.9	0.83
	3 min	0.89	0.89	0.9	0.89	0.9	0.84
	0.5 min	0.77	0.85	0.8	0.83	0.8	0.85
FACT2	10 min	0.00	0.03	0.1	0.03	0.2	0.20
	3 min	0.00	0.00	0.2	0.00	0.3	0.33
	0.5 min	0.10	0.00	0.7	0.03	0.6	0.33
MAE	10 min	669.9	114.3	100.6	112.8	97.6	188.7
	3 min	692.6	147.3	114.7	145.7	109.4	177.3
	0.5 min	658.0	169.0	98.1	167.4	92.4	161.2

from the source and underestimate it at downwind distances less than 200 m, as encountered by Vieira de Melo et al. (2012). The Högström and PG schemes underestimate the lateral dispersion; however, as observed by Irwin (1983) that evaluated lateral dispersion schemes in farther downwind distances from the source (above 1 km), this bias could be reversed beyond 200 m. However, extrapolation of the results to greater downwind distances cannot be accomplished without some uncertainty (Irwin, 1983).

The respective evaluation also highlights the effect of large turbulent eddies on plume spreading at greater distances and longer averaging times, which increase the lateral dispersion and decrease the arc maximum concentration. Moreover, the results support the conclusion that the averaging time strongly affects the models' ability to predict the plume's lateral dispersion and maximum concentrations. This reveals a need to embed the influence of averaging time in the model formulations and also the use of more sophisticated techniques such as Large Eddy Simulation (LES), and one particle Lagrangian models such as developed by Thomson (1990), Franzese (2003), Mortarini *et al* (2009) and Manor (2014).

There are important concerns about these results due to the limited database available to investigate the effects of averaging time. The use of meteorological data farther from the site could also reduce the representativeness of the present evaluation. This last drawback is an important limitation, specially, in small time scales evaluated as used in this work. The development of a more robust dataset that comprises various atmospheric conditions and averaging times would allow us to reach more conclusive results.

Conflict of interest

The authors declare that there are no conflicts of interest.

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