

Reflections on Odour Concentration Fluctuations and Nuisance

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In general, odours are perceived by humans through a single respiratory act having a duration of approximately 5 seconds. Moreover, odour concentration varies almost randomly over time, as any other physical variable in a turbulent environment. For this reason, both field measurements and dispersion models used for odour impact assessment purposes should be able to refer to short sampling times, instead of considering concentrations averaged over prolonged periods (i.e. 1 hour). This paper has the aim to investigate the problem of odour concentration fluctuations and their correlation with odour nuisance, thereby briefly discussing the problem of turbulence based on some theoretical considerations and of a few experimental results. The experimental results relevant to the continuous monitoring of instantaneous H₂S concentration in ambient air clearly show that peak concentration exceed 1-hour average concentrations by almost 1 order of magnitude. Finally, the paper discusses the possible implementation of a dispersion model specifically dedicated to odour nuisance evaluation, thereby investigating different solutions to account for concentration fluctuations. The investigation related to the peak-to-mean approach, currently being the most widely used method to evaluate peak concentrations from 1-hour averaged concentrations on a regulatory level, allowed to point out the drawbacks of this methodology, which, despite its simplicity and ease-of-use, is unable to accurately describe the concentration fluctuations phenomena.

1. Introduction

In general, odours are perceived by humans through a single respiratory act having a duration of approximately 5 seconds. From this almost instantaneous analysis, the human olfaction is capable of perceiving a positive or negative olfactory sensation, in the latter case generating a "nuisance".

Moreover, odour concentration varies almost randomly over time with peaks and null values that follow one another apparently without a logic, as any physical variable in a turbulent environment.

For this reason, both field measurements and dispersion models used for odour impact assessment purposes should be able to refer to short sampling times, instead of considering concentrations averaged over prolonged periods (i.e. 1 hour) (Schauberger and Piringer, 2012).

Regarding experimental field measurements, these should be able to capture turbulent fluctuations. From these, it should be possible to establish a characteristic fluctuation (peak concentration) to be correlated to odour nuisance.

By means of dispersion modelling it is necessary to reproduce at least the statistics of fluctuations in order to estimate: i) the average concentration, ii) the statistical distribution of instantaneous concentrations, iii) the peak concentration value.

This work has the aim to point out the problem of the odour concentration fluctuations and their correlation with odour nuisance, thereby briefly discussing the problem of turbulence on the basis of the theory and of some experimental results, and finally proposing some possible models that can be applied in order to take such fluctuations into consideration in odour impact assessment studies.

2. Theoretical considerations about concentration fluctuations

2.1 Atmospheric turbulence

Odour nuisance is related to the emission of odorous compounds from human activities or natural sources. Such emissions can be constant or variable over time. However, even in case of constant emission, odour nuisance can be perceived differently at ground level due to atmospheric conditions, such as wind speed and direction, and turbulence. Turbulent fluctuations, i.e. deviations from the average value depending on the turbulence of the Planetary Boundary Layer (PBL), are thus responsible for the fluctuations of the different scalar variables that characterize the atmosphere, thereby including the concentrations of the odorous substances.

Besides turbulent fluctuations, which are always present in the PBL, in the near-field, i.e. close to the emission point, fluctuations also derive from the natural oscillation of the air, mainly in the horizontal direction, that typically occur in the atmosphere especially in stable conditions (sub-meso motions).

The emitted substances are dragged by turbulent eddies of big dimensions generating the typical oscillating plumes (meandering).

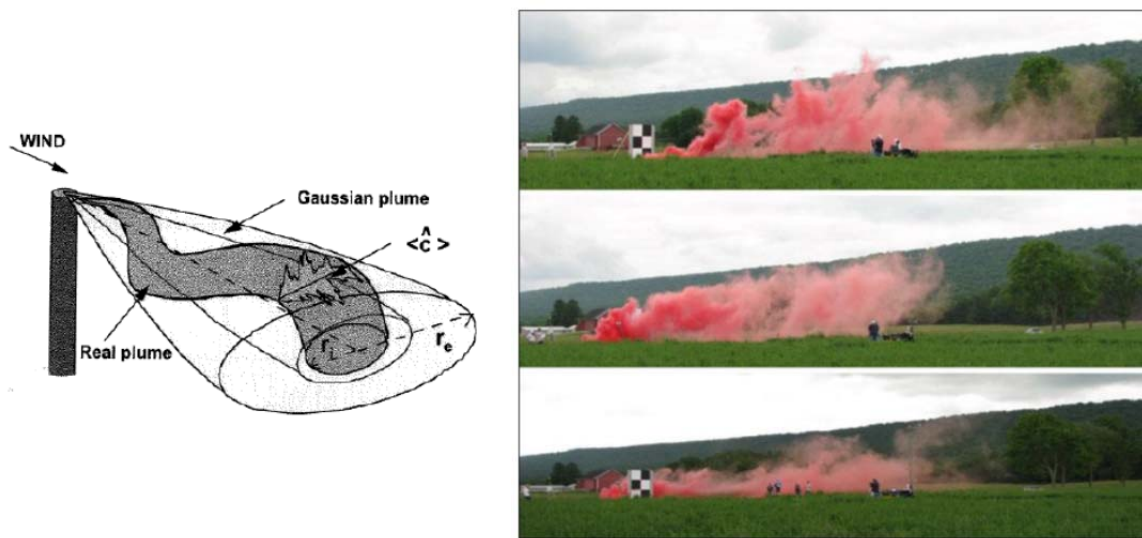


Figure 1: Examples of oscillating plume.

This apparently stochastic variability of the meteorological variables in the PBL derives from the intrinsic chaotic character of the natural laws that rule atmospheric turbulence.

From a quantitative point of view, each meteorological variable is assimilated to a stochastic process, which is completely characterized by:

- the Probability Density Function (PDF) and the relevant Cumulative Density Function (CDF), or
- the infinite statistical moments (i.e. average value, variance, etc.).

The statistical distribution of the instantaneous concentration shall thus be investigated in order to quantify the fluctuations.

2.2 Evaluation of the instantaneous concentration fluctuations

In general, from the elementary measurements, two fundamental parameters can be derived.

The first parameter is the “concentration intensity” (i_c), which is the ratio between the standard deviation of the concentration and the average concentration, as shown in Eq. (1):

$$i_c = \frac{\sigma_c}{C} \quad (1)$$

The second parameter is the so called “peak-to-mean” ratio, which can be defined as:

$$\Psi = \frac{C_p}{C} \quad (2)$$

Where C_p is the value of the p -th percentile.

If the statistical distribution of the instantaneous concentration is known, then the relationship between concentration intensity and peak-to-mean ratio can be defined.

Up to now, there is no univocal definition of the best statistical distribution to be applied (Myline and Mason, 1991), but literature studies point out that i) Log-normal, ii) Weibull, and iii) Gamma distributions are statistical distributions with two parameters that usually well describe experimental results (Finn et al., 2010; Yee et al., 2009). In a very recent and interesting study by Oetti and Ferrero (2017), the authors compare the experimental results described in different works and finally apply a slightly modified Weibull distribution in order to develop a model to assess odour hours for regulatory purposes.

For each distribution with two parameters, the knowledge of the average and of the variance allows the estimation of the two parameters and thus the complete definition of the distribution itself.

On the other hand, the choice of the statistical distribution significantly affects the definition of the peak-to-mean ratio, as shown in Figure 2.

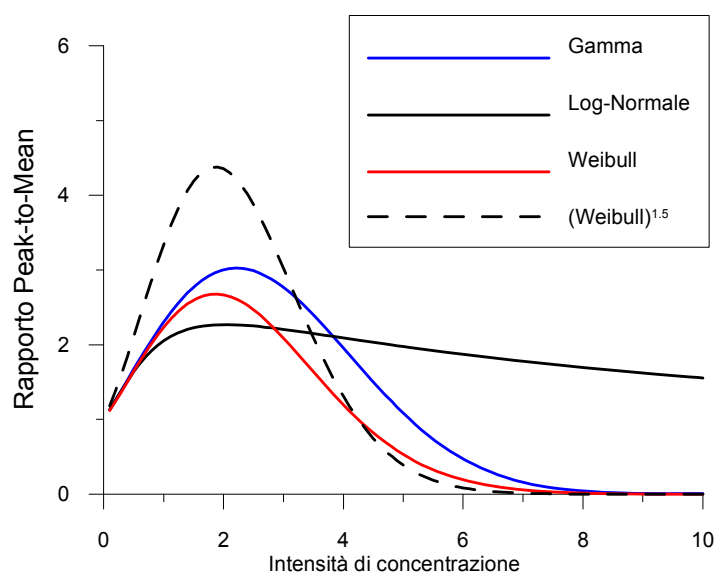


Figure 2: Dependence of the peak-to-mean ratio from the concentration intensity in function of the statistical distribution considered.

3. Experimental: On field observations of H₂S concentration fluctuations in ambient air

A first experimental campaign was carried out in order to verify the entity of the concentration fluctuations of gaseous pollutants in proximity of an open pit used for the storage of pulp and paper residues, located in the municipality of Soriano al Cimino in the province of Viterbo, in Central Italy.

The substance monitored was H₂S, using an analyzer that allows the determination of the instantaneous H₂S concentration, which was evaluated considering an averaging time of 5 seconds. An example of the results of the first monitoring campaign, carried out in February 2016, is shown in Figure 3. In Figure 3, the time step on the abscissa is 1 hour, from which it is clearly visible that sub-hourly concentrations are in most cases very significant. As an example, the H₂S measurements carried out on the 24th February 2016 (right part of the figure), show that the instrument higher detection limit (500 µg/m³) is often exceeded for short periods of time.

A second experimental campaign was then carried out in the municipality of Ceccano, a medieval town (ca. 23000 inhabitants) in the province of Frosinone, having some productive activities located within a radius of 2.5 km from the town centre, during the Autumn of 2017.

During this second campaign, which again involved the continuous measurement of H₂S, instantaneous concentration were plotted against hourly averaged concentrations in order to better highlight the relevance of the concentration fluctuations (Figure 4). It should be noted that the concentrations on the ordinate of Figure 3 are reported in a logarithmic scale: it is possible to observe that peak concentrations are in most cases 1-2 orders of magnitude higher compared to the hourly averaged concentrations.

Regarding odour impact, this in turn means that in situations on which the hourly averaged concentrations are below the odour threshold concentration, significantly higher concentrations may be registered if referring to shorter time periods, thereby resulting in an odour nuisance, as has been already widely discussed in the scientific literature on the matter (Schauberger and Piringer, 2012).

It is therefore necessary that odour dispersion models properly account for these fluctuations in order to be able to effectively describe or predict odour impacts.

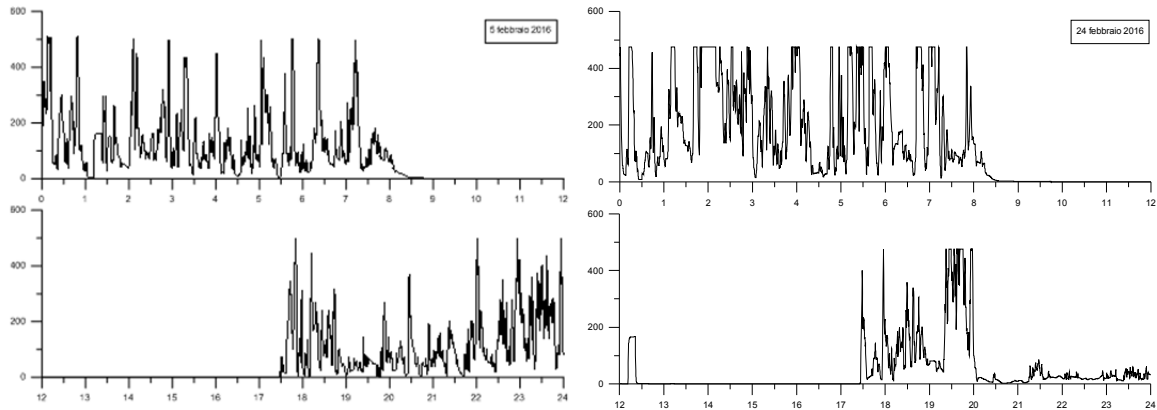


Figure 3: Example of results of the first monitoring campaign: H_2S instantaneous concentrations measured on February 5, 2016 (on the left) and February 24, 2016 (on the right). Concentrations are in $\mu g/m^3$.

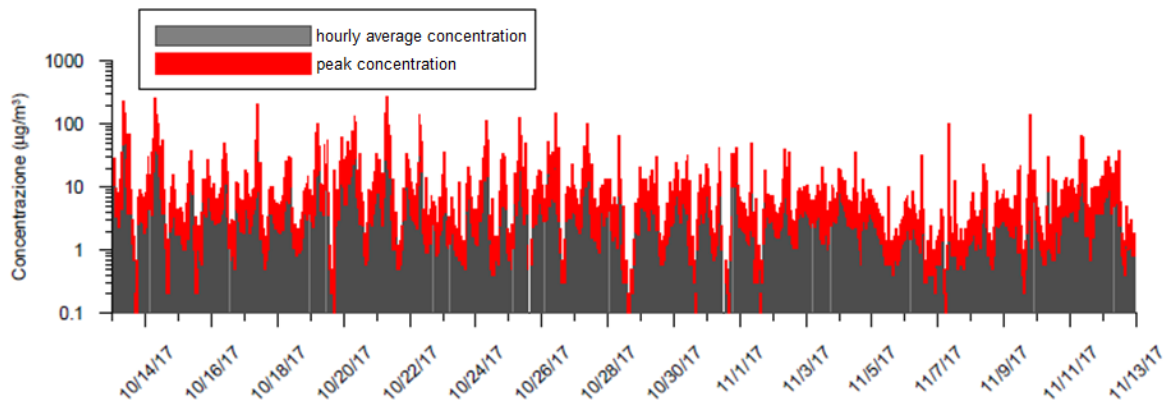


Figure 4: Plots of the hourly averaged H_2S concentrations in $\mu g/m^3$ (grey) vs. instantaneous concentrations (red) in a logarithmic scale.

4. Discussion: Development of dedicated models for odour nuisance assessment

4.1 General requirements

Based on the previous considerations, it becomes clear that a suitable dispersion model to be applied for odour impact assessment should be able to account for odour concentration fluctuations, in order to overcome the limitations of considering only hourly averaged data, which would lead to significant underestimation of the effective odour perception by the population and thus of the odour nuisance.

Thus, besides the general desirable characteristics for air dispersion models, including the fact of having solid mathematical and physical basis, and the capability to properly treat variable emissions, complex orography and both stable and convective atmospheric conditions, dispersion models specific for odours should fulfil some additional requirements.

More in detail, a model suitable for the characterization of odour nuisance, should ideally comprise the following elements:

- A model dedicated to the temporal evolution of odour emissions;

- A “normal” dispersion model capable of providing the odour concentration values for any knot of the sampling grid;
- A model capable of recreating or predicting the spatial and temporal distribution of the odour concentration variance and thus, by using the average concentration fields, to provide the concentration intensity field;
- A statistical model describing the stochastic behaviour of the instantaneous concentration (two-parameter distribution: Log-Normal, Weibull or Gamma).

The most common method used for the purpose of accounting for concentration fluctuations up to now is the semi-empirical “peak-to-mean” approach, which does not fulfil the last two requirements.

4.2 The “peak-to-mean” approach

Different peak-to-mean models can be adopted for estimating peak concentrations. All those models are based on the assumption that, once the average odour concentration in one point of the grid is known, the peak concentration can be obtained directly by multiplying the average concentration by a peak-to-mean ratio, as defined in Eq. (2).

Different variants of this approach have been proposed in literature.

The simplest method is the adoption of a constant peak-to-mean ratio. This is for instance the case for Germany ($\Psi=4$) or for the regional guidelines on odour in Italy ($\Psi=2.3$) (Brancher et al., 2017). Despite its simplicity and ease of application, this model is quite unrealistic and not scientifically supported (Oetl and Ferrero, 2017).

Another possibility is to consider Ψ as a function of atmospheric stability and of the distance from the source, as it is the case for the Austrian regulation (Brancher et al., 2017). This dependence can be expressed by the following relationship:

$$\Psi(x) = 1 + (\Psi_0 - 1) \cdot \exp\left[-\beta \cdot \frac{x}{U \cdot T_L}\right] \quad (3)$$

Where U is the average wind speed, β is a constant, and T_L the Lagrangian time scale. This model is a little bit more complex than the previous one, trying to reproduce some laboratory and field result, but still consists of a simple post-processing of the concentration fields obtained by means of a dispersion model.

Nonetheless, it entails a fluidodynamic inconsistency, due to the fact that according to this model, at far distance from the source ($x \rightarrow \infty$), the peak-to-mean ratio tends to a constant value of 1 ($\Psi \rightarrow 1$), meaning that the peak concentration is equal to the average concentration. This in turn would mean that at a far distance from the source atmospheric turbulence is no longer present, which is obviously untrue.

Finally, there are also some literature works proposing to introduce the variability of Ψ with the transversal direction y (Best et al. 2001):

$$\Psi(x, y) = 1 + (\Psi_0 - 1) \cdot \exp\left(-0.7317 \left[\frac{x}{UT_L}\right]\right) \cdot i_y(y) \quad (4)$$

With:

$$i_y(y) = \exp\left(\frac{y^2}{a\sigma_y^2}\right) \quad (5)$$

Where a is set equal to 2 and σ_y indicates the transversal dispersion parameter (Best et al., 2001). As can be seen from Eq. (4) and (5), the dependence from the transversal distance is exponential, and increases with the distance from the plume barycentre. This gives that Ψ decreases with x , while it increases with y , as confirmed by experimental observations.

Thus, the introduction of the transversal dependence of the function $\Psi(x,y)$ tries to account for experimental evidences, but on the other hand it is difficult to implement, especially if the adopted model is not Gaussian and thus meteorology is no longer homogeneous on the horizontal direction.

Therefore, it can be concluded that peak-to-mean models can be usefully applied in order to gain information about the order of magnitude of the phenomenon, thereby knowing that they are not able to accurately describe the concentration fluctuation phenomena.

4.3 Alternative approaches

Considering the model aspects related to the description of the spatial and temporal evolution of the concentration variance, it is possible to find out that some quite convincing solutions have been proposed in the scientific literature (Oettl and Ferrero, 2017), which seem to be based on more solid fluidodynamic and statistical considerations compared to the peak-to-mean approach.

Without getting too much into detail, which would be outside the scope of this paper, the following categories of modellistic philosophies can be mentioned:

- Models for the transport of variance
- Fluctuating plume models
- Micromixing models
- Lagrangian 2-particle models

Usually, such modellistic philosophies are inserted within the context of a Lagrangian Particle Model (LPM) and, from the point of view of the mathematical implementation, it is logical to presume that they might represent in the next future the natural evolution of the LPMs that are currently used in the context of atmospheric pollution, when atmospheric chemistry is not fundamental.

Thus, the main problems to be solved for the realization of a LPM specifically dedicated to odour nuisance are basically focused on one hand on the choice of the modellistic philosophy to be adopted, and on the other hand on the adoption of a suitable statistical model for the description of the fluctuations.

5. Conclusions

This work discusses the basic concepts related to the problem of odour concentration fluctuations and their correlation with odour nuisance, thereby pointing out the specific necessity to account for such fluctuations in dispersion models dedicated to odour impact and/or odour nuisance assessment. As a matter of facts, fluctuations are particularly important in the case of odour concentration, since the exceedance of the odour threshold also for a short period of time (the time of one breath is estimated to be of about 5 seconds), can already be a cause of discomfort.

The paper presents both some theoretical considerations about concentration fluctuations and a few experimental results regarding the continuous measurement of instantaneous H₂S concentration in ambient air in two locations in Central Italy, showing clearly that peak concentration can exceed 1-hour average concentrations by almost 1 order of magnitude.

Furthermore, the paper discusses the possible implementation of a dispersion model specifically dedicated to odour nuisance evaluation, considering different solutions to account for concentration fluctuations.

The limitations of the simple approach that is currently most widely used for this purpose, i.e. the peak-to-mean ratio models, are discussed, concluding that such models are useful in order to gain information about the order of magnitude of the phenomenon, without allowing to describe it in an accurate way.

Finally, some alternative solutions to be implemented in Lagrangian Particle Models are also mentioned, which will require deeper studies in the next future.

References

- Best P.R., Lunney K.E., Killip C.A., 2001, Statistical elements of predicting the impact of a variety of odour sources, *Water Science and Technology*, 44, 157-164.
- Brancher M., Griffiths K.D., Franco D., de Melo Lisboa H., 2017, A review of odour impact criteria in selected countries around the world, *Chemosphere*, 168, 1531-1570.
- Finn D., Clawson K.L., Carter R.G., Rich J.D., Biltoft C., Leach M., 2010, Analysis of urban atmosphere plume concentration fluctuations, *Boundary-layer meteorology*, 136, 431-456.
- Mylne K.R., Mason P.J., 1991, Concentration fluctuation measurements in a dispersing plume at a range of up to 1000 m, *Quarterly Journal of the Royal Meteorological Society*, 117, 177-206.
- Oettl D., Ferrero E., 2017, A simple model to assess odour hours for regulatory purposes, *Atmospheric environment*, 155, 162-173.
- Piringer M., Knauder W., Petz E., Schaubberger G., 2015, A comparison of separation distances against odour annoyance calculated with two models, *Atmospheric Environment*, 116, 22-35.
- Schaubberger G., Piringer M., 2012, Assessment of separation distances to avoid odour annoyance: interaction between odour impact criteria and peak-to-mean factors. *Chemical Engineering Transactions*, 30, 13-18.
- Yee E., Wang B.C., Lien F.S., 2009, Probabilistic model for concentration fluctuations in compact-source plumes in an urban environment, *Boundary-layer meteorology*, 130, 169-208.