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Abstract: Waste Water Treatment Plants are known to have significant
emissions of several pollutants from the different treatment phases.
Among such pollutants, volatile organic and inorganic compounds, often
having low odour detection thresholds, cause odour nuisance to the
population. One of the purposes of the present work is to determine which
are the more suitable methodologies to assess the odour emissions from
liquid passive area sources, by means of a thorough study of the models
capable of describing the volatilization phenomena of the odoriferous
compounds from such sources. Several different models were evaluated for
the open field emission, selecting the most appropriate one. Moreover,
the models describing volatilization under a forced convection regime
inside a wind tunnel device have been investigated, in order to describe
the situation inside this sampling device, typically used for sampling on
liquid sources. By means of experimental tests involving pure liquid
acetone and pure liquid butanone, it was verified that the model more
suitable to describe precisely the volatilization inside the sampling
hood is the model for the emission from a single flat plate in forced
convection and laminar regime, with a fluid dynamic boundary layer fully
developed and a developing mass transfer boundary layer. The
proportionality coefficient for the model was re-evaluated in order to
account for the specific characteristics of the adopted wind tunnel
device. Due to the differences between the fluid dynamic conditions in
the open field and inside the hood, it was deemed useful to devise a
correlation that - according to the flush rate adopted inside the chamber
- is capable of computing the wind velocity at a 10 m height that would
cause the same emission flux that is estimated from the analysis of the
sample collected with the wind tunnel.
Finally, the field of application was clearly defined for the considered
models during the project, discussing the two different kinds of
compounds commonly found in emissive liquid pools or liquid spills, i.e.
gas phase controlled and liquid phase controlled compounds.

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Assessing Odour Emissions from Liquid Area Sources

Federico Lucernoni, Laura Capelli, Valentina Busini, Selena Sironi

Our work discusses a methodology for the determination of emission rates of pollutants like VOCs and odours from liquid area sources like waste water treatment plant tanks. The method entails sampling on the (simulated) liquid source with a Wind Tunnel device, calculating the Specific Emission Rate (SER) and finally rescaling the SER in order to be representative of the real conditions, outside the device, in the open field.

In the research, laboratory experiments were performed adopting a Wind Tunnel and a simulated liquid pool, investigating the emissions of two VOCs, acetone and butanone. We study a particular kind of sampling device, a Wind Tunnel, for field applications on liquid passive are sources, such as waste water treatment tanks. The present research is part of the ongoing effort in different parts of the world aimed to gaining a better understanding of how the hood devices work, what happens exactly inside them during sampling and how to use the obtained data in order to be able to obtain meaningful conclusions and insights concerning the problem of emissions assessment from area sources. Such problems are addressed and discussed also in other papers recently published on Atmospheric Environment (Parker et al., 2013).

The main results of the present study are highlighted, discussing how the proposed procedure for rescaling the emission rate to the open field scenario can be really useful, especially for atmospheric dispersion modelling purposes.

As novelty of this work and its contributions to the field of liquid surfaces emissions assessment and pollutants dispersion modelling, please consider our paper "Assessing Odour Emissions from Liquid Area Sources" for publication on Atmospheric Environment Journal.

We hope this paper will be of interest for the readers of the journal.

Best regards,

Federico Lucernoni and Laura Capelli

Highlights for the article "Assessing Odour Emissions from Liquid Area Sources":

- Liquid area surfaces are significant sources of odours and VOCs emissions.

- Lawmakers concur that the emissions should be monitored.

- There are no widely accepted methodologies for emissions assessment.

- The paper tries to provide a method for evaluating liquid sources.

- The procedure entails also the recalculation of the emission in the field.

1   **ASSESSING ODOUR EMISSIONS FROM LIQUID AREA SOURCES**

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7

8   **ABSTRACT**

9   Waste Water Treatment Plants are known to have significant emissions of several pollutants from the

10   different treatment phases. Among such pollutants, volatile organic and inorganic compounds, often having

11   low odour detection thresholds, cause odour nuisance to the population. One of the purposes of the

12   present work is to determine which are the more suitable methodologies to assess the odour emissions

13   from liquid passive area sources, by means of a thorough study of the models capable of describing the

14   volatilization phenomena of the odoriferous compounds from such sources. Several different models were

15   evaluated for the open field emission, selecting the most appropriate one. Moreover, the models

16   describing volatilization under a forced convection regime inside a wind tunnel device have been

17   investigated, in order to describe the situation inside this sampling device, typically used for sampling on

18   liquid sources. By means of experimental tests involving pure liquid acetone and pure liquid butanone, it

19   was verified that the model more suitable to describe precisely the volatilization inside the sampling hood

20   is the model for the emission from a single flat plate in forced convection and laminar regime, with a fluid

21 dynamic boundary layer fully developed and a developing mass transfer boundary layer. The

22 proportionality coefficient for the model was re-evaluated in order to account for the specific

23 characteristics of the adopted wind tunnel device. Due to the differences between the fluid dynamic

24 conditions in the open field and inside the hood, it was deemed useful to devise a correlation that –

25 according to the flush rate adopted inside the chamber – is capable of computing the wind velocity at a 10

26 m height that would cause the same emission flux that is estimated from the analysis of the sample

27 collected with the wind tunnel.

28 Finally, the field of application was clearly defined for the considered models during the project, discussing

29 the two different kinds of compounds commonly found in emissive liquid pools or liquid spills, i.e. gas

30 phase controlled and liquid phase controlled compounds.

31

34

35 **1. INTRODUCTION**

36 Waste Water Treatment Plants (WWTP) are known to have significant emissions of several pollutants from

37 the different treatment phases (Capelli et al., 2009a; Prata Jr. et al., 2016a; Santos et al., 2012). Among

38 them, volatile organic and inorganic compounds, often having low odour detection thresholds, are typically

39 a cause of odour nuisance to the near-living population (Capelli et al., 2009a; Parker et al., 2013; Prata Jr. et

40 al., 2016a). Whereas point source, such as stacks (Bockreis and Steinberg, 2005; Capelli et al., 2013) are

41 typically subject to periodical emission monitoring, this is not the case for area sources, such as WWTP

42 tanks, which, however, often represent the main source of VOCs and odour emissions in this kind of plants

43 (Capelli et al., 2008; 2009a). For this reason, area sources should be considered as well as point sources for

44 odour emission characterization and impact assessment, as already provided by some regulations on the

45 matter (D.g.r. n. IX/3018, 2012; VDI 3880, 2011). However, assessing odour emissions from area sources

46   such as wastewater treatment tanks, i.e. the so-called "passive" area sources, which means without

47   outward flow, is typically a rather complicated task, since there is no straightforward nor established

48   procedure (Capelli et al., 2013). First of all, already choosing the most appropriate technology for sampling

49   is difficult among the different ones that have been proposed and are currently applied for this scope

50   (Muezzinoglu, 2003; Blunden and Aneja, 2008; Beghi et al., 2012; Rumsey et al., 2012; Hentz et al., 2013;

51   Hudson and Ayoko, 2008; Bliss et al., 1995; Kim and Park, 2008). In recent studies there is evidence that so-

52   called "hood methods", entailing an enclosure of some sort (e.g., wind tunnels), whereby emission rates

53   are derived from the data regarding the concentration at the outlet of the sampling device combined with

54   the dimensions of the device and the operating conditions, should be preferred for this purpose (Hudson

55   and Ayoko, 2008; Capelli et al., 2013). There are different types of dynamic hoods, mainly distinguished

56   between Flux Chambers (FC) (Klenbusch, 1986; Prata Jr. et al., 2016a) and Wind Tunnels (WT) (Smith and

57   Watts, 1994; Jiang et al., 1995; Capelli et al., 2009b; Parker et al., 2010), the latter being considered for this

58   study. With hood methods, the assessessment of the odour emission rate (OER) involves 3 phases: on-site

59   sampling (Capelli et al., 2009b; Koziel et al., 2005; Sironi et al., 2014a), sample analysis (CEN EN 13725,

60   2003) and data elaboration (Lucernoni et al., 2016; Ranzato et al., 2012). With the odour concentration it is

61   possible to evaluate the Specific Odour Emission Rate (SOER), that is the odour units emitted from the

62   source per surface and time unit [$ou/m^2/s$] referred to the neutral sweep air flow rate used during sampling

63   (Capelli et al., 2009b).

64   The main problem is that emission rates, calculated as above-mentioned (Capelli et al., 2013), refer solely

65   to the specific sampling conditions inside the hood, and therefore are not representative of the effective

66   emissions from the source in the open field when subject to natural ventilation (Hudson and Ayoko, 2009;

67   Leyris et al., 2005). In order to evaluate the SOER occurring in the open field at different wind conditions, it

68   is necessary to scale the value obtained for the hood to the real situation, thereby adopting a suitable

69   correlation. While sampling and analysis methodologies are fairly established (analysis more than sampling,

70   as previously discussed), the elaboration of the datum is still an open issue. The studies of Sohn et al.

71   (2005) and Sironi et al. (2014b) propose to use a correlation for the re-calculation of the SOER for the open

72 field based on the Prandtl's boundary layer theory for laminar flow, assuming a proportionality of the SOER

73 measured at the sampling conditions (i.e. sweep air velocity) and the SOER at the actual wind speed with

74 the square root of the ratio between actual wind speed and sweep air velocity:

75
$$SER_{field} = SER_{WT} \left(\frac{U_{10}}{U_{WT}}\right)^{0.5}$$
(1)

76 The adoption of a dependence with the square root of the air velocity – typical of the laminar flow – does

77 not account for the fact that in real situations the flow above a liquid area surface is typically turbulent

78 (Sutton, 1934). Moreover, this approach clearly entails a big approximation of the real situation, since by

79 relating the actual wind speed – conventionally measured at a height of 10 m above ground – with the

80 sweep air speed inside the wind tunnel, on one hand it is assumed that the wind profile is constant with the

81 height, which is typically not the case, the wind profile can be described by means of specific mathematical

82 relationships (Bonan, 2005; Cook, 1997; Drew et al., 2013; Tieleman, 2008), and on the other hand it

83 doesn't consider the difference between open-field conditions and forced convection inside an enclosure

84 (WT). For these reasons, this work aims to investigate a suitable model to relate the datum obtained from

85 the WT to the real situation. For this purpose, a suitable model based on Prandtl's boundary layer theory

86 accounting for the volatilization inside the hood (Perry, 1997; Incropera et al., 2007) was experimentally

87 verified and then related to a model describing the volatilization in the open field, thereby referring to the

88 semi-empirical models existing for the estimation of emissions from liquid pools as a function of the wind

89 speed. This relationship allowed to establish a new correlation for the recalculation of the SOER to the

90 actual wind speed in the open field, thereby discussing reliability and field of application of this new

91 proposed model.

92

93 **2.   MATERIALS AND METHODS**

94 2.1. The Wind Tunnel device

95 The WT adopted for this work was designed and developed by the Olfactometric Laboratory at Politecnico

96 di Milano. The structure of the hood is described in detail by Capelli et al. (2009b) and is depicted in Fig. 1

97 and 2. The central body has a 25x50 cm base section and is 8 cm high. The hood is open at the bottom and

98   is laid on top of the emissive surface. The body has two converging sections at the extremes, connected to

99   the inlet and outlet of the chamber that can be closed with specific fittings allowing to feed the neutral air

100 at the inlet and collect the sample at the outlet. The WT is made of PVC and is equipped with buoyant parts

101 that allow sampling on liquid sources.

102

103 *Figure 1: The LabOlf Wind Tunnel*
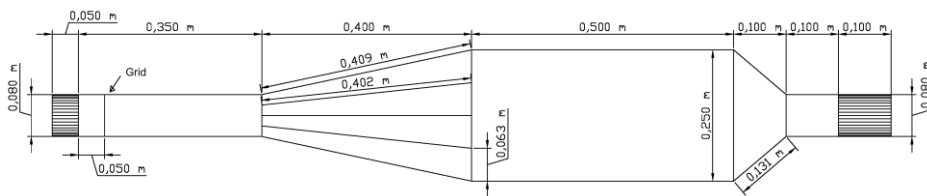
104

105 *Figure 2: The Wind Tunnel scheme*

106 2.2. <u>Volatilization model for the open field</u>

107 In the scientific literature there are several models that have been proposed in order to describe the

108 volatilization phenomena of chemical compounds from liquid pools in the open field. Such models, typically

109 developed in the field of industrial safety, for the most part rely on the theory developed by Sutton (1934).

110 Sutton for his research (1934) considered a pool of a pure liquid volatile compound, subject to the wind

111 action in the open field and he evaluated the dependence of the emission rate on the major factors that

112 can influence the phenomenon. The resulting correlation can be expressed as:

113 $$SER = K\, U_h^{0.78}\, a^{0.89}\, x_0^{-0.11}$$ (2)

114 Where: SER is the Specific Emission Rate; K is the proportionality constant; U_h is the wind speed at the

115 considered height; a is a constant depending on the physical characteristics of the atmosphere; x_0 is the

116 characteristic dimension of the pool in the leeward direction.

117 Relying on Sutton's dissertation (1934), several later studies have been able to determine semi-empirically

118 the constant terms K and a (Kawamura and MacKay, 1987; Committee for the Prevention of Disasters,

119 1992). The values of these two parameters are usually what distinguish one model from the other. Among

120 the variety of this kind of models, here the one adopted in the text "Methods for the calculation of physical

121 effects" (Committee for the Prevention of Disasters, 1992) is considered because it has been validated by

122 many authors and with different sets of data (Pasquill, 1943; Clancey 1974; Jeulink, 1983; MacKay and

123 Matsugu, 1973). According to this model the emission in the open field can be expressed as:

$$SER = 0.002\ U_{10}{}^{0.78} X^{-0.11}\ M_i\ \frac{Pv(T_{pool})}{RT_{pool}} \tag{3}$$

125 Where: SER is the Specific Emission Rate in [kg/m^2/s]; M_i is the molar mass of the evaporating compound

126 in [kg/mol]; $P_v(T_{pool})$ is the vapour pressure of the liquid compound at the surface temperature in [Pa]; R

127 is the universal gas constant equal to 8.314 [J/mol/K]; T_{pool} is the surface temperature of the pool in [K];

128 U_{10} is the wind velocity at 10 m in [m/h]; X is the characteristic dimension of the pool in the leeward

129 direction in [m]; Sc is the mixed Schmidt's number (i.e. air's kinematic viscosity over compound's

130 diffusivity).

131

132   2.3. Volatilization models inside the Wind Tunnel

133 Since there are no available models in the literature for the particular situation of emissions inside a WT,

134 among the various models (Perry, 1997; Incropera et al., 2007; Bejan and Kraus, 2003; Shah and London,

135 1978; Bliss et al., 1995), the one deemed more appropriate for the case at hand is the model for mass

136 transfer under forced convection over a single flat emissive surface in laminar regime. In fact, for the air

137 velocity range considered (Capelli et al., 2009b), the fluid dynamic regime is fully developed laminar. Thus,

138 the thickness of the fluid dynamic boundary layer can be assumed equal to the 50% of the hood's height

139 (Bejan and Kraus, 2003) and the mass transfer boundary layer can be computed as:

$$\delta_m = \delta_f\ Sc^{-\frac{1}{3}} \tag{4}$$

141 Where: δ_f is the fluid dynamic boundary layer thickness in [m]; δ_m is the mass transfer boundary layer

142 thickness in [m]; Sc is the mixed Schmidt's number ($\frac{D_i}{\nu}$).

143   From this correlation, it is easy to see that for compounds with $Sc > 1$, as the case under evaluation, the

144   mass transfer boundary layer develops inside the fluid dynamic one, with $\delta_m \leq \delta_f$ and therefore it is

145   possible to assume that the top wall of the hood has a negligible effect on the volatilization and is non-

146   influencing for the mass transfer phenomenon. Anyway, for the sake of comparison, the model for the

147   situation of internal flows between to emissive plates in laminar regime was considered as well (Perry,

148   1997; Incropera et al., 2007).

149   It is possible to write the mass balance between the inlet and outlet of the WT, obtaining:

150
$$Q\,C_{out} = Q\,C_{in} + K_{c,ave}\,(C_i - C_B)\,A \tag{5}$$

151   Where: Q is the neutral air flow rate flushed in the WT in [m^3/s]; C_{out} is the emitted compound

152   concentration at the outlet in [mol/m^3]; C_{in} is the emitted compound concentration at the inlet in

153   [mol/m^3], that is 0 if neutral air is used; $K_{c,ave}$ is the convective mass transfer coefficient, averaged over the

154   exchange length, in [mol/m^3]; A is the base area of the WT in [m^2], C_i is the gas-liquid interface

155   concentration of the emitted compound in [mol/m^3]; C_B is the emitted compound concentration in the bulk

156   of the gas phase inside the hood in [mol/m^3], which can be assumed equal to 50% of the outlet

157   concentration, taking it as the average between inlet and outlet concentrations with a 0 inlet concentration

158   (Bejan and Kraus, 2003). Thus:

159
$$C_{out} = \frac{K_{c,ave}\,C_i\,A}{\left(Q + \frac{K_{c,ave}\,A}{2}\right)} \tag{6}$$

160
$$SER = \frac{C_{out}\,Q\,M_i}{A} \tag{7}$$

161   The values of $K_{c,ave}$ change from the "single surface" model to the "parallel surfaces" model, as here

162   reported in Eq. (8) and Eq. (9) respectively (Perry, 1997; Incropera et al., 2007):

163
$$K_{c,ave} = 0.664 \left(\frac{D_i^4}{L_{WT}^3\,\nu}\right)^{\frac{1}{6}} U_{WT}^{1/2} \tag{8}$$

164
$$K_{c,ave} = 1.40 \frac{D_i^{\frac{2}{3}}\,U_{WT}^{\frac{1}{3}}}{L_{WT}^{\frac{1}{3}}\,h^{\frac{1}{3}}} \tag{9}$$

165 Where: D_i is the compound's molecular diffusivity in air in [m^2/s]; L_{WT} is the length of the WT central body

166 in [m]; ν is the air's kinematic viscosity in [m^2/s]; h is the height of the WT in [m]; U_{WT} is the air velocity

167 inside the hood in [m/s].

168

169 2.4. The experimental tests

170 In order to evaluate which volatilization model suits better the case under study and describes more

171 accurately the emission phenomenon inside the WT, it was decided to perform a set of experimental runs

172 with evaporating pure compounds. It was decided to adopt acetone first, as it is easily detectable by a GC-

173 TCD and is volatile enough to be considered a gas-phase controlled compound for emissions (Fingas, 1998;

174 2013) and it is also a reasonably safe compound to use. Then, aiming to have further confirmation of the

175 validity of the model for different substances, the experiments were performed also with another

176 compound capable of providing a confirmation of the outcomes obtained with acetone. The chemical

177 chosen for this second set of experiments is butanone, also a compound that can be easily detected via GC-

178 TCD and reasonably safe.

179 For the tests, a small PE tank filled with the pure liquid compound, exactly fitting the WT central body, was

180 placed under the hood simulating the liquid source. Then, the cylinder-fed neutral air was flushed through

181 the inlet of the chamber, at different velocities, ranging from 0.0096 to 0.0527 m/s. In order to have a

182 uniform and reliable collection of the gaseous sample at the outlet, a PET tube, equipped with a sampling

183 port, was connected to the outlet of the WT. The sample was collected by means of a Nalophan® bag and a

184 sampling vacuum pump (Capelli et al., 2009b; Koziel et al., 2005; Sironi et al., 2014a). The sample was then

185 analysed by means of Dynamic Olfactometry (CEN, 2003) in order to determine its odour concentration.

186

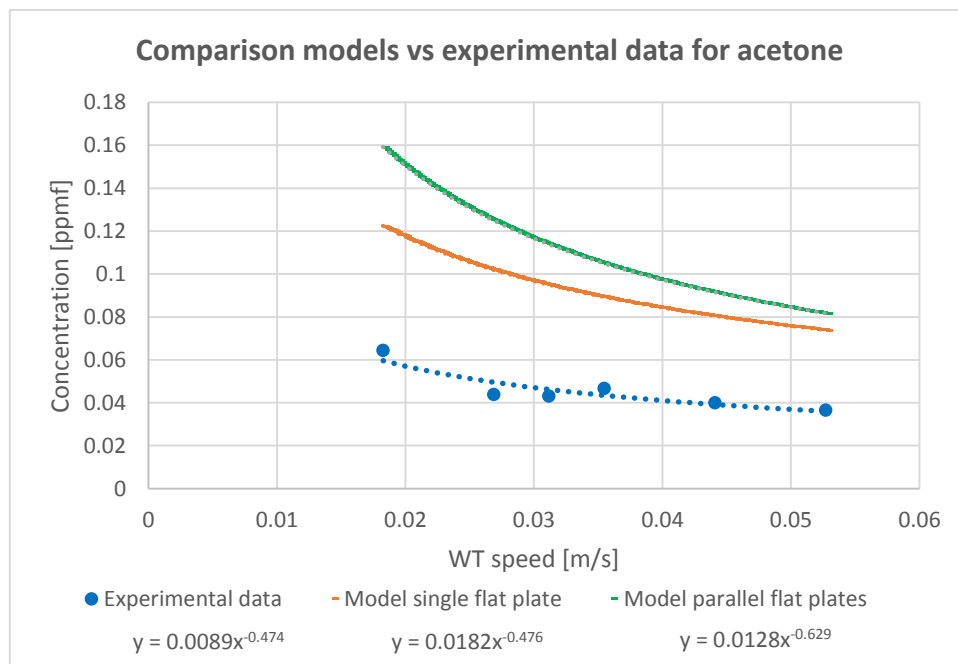187 **3. RESULTS AND DISCUSSION**

188 3.1. Experimental results and model validation

189 The samples analysis via GC-TCD allowed to obtain the outlet concentrations expressed as acetone or

190 butanone ppm for each test. Since the concentration is a function of pool's surface temperature, for each

191 datum the value was expressed also as ppm fraction [ppmf], normalizing the values dividing by the

192   saturation concentration for the compound at the surface temperature of that specific run. This procedure

193   allows to compare results obtained in different days with different temperatures and confront the

194   experimental data with the forecasts of the two models considered (i.e. single and double plate). Fig. 3 and

195   Fig. 4 report the values of concentration expressed as ppm fractions [ppmf], in function of the speed of the

196   neutral flushing air inside the WT. The experimental data, for clarity's sake, are reported in Table 1 and

197   Table 2 for acetone and butanone respectively.

Comparison models vs experimental data for acetone

Experimental data: $y = 0.0089x^{-0.474}$
Model single flat plate: $y = 0.0182x^{-0.476}$
Model parallel flat plates: $y = 0.0128x^{-0.629}$

198

199   *Figure 3. Comparison model-experimental data for acetone*

200   *Table 1: Experimental data for acetone*

Q,air [l/h]	v,WT [m/s]	C,exp [ppmf]
1210	0.01826	0.06445
1790	0.02687	0.04396
2070	0.03117	0.04322
2360	0.03548	0.04672
2930	0.04409	0.03997
3500	0.0527	0.0366

201

Comparison models vs experimental data for butanone

Concentration [ppmf] vs WT speed [m/s]

● Experimental data   – Model single flat plate   – Model parallel flat plates
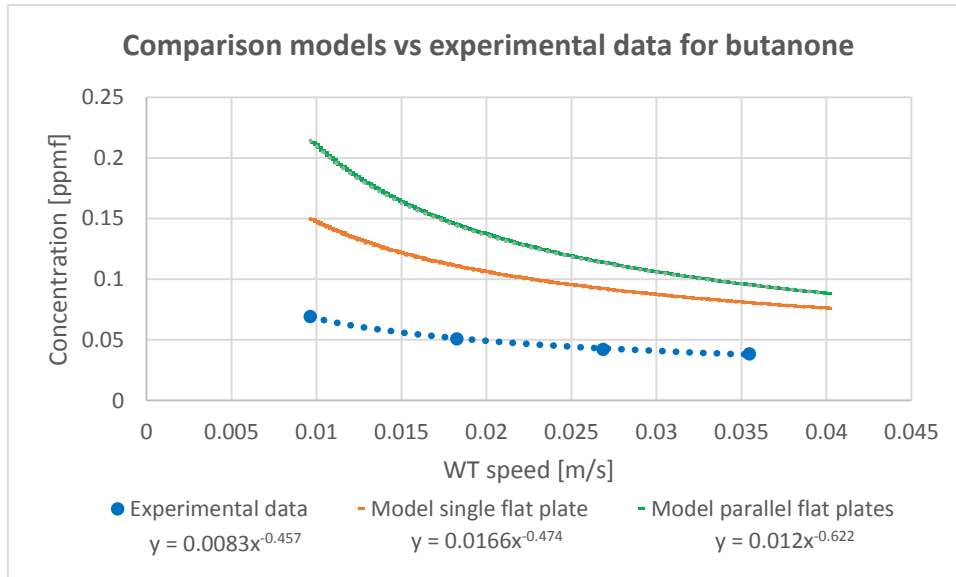 y = 0.0083x^-0.457   y = 0.0166x^-0.474   y = 0.012x^-0.622

202

203   *Figure 4: Comparison model-experimental data for butanone*

204   *Table 2: Experimental data for butanone*

Q,air [l/h]	v,WT [m/s]	C [ppmf]
640	0.00964	0.0692
1210	0.01826	0.051
1790	0.02687	0.04228
2360	0.03548	0.03855

205

206   It is possible to observe how the exponential dependency of concentration on air velocity obtained through

207   the experiments is well described by the volatilization model for the single flat plate geometry. However, it

208   is possible to see also an offset between experimental points and model prediction that seems to

209   overestimate the concentration. Since mass transfer models have a semi-empirical nature and the peculiar

210   case of the WT is somewhat different from the flat plate geometry, it was deemed reasonable to evaluate a

211   new proportionality coefficient for the WT geometry that would fit better the situation at hand,

212   substituting it to the value of 0.664 present in the original model. This would optimize the data-fitting of

213   the model when applied to the specific case of the WT device. The values obtained are 0.33 for acetone and

214   0.30 for butanone. Therefore, an average value was considered of 0.315. Inserting this new coefficient into

215   the equation for the definition of the convective mass transfer coefficient, a new expression is obtained:

216
$$K_{c,ave} = 0.315 \left(\frac{D_i^4}{L_{WT}^3 \, \nu} \right)^{\frac{1}{6}} U_{WT}^{1/2} \tag{10}$$

3.2. Correlation between wind speed in open field and air velocity inside the Wind Tunnel

Once the volatilization model has been defined, it was possible to proceed to the derivation of the formula for the estimation of the wind speed in the open field, at 10 m, that would cause the same emission rate from the source as the one obtained with the WT device $U_{10}{}^*$. This is the velocity that will be used for the recalculation of the SER, at the actual value of the wind speed recorded at 10 m by a meteorological station, starting from the WT data. The equations for the assessment of the SER in the two cases are equalled, adopting Eq. (3) for the open field and Eq. (5)-(7) for the WT, yielding:

$$0.002 U_{10}^{*0.78} X^{-0.11} M_i C_i = \frac{0.315 \left(\frac{D_i^4}{L^3 \nu}\right)^{\frac{1}{6}} U_{WT}^{\frac{1}{2}} C_i}{Q + 0.315 \left(\frac{D_i^4}{L^3 \nu}\right)^{\frac{1}{6}} U_{WT}^{\frac{1}{2}} \frac{A}{2}} Q M_i \tag{11}$$

It is possible then to get the value of the velocity at 10 m ($U_{10}{}^*$), as a function of the velocity in the WT (U_{WT}), as here shown:

$$U_{10}{}^* = \sqrt[0.78]{157.5 \frac{L^{-0.39} \nu^{-\frac{1}{6}} U_{WT}^{0.5} h}{h D_i^{-0.67} + 0.1575 L^{0.5} \nu^{-\frac{1}{6}} U_{WT}^{-0.5}}} \tag{12}$$

From the application of such correlation, the emission rate value can be then scaled according to the actual wind velocities of interest, as depicted in Eq. (13):

$$SER_{field} = SER_{WT} \left(\frac{U_{10}}{U_{10}{}^*}\right)^{0.78} \tag{13}$$

This correlation relies on the exponential dependency between the emission rate and the wind velocity for turbulent conditions. In order to investigate the differences between the proposed model and the SER scaling procedure commonly adopted (Sohn et al., 2005; Sironi et al., 2014b), it was deliberately chosen a concentration of 1000 ppm at the outlet of the WT, with an air speed inside the chamber of 0.035 m/s. The chosen parameters values are summarized in Table 3:

Table 3: Parameters values chosen for the comparison

v,WT [m/s]	c,out [ppm]	SER,WT [mol/m2/s]	U,10 range [m/s]
0.035	1000	0.000233	0-5

239   By applying the model and comparing the forecast with the new model and the one obtained with the

240   "regular" procedure, it was possible to obtain the plots depicted in Fig. 5.
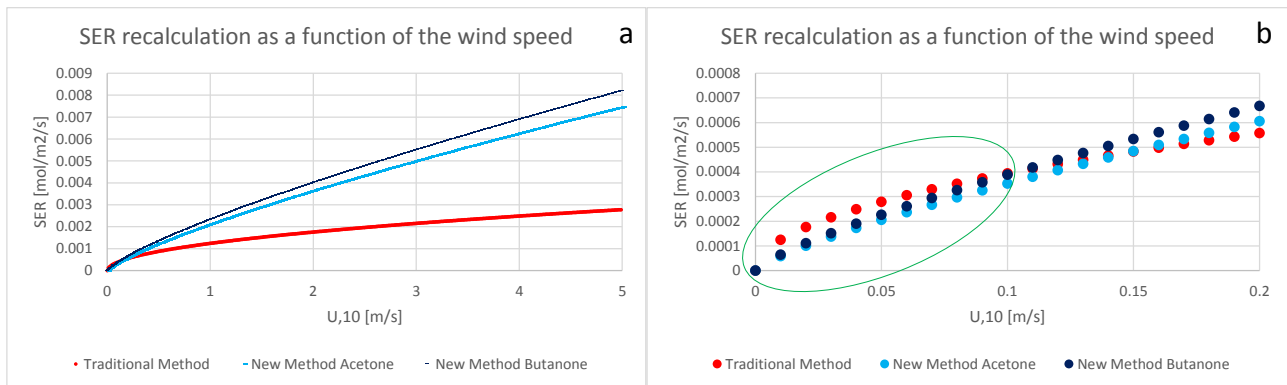
242   *Figure 5: SER recalculation method for acetone (blue line) vs SER recalculation method for butanone (purple*

243   *line) vs traditional SER recalculation method (red line) as a function of the wind speed (Fig. 5a). Zoom for the*

244   *0-0.2 wind speed U,10 range (Fig. 5b).*

245   It is possible to see that the two new models behave quite differently with respect to the old one,

246   forecasting SER values that become more distant as the wind speed at 10 m increases. The new models

247   indicate much higher emissions for strong winds with respect to the "regular" method. This difference can

248   be explained as mostly due to the different exponent used for the SER recalculation in the two

249   methodologies. In fact, the "regular" method considers the air speed inside the WT equal to the wind speed

250   in the open field at the same height and relies on the scaling formula reported in Eq. (1). Looking at Fig. 5b,

251   it is possible to see how for low wind speeds the new models actually forecast lower emissions with respect

252   to the traditional method. This is due to the fact that for such speed range, the velocities are too low for

253   the new exponent (0.78) effect to be predominant and it is possible to observe how the wind speed

254   recalculation formula (Eq. 12) leads to forecasting lower emissions.

255   As discussed above, the most suitable exponent to use in the SER recalculation formula is 0.78, as it is an

256   expression for scaling the emission rate in the open field, in the lower atmosphere, that is typically

257   turbulent.

258

259   **4.   GENERALIZATION TO ODOUR EMISSIONS**

260 In order to be able to generalize the models previously presented to the field of olfactometry, it is

261 necessary to clearly define their field of application.

262 According to the studies carried out by Fingas (1998; 2013), the volatilization phenomena concerning

263 compounds like paraffins with a chain longer than 10 carbon atoms, are not gas phase controlled (GPC) and

264 therefore the velocity of the wind swiping the source's surface has no influence on the SER.

265 The research by Parker et al. (2010), investigating the volatilization of the compounds usually found in the

266 wastewaters of industrial activities or animal feeding operations, has highlighted that – if the liquid source

267 is a diluted aqueous solution – the solubility value for the considered compounds in water plays a crucial

268 role in determining what phenomenon is controlling the overall volatilization process. In order to compare

269 the different chemical species, the non-dimensional Henry's constant (H_{cc}) was referred to. The non-

270 dimensional Henry's coefficient (H_{cc}) can be determined from the Henry Law coefficient (H_{cp}), according to

271 Eq. 14:

272
$$H_{cc} = \frac{1}{H_{cp} * R * T} \tag{14}$$

273 Where: H_{cc} is the non-dimensional Henry coefficient; H_{cp} is the dimensional Henry coefficient in

274 [mol/m^3/Pa]; R is the universal gas constant in [J/mol/K]; T is the room temperature in [K].

275 It was found out that only for the compounds with a very low dimensionless Henry's constant (H_{cc} < 0.001)

276 the velocity of the gas sweeping the surface of the liquid surface is the controlling phenomenon in the

277 volatilization process and such compounds are defined Gas Phase Controlled (GPC). On the contrary, for

278 compounds characterized by a high non-dimensional Henry's constant (H_{cc} >> 0.001), typically

279 characterized by a low solubility in water, the effect of the wind velocity becomes negligible, compared to

280 the phenomena occurring in the liquid phase bulk. For intermediate values of the dimensionless Henry's

281 constant, both phenomena in the liquid phase and in the gas phase play an important role and should be

282 accounted for properly. In order to better understand the range of H_{cc} values for the distinction between

283 GPC, LPC and "intermediate" compounds, it is possible to refer to the dissertation proposed by Parker et al.

284 (2010). Relying on the work of Parker et al. (2010), in fact, it is possible to infer that compounds with

285 $H_{cc} < 0.0006$ can be classified as GPC, compounds with $H_{cc} > 0.0562$ can be classified as LPC and

286 compounds with $0.0006 < H_{cc} < 0.0562$ can be classified as "intermediate".

287 In Table 4, the values for the diffusivities in air are listed along with the non-dimensional Henry coefficients

288 for several odorous compounds usually found in WWTP tanks, both GPC and LPC, indicating as well for each

289 compound whether it is Gas Phase Controlled (GPC) or Liquid Phase Controlled (LPC). In order to compile

290 Table 4, the dimensional Henry coefficients were taken from R. Sander (2015). Table 4, for clarity's sake,

291 was compiled grouping the LPC compounds in the upper rows and GPC compounds in the lower rows.

292 *Table 4: Values for the diffusivities and non-dimensional Henry's coefficients for the considered odorous*

293 *compounds*

Compound	Diffusivity in air @ 20 °C [m2/s]	H,cc @ 25 °C [-]	Type
Methyl-mercaptan	1.21398E-05	0.106162606	LPC
Ethyl-mercaptan	1.01292E-05	0.144077822	LPC
Benzene	8.70165E-06	0.224121056	LPC
Toluene	7.82022E-06	0.268945268	LPC
Tetrachloroethylene	7.27439E-06	0.733487094	LPC
Orto-xylene	7.15497E-06	0.201708951	LPC
Alfa-pinene	5.85698E-06	5.451593266	LPC
Ethylbenzene	7.15497E-06	0.288155644	LPC
Butanol	8.69139E-06	0.000336182	GPC
Phenol	8.27155E-06	2.12325E-05	GPC
Butyrric Acid	8.43937E-06	8.96484E-06	GPC
Propionic Acid	9.55295E-06	7.33487E-06	GPC
Valeric Acid	9.45326E-06	3.36182E-05	GPC
Ethanol	1.18929E-05	0.000212325	GPC
Acetic Acid	1.12227E-05	1.00854E-05	GPC
Acetone	1.03057E-05	0.001613672	~ GPC
Butanone	8.89749E-06	0.002241211	~ GPC

294

295 In order to evaluate the variability of the correlation between wind speed in the open field and air velocity

296 inside the WT, in function of the change in the compound's diffusivity in air, the same calculations

297 described in par. 3.2 were performed to assess the SER for a set of odoriferous compounds typically found

298 in liquid passive area sources. The compounds considered are characterized by low values of the non-

299 dimensional Henry's constant ($< 10^{-3}$), thus fall within the field of application of the models (i.e. GPC

300 compounds).
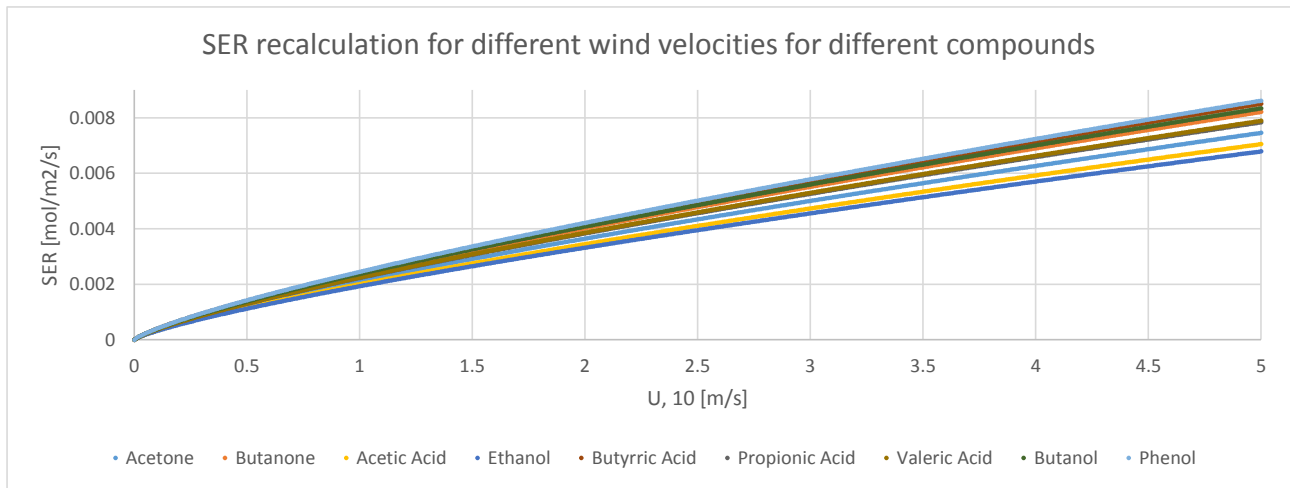
Figure 6: SER recalculation for several odorous compounds

It can be seen how Fig. 6 focuses only on the so-called Gas Phase Controlled compounds characterized by values of the non-dimensional Henry coefficient H_{cc} lower than 0.001 (GPC in Table 4). In particular, the compounds considered in Fig. 6 are: acetone, butanone, acetic acid, ethanol, butyric acid, propionic acid, valeric acid, n-butanol and phenol. The resulting SER values showed a maximum variability of 12% (Fig. 6) if GPC compounds only are considered. Such value of variability can be considered acceptable, given that, the experimental uncertainty in olfactometry can be as much as 50% (CEN EN 13725, 2003); thus, it is possible to take an averaged value of diffusivity for the application of the model to odour, this value being $9.65*10^{-6}$ m^2/s.

5. CONCLUSIONS AND FUTURE PERSPECTIVES

The need of a method to define the Specific Odour Emission Rate (SOER), after odour sampling with a WT and recalculate the SOER at the different wind speeds in the open field, implies knowing the models most suitable for describing the volatilization phenomena inside the WT and in the open field. This is why in the present work it was necessary in the beginning to analyse thoroughly the different models describing such processes. For the open field scenario, it was chosen as reference the model proposed in the textbook "Methods for the calculation of physical effects" (Committee for the prevention of disasters, 1992). Conversely, for describing the phenomenon inside the hood, as there are no models specific for the particular configuration of the Wind Tunnel, two models have been considered. The two models are the

321 ones describing the situations closer to the WT configuration. Eventually, the model for mass transfer

322 under forced convection from a single flat emitting surface in fully developed laminar motion regime and

323 developing mass transfer boundary layer, was preferred as the most appropriate to describe the real

324 situation. The decision was reached as a consequence of theoretical considerations. In order to verify the

325 applicability of the chosen model to the WT case, experimental tests were carried out with pure liquid

326 acetone and butanone. The outcomes confirmed that the mass transfer model for a single flat plate

327 forecasts a dependency of the emission on the sweep velocity equal to the dependency obtained

328 experimentally. However, the experimental concentration values are clearly lower than the model

329 prevision. Therefore, it was proposed to modify the model equation, varying the proportionality coefficient,

330 defining a value specific for the peculiar case of the WT.

331 Additional studies are necessary and should be carried out testing different compounds, GPC as well as LPC,

332 in order to improve the estimation of the suitable generalized experimental coefficient reported in Eq. (19),

333 also investigating the effects of the inlet zone on the phenomenon.

334 Furthermore, a correlation was defined that from the flush air speed in the WT, allows to assess the wind

335 velocity in open field at 10 m that would cause the same emission as the one calculated with the data

336 obtained in the WT. The correlation has the great advantage to account for the differences existing

337 between volatilization in the open field and volatilization in the WT. The formula returns a wind velocity

338 value that can then be used to recalculate the obtained SOER/SER to the wind velocities of interest: e.g., in

339 the case of an odour impact assessment with atmospheric dispersion modelling, these are the wind

340 velocities of the whole time domain of the simulation. Throughout the present work, it was taken great

341 care in the definition of the field of application of the presented models and correlations. Relying of

342 previous studies (Fingas, 1998; 2013), it is possible to say that the volatilization of low-volatility

343 compounds, is not influenced by the velocity of the wind sweeping the liquid surface. Moreover, the

344 researches by Hudson and Ayoko (2008) and Parker et al. (2010), led to concluding that for diluted

345 solutions, only for the case of compounds with dimensionless Henry's constant $< 10^{-3}$ the volatilization

346 process is controlled exclusively by the wind velocity on the surface. As a consequence of these findings, a

347 possible future development of this project would be to incorporate the models here described into a more

348   general model framework, capable of describing the volatilization of compounds that are not only

349   controlled by forced convection. In addition, it would be necessary to extend the validation of the proposed

350   models, performing other experiments with compounds different from acetone and butanone, both pure

351   and in aqueous solution. Furthermore, it would be useful to define a methodology to better compare the

352   experimental data with the forecasts of the model. This may entail carrying out chemical analysis as well as

353   olfactometric analysis. The latter has the additional criticality of performing the olfactometric analysis

354   directly in the field in ambient air, a difficult task. Finally, it would be interesting to investigate the adoption

355   of the friction velocity instead of simply the wind velocity in the model, considering the effects that waves

356   on the liquid surface may have on the friction velocity and the emission rate itself, effects that some recent

357   researches seem to indicate are actually not negligible (Prata Jr. et al., 2016b).

358

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477   **Figures Captions**

478   Figure 1. The PoliMi LabOlf Wind Tunnel.

479   Figure 2. The Wind Tunnel scheme.

480   Figure 3. Comparison model-experimental data for acetone.

481   Figure 4. Comparison model-experimental data for butanone.

482   Figure 5: SER recalculation method for acetone (blue line) vs SER recalculation method for butanone

483   (purple line) vs traditional SER recalculation method (red line) as a function of the wind speed (Fig. 5a).

484   Zoom for the 0-0.2 wind speed U,10 range (Fig. 5b).

485   Figure 6. SER recalculation for several odorous compounds.

486   **Tables Captions**

487   Table 1. Experimental data for acetone.

488   Table 2. Experimental data for butanone.

489   Table 3. Parameters values chosen for the comparison among the considered models.

490   Table 4. Values for the diffusivities and non-dimensional Henry's coefficients for the considered odorous

491   compounds.