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New developments and applications in modelling occupational exposure to airborne contaminants.

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Abstract

Occupational exposure assessment is an important stage in the management of chemical exposures. Few direct measurements are carried out in workplaces, and exposures are often estimated based on expert judgements. There is therefore a major requirement for simple transparent tools to help occupational health specialists to define exposure levels.

The aim of the present research is to develop and improve modelling tools in order to predict exposure levels.

In a first step a survey was made among professionals to define their expectations about modelling tools (what types of results, models and potential observable parameters). It was found that models are rarely used in Switzerland and that exposures are mainly estimated from past experiences of the expert. Moreover chemical emissions and their dispersion near the source have also been considered as key parameters.

Experimental and modelling studies were also performed in some specific cases in order to test the flexibility and drawbacks of existing tools. In particular, models were applied to assess professional exposure to CO for different situations and compared with the exposure levels found in the literature for similar situations. Further, exposure to waterproofing sprays was studied as part of an epidemiological study on a Swiss cohort. In this case, some laboratory investigation have been undertaken to characterize the waterproofing overspray emission rate. A classical two-zone model was used to assess the aerosol dispersion in the near and far field during spraying.

Experiments were also carried out to better understand the processes of emission and dispersion for tracer compounds, focusing on the characterization of near field exposure. An

experimental set-up has been developed to perform simultaneous measurements through direct reading instruments in several points. It was mainly found that from a statistical point of view, the compartmental theory makes sense but the attribution to a given compartment could not be done by simple geometric consideration.

In a further step the experimental data were completed by observations made in about 100 different workplaces, including exposure measurements and observation of predefined determinants. The various data obtained have been used to improve an existing two-compartment exposure model. A tool was developed to include specific determinants in the choice of the compartment, thus largely improving the reliability of the predictions.

All these investigations helped improving our understanding of modelling tools and identify their limitations. The integration of more accessible determinants, which are in accordance with experts needs, may indeed enhance model application for field practice. Moreover, while increasing the quality of modelling tool, this research will not only encourage their systematic use, but might also improve the conditions in which the expert judgments take place, and therefore the workers ‘ health protection.

Résumé

L’évaluation de l’exposition aux nuisances professionnelles représente une étape importante dans l’analyse de poste de travail. Les mesures directes sont rarement utilisées sur les lieux même du travail et l’exposition est souvent estimée sur base de jugements d’experts. Il y a donc un besoin important de développer des outils simples et transparents, qui puissent aider les spécialistes en hygiène industrielle dans leur prise de décision quant aux niveaux d’exposition. L’objectif de cette recherche est de développer et d’améliorer les outils de modélisation destinés à prévoir l’exposition.

Dans un premier temps, une enquête a été entreprise en Suisse parmi les hygiénistes du travail afin d’identifier les besoins (types des résultats, de modèles et de paramètres observables potentiels). Il a été constaté que les modèles d’exposition ne sont guère employés dans la pratique en Suisse, l’exposition étant principalement estimée sur la base de l’expérience de l'expert. De plus, l’émissions de polluants ainsi que leur dispersion autour de la source ont été considérés comme des paramètres fondamentaux.

Pour tester la flexibilité et la précision des modèles d’exposition classiques, des expériences de modélisations ont été effectuées dans des situations concrètes. En particulier, des modèles prédictifs ont été utilisés pour évaluer l'exposition professionnelle au monoxyde de carbone et la comparer aux niveaux d'exposition répertoriés dans la littérature pour des situations similaires. De même, l’exposition aux sprays imperméabilisants a été appréciée dans le contexte d'une étude épidémiologique sur une cohorte suisse. Dans ce cas, certaines expériences ont été entreprises pour caractériser le taux d'émission des sprays

imperméabilisants. Ensuite un modèle classique à deux-zone a été employé pour évaluer la dispersion d'aérosol dans le champ proche et lointain pendant l’activité de sprayage.

D’autres expériences ont également été effectuées pour acquérir une meilleure compréhension des processus d’émission et de dispersion d’un traceur, en se concentrant sur la caractérisation de l'exposition du champ proche. Un design expérimental a été développé pour effectuer des mesures simultanées dans plusieurs points d’une cabine d’exposition, par des instruments à lecture directe. Il a été constaté que d'un point de vue statistique, la théorie basée sur les compartiments est sensée, bien que l'attribution à un compartiment donné ne pourrait pas se faire sur la base des simples considérations géométriques.

Dans une étape suivante, des données expérimentales ont été collectées sur la base des observations faites dans environ 100 lieux de travail différents: des informations sur les déterminants observés ont été associées aux mesures d'exposition. Ces différentes données ont été employées pour améliorer le modèle d'exposition à deux zones. Un outil a donc été développé pour inclure des déterminants spécifiques dans le choix du compartiment, renforçant ainsi la fiabilité des prévisions.

Toutes ces investigations ont servi à améliorer notre compréhension des outils de modélisations ainsi que leurs limitations. L'intégration de déterminants mieux adaptés aux besoins des experts devrait les inciter à employer cet outil dans leur pratique. D'ailleurs, en augmentant la qualité des outils de modélisations, cette recherche permettra non seulement d’encourager leur utilisation systématique, mais elle pourra également améliorer l’évaluation de l’exposition basée sur les jugements d’experts et, par conséquent, la protection de la santé des travailleurs.

List of publications

Peer review

Bruzzi,R., Vernez,D., Droz,P.O., and De Batz,A., 2006. Beliefs and practices in the assessment of workplace pollutants. Sozial- und Praventivmedizin, 51, 5-13.

Vernez,D., Bruzzi,R., Kupferschmidt,H., De Batz,A., Droz,P.O., and Lazor,R., 2005. Acute respiratory syndrome after inhalation of waterproofing sprays: A posteriori exposure-response assessment in 102 cases. Journal of Occupational and Environmental Hygiene. 12, 250-261.

Bruzzi,R., Vernez,D., Sottas,P.E., and Droz,P.O., 2005. Exposure models in Switzerland. An overview of the present situation. Gefahrstoffe Reinhaltung Der Luft, 415-418.

Bruzzi,R., Girault,M., Vernez, D., Maillet, D., Sottas, P.E., Droz, P.O., 2007. Pertinence of a two-zone model for occupational exposure assessment. Journal of Occupational and Environmental Hygiene (Submitted).

Proceedings

Bruzzi,R., 2006. Modèle d'exposition au CO. Archives des maladies professionnelles et de l'environnement, vol. 67, pag.78.

Bruzzi,R., Vernez,D., Droz,P.O., and De Batz,A., 2006. Exposure models: current practices and ongoing development. In: Renewing a century of commitment to a healthy, safe and productive working life: 28 th International Congress on Occupational Health, Milan, Italy, June 11-16, 2006: book of abstracts. - Milan : ICOH (Scientific Committee on Education and Training in Occupational Health, 2006. - P. 190).

Outline

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Introduction

Context and Motivation

Estimating exposure is an important step in occupational health studies, both retrospective, and prospective. Preventive measures and corrective actions against pollutant exposure in the workplace are frequently based on these estimates. It may also play a key role in the recognition of occupational diseases. Exposure estimates to chronic pollutants is the traditional field of occupation hygienists and, to a lesser extent, of occupational physicians and occupational safety specialists (e.g. safety engineers).

For chemical exposure, *direct measurement* is certainly the most appropriate and objective way to obtain a reliable assessment of the exposure. It must however be emphasized that this approach suffers major drawbacks regarding cost and technical complexity. Furthermore, direct measurements only give information on the current exposure (the day of investigation) and do not allow for past exposure estimation, or exposures under other or future conditions (Nicas, 2003a).

The most simple and most widely used theoretical approach is probably the so-called *expert judgment,* the “art” of occupational hygiene. Occupational hygienists evaluate whether a potential hazard exists by observing workplace conditions and interviewing the exposed workers about the materials used, the production levels, the duration of exposure, existing preventive measures and so on. Exposure assessment is thus based on an interpretation of observations and interviews, integrated with knowledge gathered from previous similar situations, either coming from the specialist’s own experience or from literature reports.

Despite its widespread use, there is limited information on the 'expert judgement' approach (Ramachandran et al., 2003). These subjective estimates are usually unstructured opinions, difficult to explain objectively and to transfer to others (Jayjock, 1997).

However the quality of expert estimation and the variability among the experts have been explored on numerous occasions, mainly in epidemiological studies (Kromhout et al., 1987;de Cock et al., 1996;Walker et al., 2001;Benke et al., 2001;Walker et al., 2003;Ramachandran et al., 2003;Mannetje et al., 2003). In some cases, judgments of various kinds of professionals (hygienists, chemists, operators, supervisors) have been compared with each other, and also with quantitative measurements. In these studies, it has been shown that it is often difficult to make predictions, hygienists being however better than the other professionals.

In the absence of current monitoring data, *semi-quantitative methods* have been developed to estimate historical or future exposures. For example, a Job Exposure Matrix could represent a practical and less time-consuming method, using historical data through a cross classification of job titles by substances (Dosemeci et al., 1990). This approach is however limited in its details and cannot give information on specific exposure situations. Cherrie and Schneider (Cherrie and Schneider, 1999) have developed and validated a structured approach to assess exposure based on descriptive information about work activity and work environment. In this case judgments are not made on exposures themselves, but on certain parameters considered as critical, such as the intrinsic emission, the work method and the prevention techniques used.

The Estimation and Assessment of Substance Exposure (EASE) is a *semi-quantitative empirical model,* developed in England to better describe workplaces with available historical data (Cherrie et al., 2003;Creely et al., 2005;Cherrie and Hughson, 2005). It gives ranges of potential exposures based on an analysis of exposure measurements contained in the UK National Exposure Database (NEDB). In fact, a selection of exposure determinants is included in the model, and their influence is estimated based on past exposure measurements. This allows the user to make predictions using a simple description of workplaces and processes.

An example of a *Bayesian framework* was developed by Sottas et al. (Sottas et al., 2005). In this case, three different sources of available data were combined: (1) information on the exposure determinants is taken into account in a parametric physical model; (2) a nonparametric, empirical model takes advantage of retrospectively collected exposure data; (3) direct measurements are used for sampling airborne contaminants in the workplace. In practice, exposure determinants allow the construction of two concentration distributions from physical modeling and historical measurement data. Bayes' rule is employed to combine this prior knowledge with field measurements, to get a posteriori probability estimate of the exposure. An example of application of *Bayesian methods* to occupational exposure assessment is presented by Ramachandranrt al. (Ramachandran G, 1999). Based on limited historical measurements and subjective expert judgment, they presented a framework to reconstruct probability distributions of historical exposure for various groups of workers to airborne particulates. Similarly, Wild et al. (WILD et al., 2002) recently described an original method for combining expert judgment on exposure in each "exposure group" with exposure measurements. Finally, Hewett provided (Hewett et al., 2006) a Bayesian decision methods to improve professional judgment.

Deterministic models have been developed for a quantitative reconstruction of historical exposures, but may also be used for complementary or prospective exposure assessments in the future. Rong (Rong et al., 1990) takes into account the causal variables actually responsible for changes in exposure and the interdependence between mean exposures in consecutive time periods. Kauppinen (Kauppinen et al., 1994) included in the model variables related to the job, the emission of chemicals, the contact with chemicals, and other relevant determinants of exposure. The results were then more valid and reliable than the subjective assessments.

Available *physical* models, based on physico-chemical principles, such as ventilation characteristics, pollutant generation rate, and mass transport mechanisms, provide a convenient way to structure significant factors determining the levels of exposure.

Physical models can range in complexity from the very simple zero-ventilation model and multi-compartmental models to computational fluid dynamic models. The precision requirement of a model depends on how close to the “truth” the model output needs to be to make a decision. More sophisticated models are able to take into account more details, such as spatial and temporal pollutant concentrations. However, in ranking several scenarios, for example, models do not need to be too detailed. Keil (Keil, 2000b;Keil and Murphy, 2006) described a tiered approach in selecting which model to use, “considering the goal of the modeling, the availability of model inputs, and the degree of uncertainty that is acceptable”.

The notable gains of employing models are their general simplicity, quickness and their low-cost. They also may provide a screening tool in the field of chemical risk assessment. As well, models can also serve as a useful tool for predicting exposures to new substances where no direct data are available, or to provide specific ventilation requirements under different assumptions for production rate, chemical consumption or air mixing conditions (Olcerst, 1999). Buringh and Lanting (Buringh and Lanting, 1991) list a number of advantages in using models: “(1) reduction of monitoring effort; (2) insight into how different workplace variables affect outcomes, and (3) the ability to predict the effect of various control options. So model application can help hygienists to understand how exposure depends upon various parameters such as ventilation rates or emission rates”.

As a matter of fact, due to its reduced costs, there is a general increase of the expert judgment practice. So exposure assessment is often based on the knowledge acquired in previous similar situations, either coming from the specialist own experience or reported in the literature. However situations for which the specialist's own experience is not sufficient or situations, for which identical exposures are not reported in the literature, often occur.

In such cases, the problem is to identify the relevant factors – exposure determinants – that have an influence on the exposure level. Exposure determinants have been studied on many occasions, mainly in the context of epidemiological studies, but also when it is necessary to identify key parameters on which to act to reduce exposure.

Identification and quantification of determinants are based either on experimental studies or on observation of actual workplace conditions. Thus identified determinants are usually included in empirical relationships or structured in exposure model. However generalizations of this practice are relatively rare, which makes it difficult for hygienists to apply this exercise to new situations. There is therefore a need for a generalized approach to the quantification of exposure determinants, but mostly a need for a deep understanding of their impact on exposure.

In this context, the integration of relevant exposure determinants in appropriate model structures may represent a more systematic, transparent and consistent approach to exposure prediction. “Mathematical modelling can therefore be regarded as a formalization of the decision-making processes” (Karplus, 1983).

Hygienists generally prefer measurements to mathematical model, because of their relative accuracy. However in certain cases measurements may have a high degree of variability. If high quality information on exposure determinants is available and few measurements are obtainable, models may provide a more accurate estimate than sampling (Nicas and Jayjock, 2002).

Jayjock (Jayjock, 2005) believes that “exposure modeling represents the essence of the science of exposure assessment and should be considered a principal stock in trade of all industrial hygienists”.

Thus, between the different approaches previously presented, physical models have to be considered as attractive tools for gathering information on exposure levels in a decision making process.

Based on a literature review conducted in the field of exposure assessment methods, the following hypothesis may be expressed:

- occupational exposure assessment to pollutants rely more and more frequently on "expert judgments"
- expert judgments are funded, explicitly or not, on the identification and assessment of exposure factors perceived as relevant (determinants)
- emission conditions and near-field dilution are key factors of the exposure process

More general models would be very useful in exposure assessment and efforts should be done to develop and validate them. There is a strong suspicion that these assumptions are true, however these topics must be confirmed during the research project. In any case, an implementation of the existing models with parameters in accordance with the experts needs may not only encourage their systematic use but may also increase the quality of the expert judgements in regards of their reproducibility, of their accuracy and of the coherence between experts.

Objectives

This project aims to improve the conditions in which the expert judgments take place, by providing adequate tools for the practitioners. The implementation of the existing tools with parameters in accordance with the experts needs is indeed expected to promote their systematic use. Specific goal may be distinguished:

1. Investigate the exposure assessment methods and assess the key factors observed during expert judgement through literature review and questionnaires.
2. Assess the use of exposure models in terms of frequency and perceived accuracy as well as analyse the needs and the difficulty of the practitioners in using models.
3. Understand model limitations and benefits through laboratory experimentations.
4. Develop an exposure model, through a revision of the existing tools, based on observable parameters and easy accessible information, in accordance with the experts needs

This project doesn’t intend to replace measurement by expert judgement in any situation. It rather aims to allow the practitioners to treat efficiently, quickly and cost-effectively an enlarged number of workplaces exposure situations. No measurement is usually required for situations, which are clearly beyond or below the acceptance criteria.

Literature review.

Several exposure models may be used in order to assess quickly occupational exposure to a given pollutant. Among physical models, two separate steps to assess pollutant concentration can be distinguished: estimating the pollutant emission and predicting the pollutant dispersion.

Emission quantification

A systematic qualitative and quantitative evaluation of the pollutant release is a priority in fully describing a workplace exposure situation. More, emission rates are indeed required to implement deterministic exposure models and their accuracy is known to affect strongly the overall assessment performance.

There are several ways to assess emission data; these include reference to literature (i. e. existing emission factors), practical approach (such as mass balance or field measurements or tracer gas methods) and empirical expressions (or specific emission model).

Emission factors, describing the amount of pollutant released per unit quantity of time can be very easily integrated. Agencies, such as the US Environmental Protection Agency, or the Swiss OFEFP have established global emission factors for a large number of industries. Different ways to assess emission factor have been proposed (Wadden et al., 1991;Wadden et al., 1998;Nagaraj and Sattler, 2005;Kura et al., 2006;Heung et al., 2007). However, as each workplace differs in the way the production facility is operated, the chemical composition of the used mixtures, the settings of the workplace, such as volume and ventilation rates and even differences in worker behaviour, emission factor applicability has to be systematically evaluated.

A mass balance of the working process may be used to set an average emission factor. This practical approach is based on the law of conservation of mass. For gases and vapours, the balance calculation is quite easy: the overall amount released is determined observing the difference in weight over a specified time. The situation is more problematic when the pollutant is a by-product or when the emitted amount is small. Contrarily to using existing emission factor, using field measurements has the advantage of providing the “true” emission factors specific to a given activity and exposure situation.

An alternative method for mass balance consists in monitoring the pollutant profile concentration and ventilation conditions, and combining this data in a classical dispersion model. The emission rate corresponding to the pollutant level may then be set through a backward calculation. Several example of this methodology are found in literature (Selway et al., 1980;Franke and Wadden, 1987;Wadden et al., 1995;Conroy et al., 1995;Keil et al., 2001;Raisanen et al., 2001;Lacey et al., 2006).

However, the uncertainty can be large due to the simplifying hypothesis inherent in the models on the dispersion pattern, such as the assumptions of completely mixed conditions or ideal dispersion gradients from the source.

Lastly, another approach for a mass balance is to measure, at steady state conditions, the air flow and pollutant concentration levels at each exit point of the room (Keil et al., 1997;Wadden, 2001).

Nevertheless in practice, steady state assumption is not always satisfied and exit points can not be easily identified; in these cases, sources might be placed in an experimental chamber, and measurements may be performed in a controlled setting (Tan and Flynn, 2002;Säämänen and Skrifvars, 2002).

However, deviations may exist between emissions estimated in a controlled environment versus “real” environments with different ventilation systems.

One should take notice that an assessment of emission rates based on existing emission factors or mass balance methods only provide an average emission value, and thus this approach is unable to account for dynamic emission rate.

A tracer gas method may also be used for emission source assessment. The principle consists of generating a tracer gas at a known steady emission rate, close to the pollutant emission source. The simultaneous measurement of both pollutant and tracer gas at the same point of the room allows an estimate of pollutant emission. Assuming similar dispersion patterns for both species, the pollutant mass flow may be easily deduced from tracer gas behavior. Tracer gas have been used extensively in the field of industrial hygiene to assess ventilation patterns (i.e. to determine air exchange rates or efficiency of local exhaust ventilation systems) (Shaw, 1993;He et al., 2005b;Batterman et al., 2006). However only a few applications in emission source assessment are reported in the literature (BEMER et al., 1999;BEMER et al., 2002).

The characterization of the emission source through emission models is thoroughly described in the literature (Heinsohn, 1991;Fehrenbacher and Hummel, 1996;Keil, 2000a;Guo, 2002), although its practical application is usually limited. The large number of possible mixtures and materials, which may generate pollutants, as well as the various emission conditions, (operation modes, activity) may explain this insufficiency. Actually, emission models tend to be specific and their pertinence will depend on the emission process involved as well as on the workplace situation. Moreover, their applications require in depth information on the physical and chemical properties of the chemical of interest along with characteristics on the environmental setting.

For instance, evaporation phenomena are depicted in several models. The generation rate is often expressed as a function of the exchange surface, air velocity and molecular weight (Keil, 2000a). Parameters such as diffusivity, kinematics viscosity, surface length and ambient pressure are also taken into account in some of the existing models (Jayjock, 1994;Mulhausen and Damiano, 1998). Two models are of particular interest with regards to evaporation: the exponentially decreasing model and the backpressure model. The exponentially decreasing model (Keil and Nicas, 2003) is applicable when the emission concentration is far below the solvent's saturation concentration (such as in the case of volatile organic compounds). In the backpressure model the partial vapour pressure of the substance in the room (near the evaporation zone) affects the emission rate. Lennert et al. (Lennert et al., 1997) tested the performance of 6 different evaporation models suggested for occupational hygiene.

Fehrenbacher et al. (Fehrenbacher and Hummel, 1996) presented an evaporation rate model for various activities, such as open surface tank and drumming operations.

Other models, considering particle separation mechanisms, were developed and validated to predict the amount and size distribution of dust generated by different material handling operations (Plinke et al., 1994;Lanning et al., 1995).

Dispersion model

When applying a deterministic model in exposure assessments, it is of critical importance to understand the basis of the models, their strengths and weaknesses, in order to select the appropriate values for the model parameters. A thorough understanding of the influence of the exposure determinants on the outcome is a crucial point. Therefore, it is important to record, during air monitoring, other details along with the main measured value, such as the size of the workroom and the general ventilation rate, which have demonstrated to impact directly the average pollutant concentration (Cherrie and Schneider, 1999).

Contaminant dispersion phenomena within the rooms can be influenced by complex interactions between variables as the room geometry, the direction of principal air flow, the temperature gradient, the presence of a worker and even by the movements of his arms. Several authors have considered these influences in exposure assessments (Brohus et al., 1996;Welling et al., 2000;Guffey et al., 2001;Wu and Gebremedhin, 2001;Whicker et al., 2002;Lee et al., 2006;Chang, 2006). Experiments have been carried out to understand the influence of the worker's and contaminant source's position with respect to the flow direction in determining breathing zone concentrations (Kim and Flynn, 1991;Flynn and Ljungqvist, 1995;Flynn et al., 1999;Ojima, 2005;He et al., 2005a).

The parameters in the current models, as those described in this context, do not take into account such kind of detailed information. Nevertheless, it is important to report them case-by-case and to interpret the results considering the specific circumstances by taking into account the simplifications made.

Zero-ventilation model

In assessing chemical health risk, an initial conservative exposure estimation may be carried out through a 'worst-case' point estimation, taking into account only the input variables that will result in the highest output. Simple *saturation* or *zero- ventilation* models predict such worst-case scenarios by assuming no dilution within a space via general ventilation (Jayjock, 1997;Keil, 2000a).

The zero-ventilation model calculates the concentration that would occur if there is no ventilation, no sinks and all of the mass of the chemical being considered enters the air instantaneously. The air is considered completely saturated with vapour, based on the assumption that the liquid is allowed to evaporate for a long time and that sufficient liquid is present to allow the entire room to reach its equilibrium concentration. The resulting predicted concentration is assumed to be spatially uniform throughout the room and is calculated as:

$$ppm_A = \frac{P_{A\,vap}}{P_{atm}} * 10^6 \qquad [1]$$

where P_{Avap} is the saturation pressure [Pa] and P_{atm} the ambient atmospheric pressure [Pa]

The approximation that pollutants are uniformly distributed throughout the interior space is used routinely in indoor models, as in the box model hypothesis, and its pertinence will be discussed further on. The time to reach equilibrium can be very long, especially for a large space, and it is not always the case that sufficient liquid is present to permit saturation. A final assumption is that the room is completely enclosed. It is intuitive, that with the exclusion of all loss processes, the concentration estimated would be highly overestimated.

Models including ventilation

Ventilation rate is of considerable importance in occupational hygiene. The average pollutant concentrations in a workplace are heavily influenced by the ventilation airflow patterns, as they are responsible for the transport and removal of the contaminant.

Studies investigate the influence of the change in ventilation rate on contaminant dispersion. Whicker et al. (Whicker et al., 2002) showed how lag times decrease if ventilation rate increased.

Defining pollutant transport is one of the main differences between the models presented below. They, actually, incorporate a wide range of assumptions regarding pollutant transport varying from an homogeneous instantaneous mixing (Ideal mixed model), to a series of smaller completely mixed “boxes” within a room (Two Zone Model, for two boxes), to diffusion models with continuous concentration gradients in time and space (Eddy Diffusion Model), to directional diffusion models reflecting the presence of advective flow in the room (Gaussian Plume Dispersion model) (Keil, 2000a).

One-box model

In the ideal mixed *one box model* concentrations in the workplace are calculated as a function of the emission and ventilation rates as well as the time elapsed from the start of the emission (Keil, 2000a) (Jayjock, 1997). This model relies on the concept of mass conservation and of a complete instantaneous mixing throughout a single workplace volume. According to the simplest version of this model, the volume of the workplace is modelled as one homogeneously mixed box. Generally in box models, the entire room volume is considered as the air volume available for exposure, and it is assumed to coincide with the total room volume. Moreover, if the room is not of a standard shape, a regular shape of solid geometry could be assumed to define the room. However, some authors demonstrated that room structure can slow down the mixing by creating local eddies or air pockets (Whicker et al., 2002).

The equation describing contaminant concentration in a box is developed using a mass balance equation:

$$V \cdot \frac{dC_A}{dt} = Q \cdot C_{Ain} + G - Q \cdot C_A - K_{sink} \cdot C_A \qquad [2]$$

where C_A is the uniform room concentration of A [mg/m^3], C_{Ain} the concentration of A in the incoming air [mg/m^3], Q the ventilation flow [m^3 /s], V the compartment volume [m^3], G the emission rate [mg/s],

K_{sink} the rate constant for the pollutant sink process [m^3/s].

To integrate this equation, various assumptions must be made. The rate constant K_{sink} is a property of the room that depends of the affinity between the contaminants and the surfaces of the room. It is commonplace, to assume that sink effects are negligible for occupational settings. The mass generation is assumed to be constant with time and the initial concentration and that of the incoming air are set to zero. The following equation gives a fair first estimate of average room air concentration changes with time:

$$C_A = \frac{G}{Q} \cdot \left[1 - e^{\frac{Q(t-t_0)}{V}} \right] \qquad [3]$$

At steady state (the condition in which the physical proprieties of a system do not change with time), equation [2] is reduced to:

$$\frac{dC_A}{dt} = 0 \longrightarrow C_A = \frac{G}{Q} \qquad [4]$$

This model does not provide information about the spatial dispersion of air contaminants but may nevertheless represent a practical approach in some particular exposure situations. These include complete mixing within the space being modelled, homogeneous source emissions throughout the space and long time-scale modelling. If these criteria are met, the one-box approach can often give quantitatively acceptable results (Jayjock, 1988).

For instance, in some ventilation configurations, a complete mixing may be achieved by supplying air with a high momentum outside the occupied zone or in the case of a multi-source emission homogeneously distributed throughout the space (Jayjock, 1988) (Qian et al., 2006). However airflow patterns have a strong impact on the pollutant distribution, and it was shown that the most important aspect in the contaminant removal efficiency is the relative position of the area source to the main airflow pattern and the occupied zone. (He et al., 2005a)

Thus, a deep understanding of ventilation system is recommended before applying this kind of model (Taylor et al., 2004).

Correction factors for incomplete mixing (one-box model)

Although the ideal mixture hypothesis may be useful for estimating exposure in some conditions, it is an inappropriate approximation for exposures in large workplaces (Finlayson et al., 2004). Complete mixing may not occur in large rooms, as seen in cases with volumes greater than 500m^3 (Gmehling et al., 1989). A common occurrence in an indoor situation is the short-term airborne release of a small, localized contaminant source, in a large workplace. In this case, if workers are close to the source, exposure to high pollutant concentrations can occur immediately after the release, before the contaminant is spread and dispersed in all of the volume concerned (Drivas, 1996). The well-mixed assumption is also a poor approximation for situations with a long-term continuous release in which the space never achieves a fully mixed state (Finlayson et al., 2004). Thus the mixing problem has two aspects: when one may safely apply the well-mixed assumption, and how to model pollutant concentrations when the well-mixed approximation is inadequate (Gadgil et al., 2003).

Experimental studies verify the hypothesis that mixing time (defining the earliest point after which the room concentration is essentially uniform - the relative standard deviation of concentrations equal to 10% or less) is correlated with mechanical power and provide a quantitative relationship between the mixing rate and the intensity of input energy (Baughman et al., 1994;Drescher et al., 1995). Other results suggest that people moving about in a room can induce rapid mixing (Mora et al., 2003), to the extent that the well mixing approximation may be well justified for cases with strong internal air motion. However, in the case when air movements are weak, this approximation may not be suitable and exposures will depend considerably on the spatial relationship between emission and receptor.

To account for spatial variations in concentration a *mixing factor* “m” is often introduced, as a coefficient by which the actual ventilation is multiplied to obtain a lower “effective” ventilation rate. The effective ventilation is traditionally determined from the slope of the log of concentration against time for the decay of concentration (Wadden et al., 1995;Taylor et al., 2004).

Ishizu (Ishizu, 1980) examined experimentally the introduction of a "mixing factor" on modelling imperfectly-mixed rooms. Repace and Lowery (Repace and Lowrey, 1980) proposed different mixing factors for various enclosed spaces. Other authors (Matthiessen, 1986) (Jayjock, 1988) also recommend the use of a mixing factor to account for non-homogeneous situations. Feigley et al. (Feigley et al., 2002b) employed computational fluid dynamic simulation to explore the effect of various contaminant sources, air inlet and air outlet location on mixing factor.

However, there are no theoretical or empirical selection criteria available, and uncertainty or variability associated with “m” is not well established (Keil, 2000b). More, as mixing factors can neither be predicted with precision nor be generalized, some experts discourage their use (Heinsohn, 1991).

Multi-zone models

An alternative to using mixing factors to account for the less than complete mixing in a closed space, and the higher intensity of exposure near the source, is the use of a series of conceptual well-mixed compartments to represent several mixing zones within a room. Between each compartment contaminant is transferred via a volumetric flow rate across the boundaries of the zones (Ozkaynak et al., 1982;Nazaroff and Cass, 1989). The concept can be extended to as many cells or zones as the users deem necessary, however, quantifying the value of the exchange rate for each cell becomes critical to the accuracy of the solution.

However some authors proposed different ways to evaluate the exchange rate: the Computational Fluid Dynamic (CFD) techniques, experimental measurements of velocity field and professional intuition (Haberlin and Heinsohn, 1993). Perrier et al. (Perrier et al., 2005) provided an example of a four-box model.

A more simplified workplace description is found in the *Two Zone Model* (Nicas, 1996;Nicas, 2003b;Nicas et al., 2006), which divides the room into two conceptual zones. The first near the source (near field) contains the worker's breathing zone and the second represents the rest of the room (far field). The inner-box airflow rates move simultaneously into and out of the near field, while the general ventilation moves into and out of the far field. It's also assumed that the contaminant emission rate is constant and that there are no sink terms. This description leads to the following coupled mass balance equations:

$$V_{NF} \cdot dC_{NF} = G \cdot dt + \beta \cdot C_{FF} \cdot dt - \beta \cdot C_{NF} \cdot dt$$

$$\mathrm{V_{FF} \cdot dC_{FF} = \beta \cdot C_{NF} \cdot dt - (\beta + Q) \cdot C_{FF} \cdot dt}$$

where:

$C_{FF,NF}$ are the uniform concentrations of the near and far field [mg/m^3], Q is the ventilation flow [m^3 /s], $V_{FF,NF}$ are the near and far field volumes [m^3], G is the pollutant emission rate [mg/s],

β is the airflow rate between near and far field [m^3 /s].

At steady state equations reduce to the simple form of:

$$\mathrm{C_{NF}} = \frac{G}{Q} + \frac{G}{\beta} \qquad \mathrm{C_{FF}} = \frac{G}{Q}$$

Calculation of dynamic concentrations may be found in literature (Nicas, 1996;Keil, 2000a).

The advantage of the two-zone model is that it is a first step in addressing spatial variability of concentration and its use is recommended in assessing the exposure intensity of a worker close to the source. Different studies (Rodes et al., 1991;Ohmichi et al., 2006), undertaken to compare personal sampling with general sampling, have shown that personal exposures are generally higher than general exposures. They found that typical ratios of PEM/MEM (personal monitors /microenvironment monitors) ranged from 1.58 to 13.4. This effect is also demonstrated by Furtaw et al. (Furtaw et al., 1996) who employed a two-zone model, termed a source-proximate effect (SPE) model, to fit data from measured concentrations at various distances from the source. Other experiments (Flynn and Ljungqvist, 1995) were carried out to establish the influence of worker’s presence on the contaminant dispersion in the near field.

These studies demonstrated that a reverse flow zone, produced in front of a worker, might cause high contaminant concentrations in the breathing zone. Moreover, arm movements influenced contaminant dispersion (Welling et al., 2000). Still, Cherrie (Cherrie, 2003) reviewed data about personal and area concentrations for 40 different working situations and found that 80% of the personal measurements exceeded the respective environmental measurements.

Thus, this model, simplifying spatial variability of concentration into just two compartments, may represent a useful tool in the occupational hygiene practice, which tends indeed to focus exposure assessment on two kinds of situations, individual and ambient exposures (other workers within the same room).

However, a current drawback of this model, compared to the ideal mixed model, is the need to develop criteria for defining additional parameters such the size and shape of the near zone and the air exchange rates between the two zones.

In particular the determination of the inner air exchange rate, β is poorly understood. This parameter depends on the conceptual near field geometry and the random airspeed near the source. However Cherrie (Cherrie, 1999), reported three values for this parameter: 3 m^3/min for minimal convective air flow, an intermediate value 10 m^3/min, and 30 m^3/min for maximal convective air flow. A lower air exchange rate would result in a higher concentration.

Some authors propose defining the near field volume as a hemisphere with a radius equal to the distance between the source and the human receptor (Keil, 2000a), or still equal to 1 m (Spencer and Plisko, 2007). In this case, the inter-zone airflow can be defined as the product of one-half of the free surface area enclosing the near zone times the random air velocity on this surface. The reason of applying only one-half of the surface is to maintain the mass balance of air in the near field volume. This way it is assumed that the air flows in through only half the surface and flows out through the other half. The airspeed through this surface depends on worker movement and turbulence within the work processes. By contrast, (Cherrie, 1999) assumed a fully turbulent convective air flow arising from the person's body heat to describe the inner-zone air exchange. Selection criteria or estimation methods for air speeds, to apply to various room conditions, are not readily available. However Baldwin and Maynard (Baldwin and Maynard, 1998) compared personal wind speed measurements with static wind speed measurements. The results showed that for wind speeds smaller than < 0.3 m/s, the distribution was similar in shape with a number of peaks of high speeds above 0.4 m/s from the personal anemometer, probably due to worker movement.

Another configuration of the compartments is suggested by Nicas (Nicas, 1996), who divided the room in an upper ventilated zone and a lower zone of occupancy In this case a particular ventilation scenario is taken into account, where both the supplied ventilation air and the room air exhaust are near ceiling level. Hemeon, quoted in Burton (Burton, 1999), discussed various geometries for the near field, depending on the particular work operation involved. An example is found in Nicas et al. (Nicas et al., 2006) who employed a compartment with a rectangular base of the same area as the wash basin used (the emission source), while the height coincided with the vertical distance between the wash basin and the breathing level of the worker.

Cherrie (Cherrie, 1999), used a two-compartment box model to simulate exposure concentrations for a wide range of general ventilation conditions and room sizes. The ratio of near- to far-field concentrations from the simulations ranged from unity in small poorly ventilated rooms, to 24 in large well ventilated areas. Some studies (von Grote et al., 2003;Nicas et al., 2006;Keil and Murphy, 2006;von Grote et al., 2006;Spencer and Plisko,

2007) observing a good agreement between concentration estimates and measured concentrations, proved an adequate degree of reliability in predicting exposure levels through modelling assessment.

Eddy diffusion model

The Eddy Diffusion Model has notable advantages over the previously described models as it can take into account the gradual decrease of concentration when moving away from the source (Roach, 1981). This model is based on the assumption that mass transport is driven by turbulent (or "eddy") diffusion, which is expected to dominate molecular diffusion. The eddy-diffusion model is appropriate for modelling near-field exposure from continuous emission sources in rooms without a unidirectional air draft. The assumption that no significant air velocities exist in any specific direction has been found valid near the core of a room in experiments carried out by Cooper and Horowitz (Cooper and Horowitz, 1986). Experiments by Zhang et al. (Zhang and Christianson, 1990) in a 1/4-scale test room show that air turbulence in the centre of a room is plausibly uniform for simple ventilation situations.

Concentrations are modelled both as a function of distance from the emission source and of time from the start of emission. Equation 15 represents the concentration equation for a constant emission flux G.

$$C_{t,r} = \frac{G}{4\pi Dr}[1 - \mathrm{erf}(\frac{r}{\sqrt{4Dt}})]$$

where $C_{t,r}$ is the pollutant concentration (g/m^3), G the emission flux (g/s), D the eddy diffusivity coefficient (m^2/s) (an empirical parameter), r the distance from the emission source (m), and t the time elapsed since the start of release (s). *Erfc* denotes the complementary error function. Erfc(x) is equal to zero when x = 0 and it is equal to unity when x=∞.

The steady state concentration is described by the following equation:

$$C_r = \frac{G}{4\pi Dr}$$

A key model parameter is the eddy diffusivity coefficient. This parameter describes bulk air movement caused by the motion of the room's occupants or by turbulence within emission phenomena. These eddies transport mass, so an increase of air speed associated with bulk movement will increase the amount of mass transported and reduce the spatial extent of the concentration gradients around the source. Some authors (Drivas, 1996;Fehrenbacher and Hummel, 1996;Guo, 2002) have proposed different approaches to calculate this parameter.

However, there is not much guidance available for selecting or estimating the diffusion parameter for a given air space. Keil (Keil, 2000a) has reported a number of experimentally determined values for indoor air spaces of different room dimensions and air changes per hour. Some eddy diffusion values are available in the literature (Wadden et al., 1989;Scheff et al., 1992). Measurements of diffusion coefficients in indoor industrial studies have ranged from 0.05 to 11.5 m^2/min, and displayed 0.2 m^2/min as being a typical value (Jayjock, 1997).

Another version was presented by Roach (Roach, 1981;Lennert et al., 1997) who integrated flow rate of fresh air being supplied and the concept that the stationary concentration in the air discharged at the periphery is equal to the equilibrium concentration of an ideally mixed model. Moreover, the room volume is taken into account in the R coefficient (the distance from source and the room wall). The steady state concentration, according to this model, is:

$$C_r = \frac{G}{Q} + \frac{G}{4\pi D}\left(\frac{1}{r} - \frac{1}{R}\right)$$

Gaussian plume dispersion model

The diffusion model is used for completely random dispersion, but it can also be modified to reflect the presence of advective flow in the room. Thus, the Gaussian Plume Dispersion model (Roach, 1981;Scheff et al., 1992;Lennert et al., 1997;Mulhausen and Damiano, 1998) is based on a diffusion model that takes into account the direction of air currents.

The equation for the steady state concentration becomes:

$$C = \frac{G}{4\pi Dr} e^{\left[\frac{-U}{2D}(r-x)\right]}$$

where, C is the pollutant concentration (mg/m^3), G the emission rate (mg/s), D the eddy diffusivity (m^2/s), r the distance of the worker from the source (m), x the downwind distance from the source along the centerline of the plume (m), and U the air velocity (m/s).

This is a simplification of the general air dispersion Gaussian plume model and assumes an unvarying wind direction and speed. It is also assumed that the eddy diffusivity (D) is the same in all directions and there is no plume rise. The advective diffusion model was applied in practice by Scheff et al. (Scheff et al., 1992) to translate area concentration measurements into emission rates from degreasers, where the advective air flow was found to influence the concentration pattern.

Computational fluid dynamic models (CFD)

CFD modelling is based on the solution of a non-linear set of equations for the conservation of mass, energy and momentum (Navier-Stokes equations). CFD models represent a powerful tool capable of predicting airflow patterns and pollutant concentration throughout a room over a finely spaced grid, once the appropriate boundary conditions (like pollutant generation rate, geometry of inlet and outlet ducts, ventilation volumetric flow rates throughout the room, thermal boundary conditions) are specified. However, the application of a CFD model requires specialized knowledge, experience and care in defining the grid, identifying and specifying appropriate boundary conditions, and selecting the numerical properties of the model. Moreover, the calculations are very time consuming and require large amounts of computer memory (due to restrictions on grid size) to enable adequate treatment of turbulence. Still, the post-processing analysis and visualization of the large volume of output demand considerable efforts to appreciate the output results. Thus, the complexities of CFD model applications, precludes their use for many problems of practical interest.

In academic context, however, CFD has been used as an alternative method to experimental measurements in the evaluation of physical models (Bennett et al., 2000;Bennett et al., 2003). Salim et al. (Salim et al., 2006) have demonstrated reasonable qualitative predictions, combining CFD simulations with different evaporation models.

CFD was often employed to understand the indoor airflow behavior and pollutant transport for different ventilation rates and configurations: a naturally ventilated multi-room building (Chang et al., 2006); a ventilated room containing a downdraft table (Jayaraman et al., 2006); for push-pull ventilation systems (Chern and Ma, 2007), in an enclosed space at different air flow rates (Lee et al., 2002) and for different location of source and exhaust opening (Feigley et al., 2002a). Therefore CFD application is becoming more and more widespread, but high quality comparison with field measure remains uncommon (Finlayson et al., 2004).

Research Plan

In a first phase a survey was undertaken among Swiss occupational hygienists and other professionals in order to identify the different exposure assessment methods used, the contextual parameters observed during expert judgements, and the uses, difficulties and possible developments of exposure models for field application. A questionnaire (**Annex I**) were developed for 122 occupational health professionals, members of the Swiss Occupational Hygiene Society, and for the other occupational health specialists (169 occupational physicians, 97 safety professional). Descriptive statistics and multivariate analyses were performed to analyze the results. The results were presented and discussed at Swiss Occupational Hygienists Society Erfa Tag (Bern, 2004) and at Experts' Workshop of the ISSA Health Services Section "Models and calculation methods to determine exposure to dangerous substances" (Dresden, Germany, 2004).
Results are presented in the **paper I.**

In a second step, some applications have been undertaken in order to identify difficulties with existing assessment tools, to test the flexibility and the accuracy of the traditional exposure models and to improve our understanding of modelling practice. Thus exposure assessment have been performed through experimental and modelling works, respectively for two typical exposure situations for which fields measurements were not possible: a retrospective assessment (for an epidemiological study) and a prospective analysis (for new situations or estimation of the effect of selected parameters).
The retrospective analysis has been undertaken in collaboration with the Swiss Toxicological Information Centre and the Swiss Registries for Interstitial and Orphan Lung Diseases. The

objective was to clarify the circumstances and possible causes of the observed health effects for individuals exposed to fluorinated polymers from waterproofing sprays.

We investigated through questionnaires (to 102 patients) exposure circumstances during spraying activity, such as the products involved, the duration of spraying activity and residence time after spraying, room dimensions and ventilation condition (open windows and doors). To investigate the possible relationship between exposure and health effects, perceived health effects regarding symptoms, time before occurrence, time before medical care, duration, were also asked. Still, the more objective clinical indicators (such as two non-specific markers of inflammatory response - the white blood cell count and the serum C-reactive protein – and the arterial partial oxygen pressure) were collected in a parallel physician's questionnaires.

Then, an experimental set-up has been developed to perform measurements of emission rate of several commercial sprays. We calculated over-spray emission rate from measurements of aerosol concentration and airflow in the exhaust of the chamber and from time video recording of emission phases. Collected data from questionnaire and experimental measurement were used to conduct numeric simulation. A classical two-zone model was used to assess the aerosol dispersion in the near and far field during spraying. Finally the assessed levels of aerosol exposure were compared to the exposure outcomes (health severity) in order to highlight possible dose-response relationships.

These results are presented in the **paper II.**

Compartmental and diffusion models have also been applied, in order to illustrate their usability to assess various CO professional exposures. Three different situations have been taken into account: two indoor exposures in a car garage and in a karting hall and an outdoor exposure to chainsaw exhausts. For each situation, different emission and ventilation scenarios were simulated and integrated in different exposure models, according to the situation. The profiles of concentration calculated with the models have then compared with the exposure levels found in the literature for similar situations. The results were presented at the Swiss Occupational Hygienists Society Erfa Tag (Lausanne, 2005), at the "Journée Franco-Suisse de Médecine du travail (Belfort, France 2005)" (Bruzzi, 2006).

Annex II, presents a poster on this study

Looking at research institutions, however, there is a big interest in the use of models to solve problems which are difficult to address with field measurements, in agreement with the current European and American trends (ISSA 2004). An overview of these research activities is provided in the **paper III**.

In accordance with the results obtained from the literature review and the questionnaire analysis, the second part of the research was focused on the characterization of near field exposure. To address these requirements, a simple theoretical model has been selected (a two-box model), and its hypotheses were investigated through theoretical aspects, experimental investigations, statistical analysis and computational fluid dynamic simulations. An experimental set-up was developed to perform simultaneous measurements by direct reading instruments in several points of an experimental room. Various semiconductor gas sensors have been tested and calibrated for a gas (methane 2.5 %) and an organic solvent vapor (ethanol). A constant emission was achieved with a peristaltic pump injecting ethanol on a hotplate causing instantaneous evaporation. Concentration measurements were performed sequentially in verified reproductive conditions in several points of the room and analysed using a dedicated solution. The measurements obtained were also compared to Computational Fluid Dynamic simulation results.

Paper IV points up the principal outcomes of these investigations.

The last part of this project have been focused on how gathering information on field attribution of a given exposure point on the basis of observable determinants. We selected, in collaboration with a related project focused on a Bayesian approach (Sottas et al., 2005), a series of exposure determinants, generally observed during expert judgments, such as source intensity, source directivity, air turbulence near the source, source velocity, general ventilation, room volume, measurement position. Thus, data (measurements and exposure determinants) collected for several field exposure situations, have been employed to validate a structured approach derived from Gaussian plume dispersion model to obtain a field attribution (near or far) decision index.

Paper V explains this methodology and shows preliminary results.

Results

Paper I

The results obtained from the literature review and the questionnaire analysis stressed out the need to develop a tool able to predict near source concentrations based on observable parameters. It appears that hygienists rely mostly on experience or “so-called” expert judgments, although they give little credit to this method with regards to efficiency and reliability. Long-term sampling is perceived as the most efficient and reliable method. In practice, exposure models are used scarcely to predict exposure. They come at the last rank of exposure methods proposed by the questionnaire. When asked for reasons, occupational hygienists declare in 40% of the cases that models are difficult to apply in specific practical cases; still for the 22% of them, they consider models not accurate and precise enough.

Exposure determinants associated directly with the emission process and dispersion in the near field, used in models, are not so often observed by professionals during an “expert judgment”. Nevertheless they are considered to play an important role in exposure.

A literature review indicated that several emission and dispersion model are available to practitioners. The specificity of the existing emission models and the difficulties in quantifying the ventilation parameters in dispersion models may explain the lack of enthusiasm observed. Most of the surveyed hygienists applying models (coming from research institutions) favour practical approaches to assess emission rate such as a mass balance, which in fact is only applicable to a limited number of cases.

It is believed that a better description of emission and near-field conditions may improve models and enhance their use. Almost 70% of them believe that new developments are required in order to overcome the limitations of the existing exposure models, such as an integration of factors more easily accessible to practitioners. 50% consider also that near field local phenomena are important for operator estimation and that they should be described in more details. Finally they recommend that models for emission estimation should be developed.

Paper II

In a second step exposure models have been applied to assess exposure in a retrospective epidemiological study. The reported cases involved 3 brands of sprays containing a common waterproofing mixture. A wide variability of exposure circumstances was observed: exposure time (spraying time and residence time) ranged between few minutes until 12 hours, as well ventilation condition from an exposure in a poorly ventilated rooms to outdoor situations. However, nearly all exposed individuals reported respiratory symptoms. Other effects were also reported such as digestive troubles, vomiting or abdominal pain, fever and neurological troubles. The average of clinical indicators recorded fall out of the range of acceptable values.

Overspray emission was characterized through experimental measurements: particle size distributions for different products were similar and little differences were found between the toxic products and the apparently non-toxic products commercialised afterwards.

Both resulting assessed doses and concentration levels, calculated through numerical simulations, exhibit large ranges of values of several orders of magnitude, especially for the estimated dose. No evident dose-response relationship was found between exposure indicators and health effects indicators (perceived severity and clinical indicators). A high inter-individual response variability have been observed and the exposure levels obtained indicate that the respirable mists from involved waterproofing sprays have a very low observable effect level (LOEL), compared to the non-toxic one.

These findings suggest that a simple improvement of the exposure conditions during spraying alone does not constitute a sufficient measure to prevent future outbreaks of waterproofing spray toxicity. Although no clear relation was found between exposure and effects, the use of models represented a simple and systematic tool for ranking exposure conditions.

Paper III

Despite the low overall usage of exposure models by professionals in Switzerland, there is an interest in research institution to apply and develop new techniques. In a previous study a good accordance between model predictions and measurements has been found in some field situations, such as printing, ink manufacture and cleaning operations.

In a parallel study, based on a Bayesian framework, the possibility to combine exposure measurements with information on the exposure determinants has been investigated. A physical model, the classical two-zone model, has also been employed, providing a third source of information.

More in the context of exposure prediction, it is also important to report about ongoing efforts in the area of exposure databases. Actually, a database permits since 1991, to record exposure measurements, and at the same time it may support experts in their prediction of exposure in the absence of direct measurement. On the base of observations resulting from the previous survey among Swiss occupational hygienists, a list of exposure determinants have been selected and integrated in the database, permitting a better description of exposure circumstance.

Paper IV

The main results from the investigation on a two-zone model are summarized in the following.

The simplified equations of the two-zone model may be derived from the more general advection-diffusion. These theoretical considerations have shown that concentrations in the two zones correspond to average concentrations, the inter-compartment air exchange depends directly on the definition of near field volume and no hypothesis has been made on the shape of the near field volume. Experimental measurements have evidenced that model predicted concentrations may represent a good appraisal, if near field volume is defined with caution.

Statistical analyses (Kernel density function) have been employed to define and validate (Silverman test) the irregular shape of the “new” near field that appeared to be strongly influenced by ventilation rates. Finally, comparison with Computational Fluid Dynamic has been shown a positive correlation between simulated concentration and measurements. More, the visualization of pollutant dispersion obtained by CFD confirmed the hypothesis about the existence of two compartments of irregular geometries.

These results lead us to the conclusion that from a statistical point of view, the compartmental theory makes sense and simple geometrical shapes are not always suitable to depict near field zones. The consequence is that, to get sound results from a two-zone model, field attribution should be considered as an input variable rather than a known *a priori.*

Paper V

The hypothetical improvements achieved progressively from additional information have been tested according with different statistical approaches, evaluating the agreement between models predictions and measured concentrations.

Except little cases, all selected models overestimate exposures. Further, we found that the two zones model modified – on the bases of all determinants – was the most conservative for all substances. With regard to only dimethyl ethanol amine (DMEA), model predictions are fairly comparable to measurements, especially in the case of models integrating near field observations.

Excluding the isopropyl alcohol, R^2 coefficients show a rather good correlation. Only for isopropyl alcohol no correlation was found, even between measurements and the emission estimates.

This application represents a preliminary illustration of how this kind of approach, based on exposure determinants, may support hygienists in an exposure assessment.

On the base of these results, a model calibration could be required to a better representation of exposure levels. More, further investigations will be useful to enhance the ability of this model.

Paper I

Beliefs and practices in the assessment of workplace pollutants.

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Occupational hygiene, exposure assessment, exposure determinants, exposure models, expert judgement.

Abstract

Objectives

A survey was undertaken among Swiss occupational hygienists and other professionals to identify the different exposure assessment methods used, the contextual parameters observed and the uses, difficulties and possible developments of exposure models for field application.

Methods

A questionnaire was prepared and addressed by mail to 121 occupational hygienists, members of the Swiss Occupational Hygiene Society. A shorter questionnaire was also sent to registered occupational physicians and selected safety specialists. Descriptive statistics and multivariate analyses were performed.

Results

The response rate for occupational hygienists was 60 %. The so-called expert judgement appeared to be the most widely used method, but its efficiency and reliability were both judged with very low scores by occupational hygienists themselves. Long-term sampling was perceived as the most efficient and reliable method. Exposure models were not used very much in Switzerland to predict exposure. Various determinants of exposure, such as emission rate and work activity, were however often considered important by professionals. But they were not directly included in the present exposure assessment processes. Near field local phenomena determinants were also judged important for operator exposure estimation.

Conclusion

Exposure models should be improved to integrate factors, which are more easily accessible to practitioners, including descriptors of emission and local phenomena.

Résumé

Croyances et pratiques dans l'évaluation des polluants sur le lieu de travail.

Objectifs

Une enquête a été entreprise en Suisse parmi les hygiénistes du travail pour identifier les méthodes d'évaluation d'exposition les plus utilisées, les paramètres observés pendant les jugements d’expert et leur niveau d’utilisation, ainsi que les difficultés et les développements possibles des modèles d'exposition.

Méthodes

Un questionnaire a été préparé et adressé par poste à 121 hygiénistes du travail, membres de la Société Suisse d'Hygiène du Travail (un questionnaire réduit a été également envoyé à un groupe de médecins professionnels et spécialistes en sécurité). Des statistiques descriptives ainsi que des analyses multivariées ont été effectuées.

Résultats

Le taux de réponse pour les hygiénistes professionnels était de 60 %. Le jugement d’expert est la méthode la plus usuelle, mais son efficacité et sa précision ont été jugées par les hygiénistes eux-mêmes avec des notes très basses. Le prélèvement à long terme est perçu comme la méthode la plus efficace et la plus fiable. Les modèles d’exposition ne sont pas beaucoup employés dans la pratique en Suisse. Toutefois, certains déterminants d’exposition comme l’émission et l'activité du travailleur sont souvent considérées importantes par des professionnels, mais ces paramètres ne sont pas directement inclus dans les modèles actuels. Les conditions locales ont été jugés importants pour l'évaluation d'exposition.

Conclusion

Les modèles d'exposition existants devraient être améliorés pour intégrer des facteurs plus facilement accessibles aux praticiens, ainsi que les conditions locales et les paramètres d'émission.

Zusammenfassung

Ansichten und Praktiken bei der Schadstoff-Beurteilung auf Arbeitsplätzen

Ziel

Eine Umfrage unter schweizerischen Arbeitshygienekern und anderen Spezialisten sollte Methoden zur Expositionsbestimmung und die dabei verwendeten Bezugsparameter identifizierten. Modellanwendungen, dabei auftretende Probleme und Entwicklungsmöglichkeiten wurden ebenfalls erfasst.

Methode

Ein Fragebogen wurde erstellt und an alle 121 Arbeitshygienekern der Schweizerischen Gesellschaft für Arbeitshygiene verschickt. Eine verkürzte Fassung wurde an registrierte Arbeitsmediziner und eine Auswahl von Sicherheitsspezialisten versandt. Die Resultate wurden mit deskriptiven und multivariaten statistischen Methoden ausgewertet.

Resultate

Die Antwortrate der Arbeitshygieniker betrug 60%. Das sogenannte Expertenurteil war die am häufigsten angewandte Methode, obschon dessen Effizienz und Zuverlässigkeit von den Arbeitshygienekern mit sehr tiefen Noten beurteilt wurde. Langzeitmessungen wurden als die effizienteste und zuverlässigste Methode betrachtet. Expositionsmodelle kommen dagegen in der Schweiz fast nicht zur Anwendung um die Exposition vorherzusagen. Verschiedene Faktoren der Exposition wie Emissionsrate und Arbeitsaktivität wurden dagegen von vielen Fachleute in ihre Betrachtung einbezogen, auch wenn sie nicht direkt im Expositionsbestimmungsverfahren integriert sind.

Schlussfolgerung

Expositionsmodelle sollten durch den Einbezug von Faktoren, die den Fachleuten einfacher zugänglich sind, verbessert werden. Die lokalen Rahmenbedingungen (Nahfeldphänomene?) und die Emissions-Parameter sollten in die Modelle integriert werden.

Introduction

Estimating exposure is an important step in occupational health studies, both retrospective, and prospective. Preventive measures and corrective actions against pollutants exposure at the workplace are frequently based on this estimate. It may also play a key role in the recognition of occupational disease. Exposure estimates to chronic pollutants is the traditional field of occupation hygienists and, at a lesser extent of occupational physicians and occupational safety specialists (e.g. safety engineer).

For chemical exposure, direct measurement is certainly the most reliable and objective way to obtain a reliable assessment of the exposure. It must however be stressed that this approach suffers major drawbacks regarding cost and technical complexity. Furthermore, direct measurements only give information on the current exposure (the day of investigation) and do not allow for past exposure estimation, or exposures under other or future conditions (Nicas,2003).

Because of these difficulties, the assessment of occupational exposure relies more and more frequently on different approaches of varying complexity. Table I gives a short overview of potential methods considered up to now in occupational hygiene. The most simple and most widely used approach is probably the so-called "expert judgment". Occupational hygienists evaluate whether a potential hazard exists by observing workplace conditions and interviewing the exposed workers about the materials used, the production levels, the duration of exposure, existing preventives measures and so on. Exposure assessment is thus based on an interpretation of observations and interviews, integrated with knowledge gathered from previous similar situations, either coming from the specialist’s own experience or from literature reports. Despite its widespread use, there is limited information on the ‘expert judgement’ processes. These subjective estimates are usually unstructured opinions, difficult to explain objectively and to transfer to others (Jayjock, 1997).

In the absence of current monitoring data, semi-quantitative methods have been developed to estimate historical or future exposures. For example a Job Exposure Matrix could represent a practical and less time-consuming method, using historical data through a cross classification of job titles by substances (Dosemeci et al.,1990). This approach is however limited in its details and cannot give information on specific exposure situations. Cherry and Schneider (1999) have developed and validated a structured approach to assess exposure based on descriptive information about work activity and work environment. In this study there was a reasonable association between the estimated exposure level and the measurements, with the correlation between the log-transformed measurements and estimates mostly between 0.5 and 0.9.

A more detailed model was developed in England to better describe workplaces with available historical data (Cherry et al., 2003). EASE (Estimation and Assessment of Substance Exposure) is a semi-quantitative empirical model that gives ranges of potential exposures based on an analysis of exposure measurements contained in the UK National Exposure Database (NEDB). In fact, a selection of exposure determinants is included in the model, and their influence is estimated based on past exposure measurements. This allows the user to make predictions using a simple description of workplaces and processes.

On the other hand, indoor air quality modelling represents a more systematic, transparent and consistent method to integrate numerous parameters. Available deterministic models, based on physico-chemical principles, such as ventilation characteristics, pollutant generation rate, and mass transport mechanisms, provide a convenient way to structure all significant factors determining the levels of exposure.

The Ideal Mixed Model relies very simply on the concept of mass conservation and of homogeneous concentration throughout a single workplace volume.

Table I: Main approaches to exposure assessment, its characteristics and main requirements

Type of method	Main characteristics	Main requirements
Direct measurement	objective	laboratory facility
Expert judgement	subjective	professional experience
JEM	historical	historical data
EASE	empirical	empirical model structure
One-Zone Model	physical, well mixed, compartmental	emission, air-change
Two-Zone Model	physical, compartmental, near field exposure	emission, air-change, inter-compartment flow
Eddy-Diffusion Model	physical, diffusivity	emission, diffusion coefficient
Gaussian Plume Model	physical, directivity	emission, directivity, air velocity
CFD	physical, fluid dynamic and heat transfer	emission, turbulence, momentum effects, buoyancy

This model is one of the older and more known models in occupational hygiene, and its best advantage is its simplicity (Keil 2000). A more complicated workplace description is found in the Two Zone Model (Nicas 1996, 2003, Cherry 1999), which divides the room into two conceptual zones, one near the source (near field) and the other represented by the rest of the room (far field). The Eddy Diffusion Model has notable advantages over the previously described models as it can take into account the gradual decrease of concentrations when moving away from the source (Roach 1981, Wadden et al. 1989). The Gaussian Plume Dispersion model (Mulhausen 1998) is based on a diffusion model that takes into account the direction of air currents. Tools developed by the Environmental Protection Agency (Daniels et al., 2003), as ChemSTEER, Multi-Chamber Concentration and Exposure Model or Wall Paint Assessment Exposure model, are based on several of the above models, and represent therefore combinations of them. Finally Computational Fluid Dynamic (CFD) (Bennet et al., 2003) is a powerful tool that makes it possible to estimate the pollutant's concentration everywhere in a workplace, once the appropriate boundary conditions (like pollutant generation and air flow throughout the room) are specified.

These models can be used to provide specific ventilation requirements under different assumption for production rate, chemical consumption or air mixing conditions (Olcerst, 1999). Models have also been developed for a quantitative reconstruction of historical exposures (Rong et al., Cherry et al. 1999, Kauppinen1994).

The work presented here is part of a larger research project aimed at improving workplace exposure estimations through modelling techniques. The objective is to improve the conditions under which the "expert judgments" take place, by developing (through a revision of the existing models) an assessment tool in accordance with the experts' needs, based on parameters, which are simple and more easily accessible. To identify current job practices as well as the parameters, which are more easily accessible during field investigations, a questionnaire has been proposed to the members of the Swiss Occupational Hygiene Society. The questionnaire explores the methods used in Switzerland to assess chronic and sub-acute exposure to pollutants at workplaces, and identifies the key factors involved in the emission and dispersion phenomena, which are used by practitioners during an exposure assessment.

Methods

In a first phase, a questionnaire was sent to the 121 members of the Swiss Society of Occupational Hygienist. The questionnaire was structured into five different sections specifically targeted at:

(1) appreciate the practitioners' background and basic activities in the field of occupational hygiene and/or exposure assessments,

(2) identify the assessment methods, which are used and perceived as more efficient and more reliable to assess chronic and sub-acute exposure chemical pollutants (such as gas, vapour or dust) at the workplace,

(3) compare the relative importance of the parameters (and their utilisation frequency) observed by the specialists to assess the exposure situation (chronic and sub-acute exposures) during expert judgement (without any objective measurements or empirical or theoretical exposure models),

(4) identify the physico-chemical parameters considered as most relevant by practitioners during quantitative exposure assessment,

(5) assess the use of emission and dispersion models in terms of frequency and perceived accuracy and efficiency, and analyse the needs and the difficulty of the practitioners in using exposure models.

Most questions were multiple-choice questions, with predefined frequency classes or ranks going from 1 (lowest) to 6 (highest).

In a second phase, a similar questionnaire, but reduced to sections 1 to 3, was sent to a selected group of 95 members of the Swiss Society of Occupational Safety involved in exposure assessment and to 169 occupational physicians, members of the Swiss Society of Occupational Medicine.

Global results were analysed by descriptive statistics. In some cases they were analysed by groups in order to identify differences. Then the Chi square test was performed to find possible dependencies between two variables, followed by a factorial analysis of correspondences if necessary. The p-values reported in the text are those obtained from the Chi Square test of dependency.

We report here results obtained after the analysis of occupational hygienists questionnaires. Selected information obtained from the other occupational health specialists is also presented when needed.

Results and discussion

Seventy-seven questionnaires were returned by occupational hygienists. Five of them, which were returned by hygienists not involved in exposure assessment, were blank ones. Positive response rate was therefore 59.5 %. It should however be noted that not all the returned questionnaires were filled completely (only 50 %).

1) Background Information

Surveyed occupational hygienists were equally distributed into the following job categories: advisory or consulting body, industry/service and authority. Fifteen percent could not identify themselves in these categories (most of them in academic research) and some fell into more than one category.

Distribution of the occupational hygienists in various economic sectors is shown in Figure 1. Most occupational hygienists (35 %) are employed in the pharmaceutical and chemical industry. Data about initial training also indicates a similar trend: 59 % of the occupational hygienists have had a first training in chemistry, 11 % in environmental science, 10 % in biology, 8 % in medicine, and 12 % in other fields.

Fifty percent of the occupational hygienists followed the single postgraduate course existing in Switzerland, 9 % followed other international specialized training/courses, and the others specialized through on-the-job training. Seventy-three of the respondents were certified by the Swiss Occupational Hygiene Society.

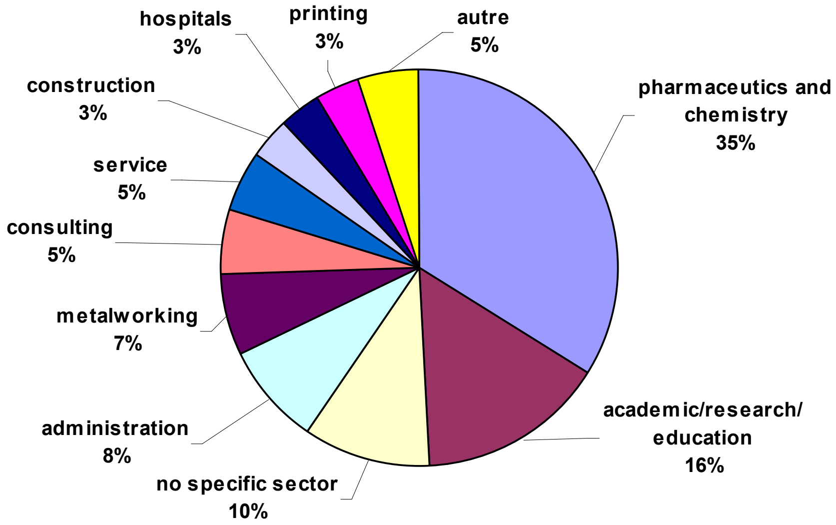

Figure 1: Distribution of the occupational hygienists in the different economic sectors

Most surveyed occupational hygienists have a relatively short experience in the profession, with 50 % having less than 10 years. This has to be put in relation with the only recently introduced legislation in Switzerland, which requires companies to call on occupational hygienists (introduced in 1996, implementation deadline in 2000). A dependency was found between the experience and the way the hygienists get specialized. Hygienists with less than 8 years of experience get specialized through postgraduate course (p value = 0.02) whereas those who have more than 8 years get specialized through practice (p value < 0.01).

An occupational hygiene activity is the main occupation of only 60% of the hygienists. This changes according to the economic sector: for industry/services or authority categories almost 80 % of the hygienists have occupational hygiene as their main activity. The frequencies reported for exposure assessment activities are shown in Figure 2. 34% of hygienists perform exposure assessment weekly or daily. This frequency is linked to the time spent in occupational hygiene activities, 60% of the hygienists whose main activity is occupational hygiene report they perform workplace exposure assessments weekly or daily. Finally, 50 % of the occupational hygienists report assessing exposures in all kinds of environments. As shown in Figure 3, industrial environments are the main focus of exposure assessment activities.

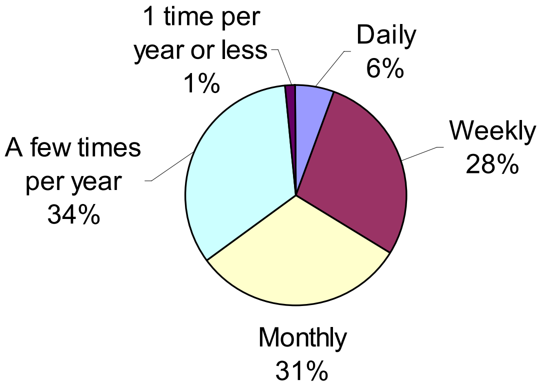

Figure 2: Distribution of the frequency of activity of occupational exposure assessment

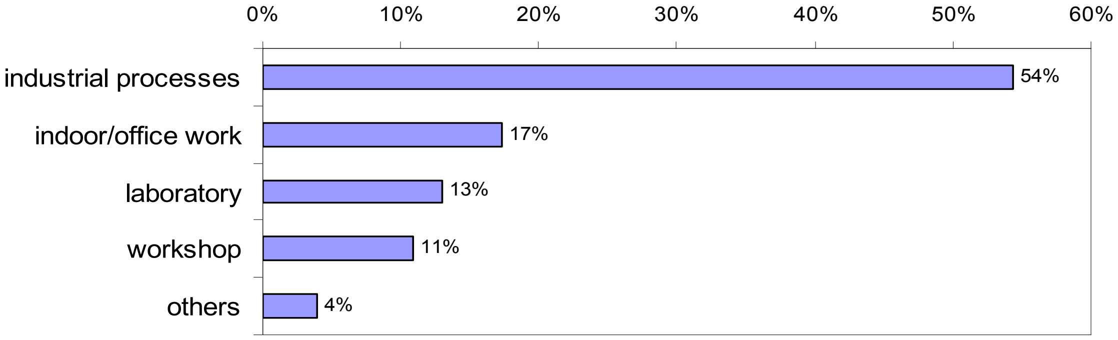

Figure 3: Different workplace environments assessed by hygienists.

2) Methods for Assessing Workplace Exposure

Figure 4 presents the frequencies of use for the different exposure assessment techniques. Exposure models and biological monitoring are seldom used: 60 % of occupational hygienists have never used models while 52 % have never made use of biological monitoring. The results obtained for those making use of models are hardly more encouraging. 30% of hygienists report using model and biological monitoring rarely (in less than 10% of exposure assessment).

These results were somehow expected for biological monitoring, which falls traditionally in the field of occupational physicians. However, the results obtained for exposure models are

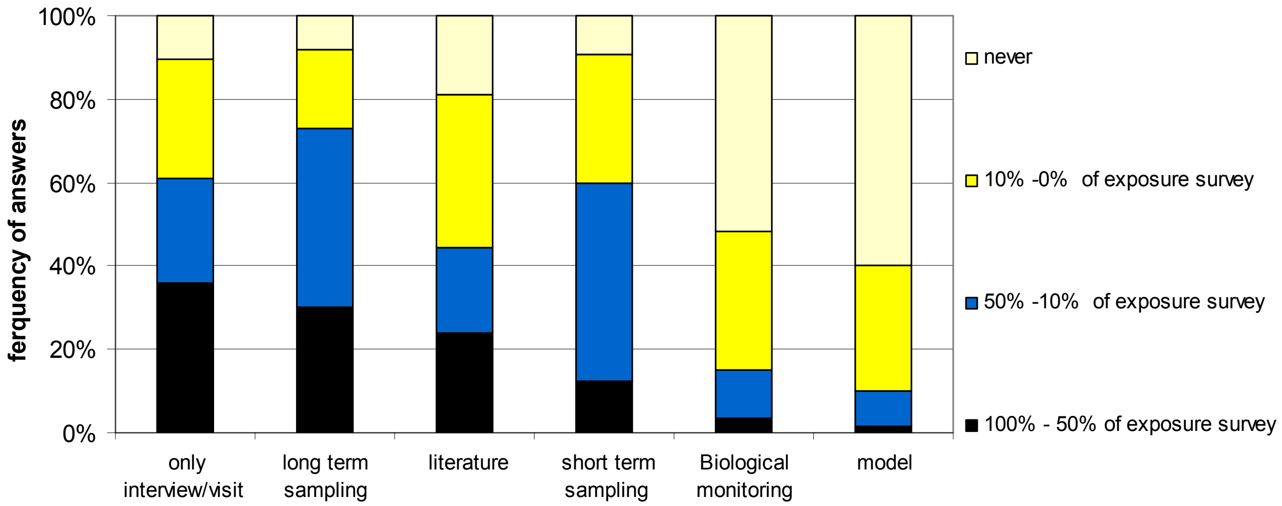

Figure 4: Frequency of use of the different exposure assessment techniques.

surprising. It appears that a significant number of occupational hygienists are unfamiliar with the existing modelling tools and with the modelling capabilities. 30% of them were indeed unable to give a ranking of the models' efficiency and reliability.

The most frequently used exposure assessment techniques are the interview/visit (expert judgement) and the long term sampling. Although the interview/visit method obtained the higher score in frequency of use, it obtained almost the lowest score in efficiency and reliability (only exposure models get a lower score).

Experience plays a significant role in field practice. On the one hand, hygienists having an initial formation in chemistry tend to score expert judgement as less efficient (p value = 0.01). On the other hand, hygienists with more than 8 years of experience frequently use expert judgment frequently (p value < 0.01) and tend to score it as more efficient (p value < 0.01). It is interesting to note that hygienists who have less than 8 years of experience make use of literature more often (p-value = 0.04) and believe it to be more efficient than expert judgment alone (p value = 0.02).

For most of the surveyed hygienists, long-term sampling obtained the best scores, both with regard to efficiency and to reliability. Unsurprisingly, occupational hygienists used exposure measurements (p value = 0.01) whereas occupational physicians employed the biological monitoring more often (p-value = 0.01) and at the same time found it more efficient (p value = 0.02).

Looking at these results after sorting by economic sector, experience or initial training, did not show any evident trend. In the case of safety specialists, exposure judgment is even more used compared to other methods, while literature information is not considered at all.

3) Use of the expert judgement

We have seen previously that occupational exposure assessment relies most frequently on employee interview and/or workplace visit, a so-called "expert judgment". This procedure is often seen as a “black-box” process, a mental process, which is not easily transferable to others (Jayjock, 1997, Schneider 2002). This is also reflected by the fact that, despite its frequent use, specialists have little confidence in it.

To clarify this process, occupational hygienists were asked about the frequency of use of several exposure determinants and their perceived influence on exposure. Eighteen different factors were considered in the questionnaire. They could be divided into 4 classes:

- workplace: room size and shape, natural ventilation, forced ventilation, air currents and direction of air currents within the room;
- emission: rough mass balance, evaporation area, vapour pressure or boiling point for volatiles, composition and dilution, presence of air jet at the source, type of emission process (e.g. grinding, spraying);
- worker’s activity: method and degree of manual handling, frequency of activity intensity , use of personal protective equipment;
- general: general cleanness, sensations such as odours or irritation, movement of people/objects in the room and air temperature gradient in the room.

The results for the 4 groups of parameters are presented in Figure 5. It is shown that occupational hygienists frequently use parameters associated with the worker's activity and also believe them to have an important influence on exposure. These parameters are furthermore easily observable. Parameters of the workplace itself are also very often used by most occupational hygienists, although they are considered to have less influence on exposure. It is interesting to note that these parameters may control exposure only indirectly, by a dilution in the far field, but that they can be easily evaluated.

On the other hand, the elements associated directly to the emission process are not so often observed during an "expert judgement", although they were considered quite important. (These parameters are difficult to quantify, but they play a key role in exposure).

Finally, some parameters describing the general conditions in the workplace, such as air temperature, temperature gradient, movement of people, are perceived as not important and are rarely used.

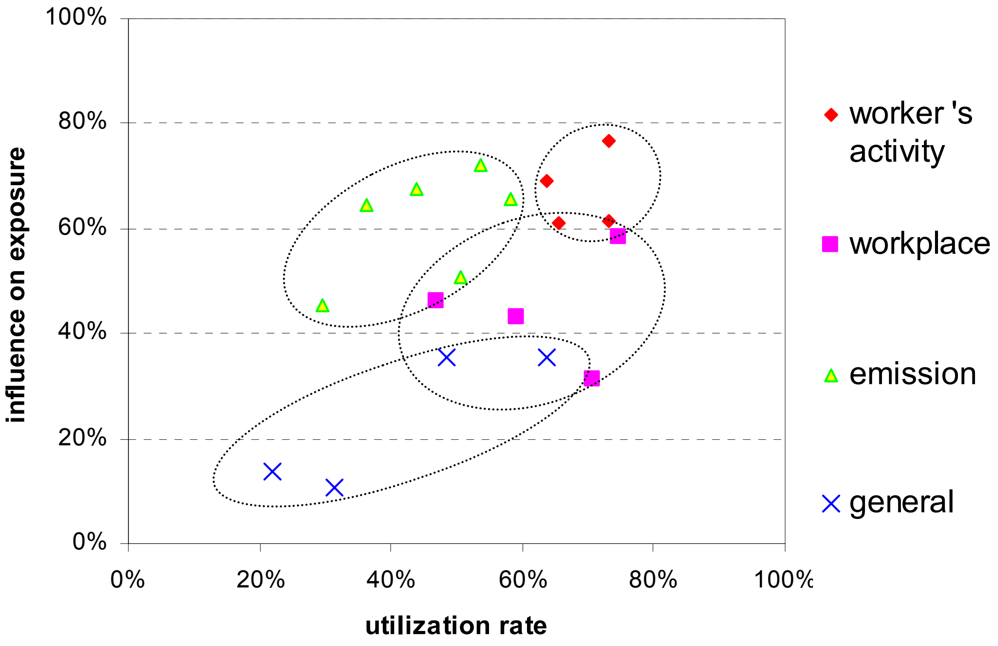

Figure 5: Influence on exposure (% of hygienists answers between score 5 and 6) versus utilisation rate (% of hygienists answers with an utilisation rate between 50 and 100 %) for the different groups of parameters

4) Relevant exposure parameters

This section of the questionnaire was designed to describe how important occupational hygienists consider the various physico-chemical parameters that control emission and dispersion of pollutants.

For solvents, most occupational hygienists selected the higher scores (between 5 and 6) for all factors proposed (vapour pressure, surface of evaporation, air temperature, ventilation near the source, agitation), except for molecular diffusivity. In the case of aerosols, the parameter, which was judged of primary importance, was particle size and distribution. In fact, aerodynamic behaviour of aerosols (such as settling over time, penetration and deposition in the lungs) is strongly dependent on particle size. Still, parameters such as the air velocity and direction at the source, as well as the separation forces associated with the process (grinding, air jet pressure…) also obtained high scores. It is clear that the emission of aerosols is closely related to the energy given to the generation process, such as separation forces (as fracture, abrasion, agitation, for dry aerosols, or atomisation and spraying for the liquid droplet); but it could also depend on the property of the specific material, such as the cohesion forces (the degree of dustiness in the case of a solid, the surface tension forces in the case of a liquid) (Vincent 1995, Reist 1993)

Local ventilation was considered the most effective control measure, controlling worker exposure at the source and preventing migration into the room environment. General ventilation was also judged important, as it ensures dilution of pollutants by providing properly conditioned air.

5) Use of exposure models

The use of predictive models, either of semi-quantitative (e.g- Job Exposure Matrix, EASE) or physical nature (e.g. compartmental, diffusion model) is clearly underdeveloped. 60% of the occupational hygienists never make use of models to assess occupational exposure situations, relying exclusively on qualitative expert judgment or measurements. The reasons given for not using models were mostly their limitations. 40% of them reported difficulties in representing real-life work situation in terms of model parameters. 22% of them invoked the lack of accuracy/precision and the time-consuming process required. Still, it must be stressed that 16% of the hygienists reported they didn't use the predictive model because they didn't know it.

Understanding the use of predictive models amongst practitioners was a prime concern in this study. A full section of the questionnaire was therefore dedicated to this specific topic. Unfortunately, only 28,5 % of the hygienists filled this section. This may easily be explained by the fact that models appear to be used to a limited extent. Moreover, most of the questions implied a relative ranking thus requiring simultaneous knowledge of several of them. The number of answers obtained is too scarce to conduct a statistical analysis or get conclusive results, although some tendencies can be observed.

One concern in the use of models is the difficulty to assess the emission rate correctly. It is interesting to note that hygienists using models favour practical approaches to estimate emissions (i.e. through mass-balance or measurement). As shown in Figure 6, emission rates are usually estimated either through mass-balance, measurement in exhaust air or by using data reported in the literature. The use of specific models is less common, which suggests a limited confidence in the existing predictive emission tools.

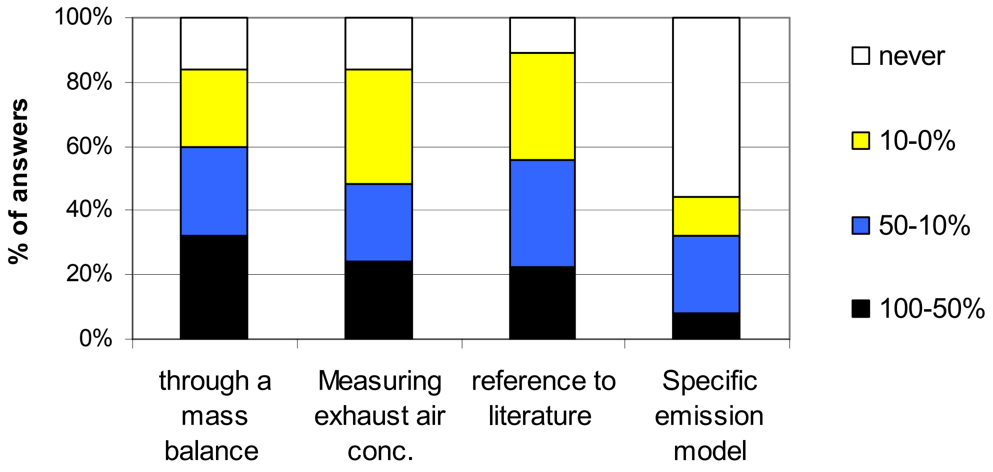

Figure 6: Distribution of utilisation rate of the different methods to identify the generation rate when the exposure is assessed through a model (n=25).

It is assumed that two factors play a key role in the selection of a modelling tool: its accuracy, which should fit with the level of precision required in the assessment, and its effectiveness, namely its capacity to produce usable results at the lowest investment costs (time, resources…). On the one hand, banding approaches (job exposure matrix), compartmental models (ideally mixed, two-zone model), and other physical models supported by user-friendly tools (EPA's tools) are considered as the most efficient because of their straightforwardness. On the other hand, physical models with a certain degree of complexity (two-zone model, EPA's tools, Gaussian model) are considered as the most accurate. Trivial physical models (e.g. ideally mixed) or models working as "black-box" for the user (e.g. EASE) are judged of poor accuracy. This tendency to give more confidence to models based on explicit and comprehensive hypotheses is not verified in the case of computational fluid dynamics. Although it is much more detailed and comprehensive than other methods, CFD is judged of mean accuracy. This result reflects perhaps the lack of confidence practitioners show in using such a complex tool correctly rather than their lack of confidence in the model itself. The number of practitioners acquainted with CFD is unfortunately too limited to draw any conclusion.

Finally, 33% of the hygienists estimated that no further development of models was required, as monitoring was a better alternative anyway, while 67% believe that new developments are required in order to overcome the limitations of the existing exposure models. The two enhancements, which are referred to more frequently are: a better fitting between field and models parameters (70 % of them) and, a better representation of dispersion phenomena near the emission source (50 % of them).

Conclusion

The present survey among Swiss occupational hygienist and other professionals showed that the “expert judgement” is the most widely used method to assess airborne exposure in Switzerland. Looking at exposure determinants, occupational hygienists observe the parameters related to worker’s activity more frequently, as they believe that these factors play a key role in exposure. The parameters associated with the emission and the pollutant behaviour near the source, are also judged very important, but seldom used because of their limited availability during field investigations.

A quantitative characterization of chemical emission sources is not a common practice in the field of occupational health and, consequently is underdeveloped (Jayjock 2005). Description of the pollutant behaviour near the emission source is of particular interest as it is also stated as a prime cause of inaccuracy in the current physical models available. Most models, particularly compartmental models, do indeed take local conditions into account to a very limited extent. Local ventilation conditions or the worker's position are usually either oversimplified or not considered.

Both emission and dispersion models are used only rarely. This is probably linked to the perceived low efficiency and reliability of the existing models. To use deterministic models, even the simpler ones, certain basic parameters must be estimated such as generation rates or ventilation conditions, and in certain cases these estimations could be a serious obstacle. In addition, occupational hygienists also felt that model predictions are not so accurate and precise. It's clear that the precision of a model depends on how much it can adapt to different specific situations, but it's also important to consider how close to the "truth" the model output needs to be to make a decision.

However, about 70 % of the occupational hygienists using models agreed on the necessity to develop models further. They think that the most beneficial improvements of exposure models would be to include input parameters, which are more accessible during field investigations. Near source phenomena should also be taken into account more.

Despite this low overall usage of exposure models by practitioners in Switzerland, there is an interest in research institution to apply and develop new techniques (Bruzzi 2005, Sottas 2005, Vernez 2005) in agreement with the current European and American trends (ISSA 2004). As a result of this questionnaire, future models should be more concentrated on near field conditions and at the same time they should integrate some parameters, which are more easily available during practical surveys.

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References

1. Bennett J.S., Feigley C.E., Khan J.A., et al. (2003). Comparison of Emission Models with Computational Fluid Dynamics and a Proposed Improved Model. Am Ind Hyg Assoc J 64: 739-754.
2. Bruzzi R., Vernez D., Sottas P.E. & Droz P.O. (2005). Exposure models in Switzerland. An overview of the present situation. Gefahrstoffe Reinhaltung der Luft (in press).
3. Cherrie J.W. (1999). The Effect of Room Size and General Ventilation on the Relationship Between Near and Far Field Concentrations. Appl Occup Environ Hyg 14(8): 539-546.
4. Cherrie J.W., Schneider T. (1999). Validation of a New Method for Structured Subjective Assessment of Past Concentrations. Annals of Occupational Hygiene 43 (4): 235-245.
5. Cherrie J.W., Tickner J., Friar J., et al. (2003). Evaluation and further development of the EASE model 2.0. Norwich, Her Majesty's Stationery Office.
6. Daniels W., Lee S., Miller A. (2003). EPA's exposure assessment tools and models, Appl Occup Environ Hyg 18(2): 82-86.
7. Dosemeci M., Stewart P.A., Blair A. (1990). Tree Proposal for Retrospective, Semiquantitative Exposure Assessments and their Comparison with other Assessment Methods. Appl Occup Environ Hyg 5(1): 52-59.
8. Issa Health Service Section (2004). Models and calculation methods to determine exposure to dangerous substances. Conference Review, Dresden: CDROM.
9. Jayjock M.A(1994): Backpressure modelling of indoor air concentrations from volatilising sources. Am Ind Hyg Assoc J 55: 230-235.

10. Jayjock M.A. (1997). Modelling Inhalation Exposure. In: Di Nardi S.R. The Occupational Environment – its Evaluation and Control. Fairfax, AIHA: 69-74.

11. Jayjock M.A, (2005). Exposure Modelling Research – First Things First. The Synergist 16(2): 34.

12. Keil, C.B. (2000). Mathematical models for estimating occupational exposure to chemicals. Fairfax: AIHA.

13. Mulhausen J. R., Damiano J. (1998). Strategy for Assessing and Managing Occupational Exposure. Fairfax: AIHA.

14. Nicas M. (1996). Estimating Exposure Intensity in an Imperfectly Mixed Room. Am Ind Hyg Assoc J 57: 542-550.

15. Nicas M. (2003). Using mathematical models to estimate exposure to workplace air contaminants. Chem Health Saf (January February): 14-21.

16. Olcerst, R. (1999). "Use in well ventilated area?". Am Ind Hyg Assoc J 60:377-383.

17. Reist P. C. (1993). Aerosol Science and Technology. New York: McGraw-Hill.

18. Roach S. A. (1981). On the role of turbulent diffusion ventilation, Annals of Occupational Hygiene (24)1: 105-132.

19. Rong C. Y., WaiYuan T., Mathew R. M., et al. (1990). A Deterministic Mathematical Model for Quantitative Estimation of Historical Exposure. Am Ind Hyg Assoc J. 51 (4):194-201.

20. Schneider T. (2002). Improving exposure assessment requires measurements and modelling. Scand J Work Environ Health 28(6): 367-369.

21. Sottas P.E., Bruzzi R, Vernez D., et al. (2005). A Bayesian framework for combining different occupational exposure assessment methods. Proceeding of IOHA Conference.

22. Kauppinen T.P., Pannett B., Marlow D.A., et al. (1994). Retrospective assessment of exposure through modelling in a study on cancer risks among workers exposed to phenoxy herbicide, chlorophenols and dioxins. Scand J Work Environ Health, 20: 262-271.

23. Vernez D., Bruzzi R., Kupferschmidt H., et al. (2005). Acute respiratory syndrome after inhalation of waterproofing sprays : a posteriori exposure response assessment in 102 cases, submitted in J Occup Environ Hyg.

24. Vincent J. H. (1995). Aerosols Science for Industrial Hygienists. New York: Pergamon Press.

25. Wadden R.A., Scheff P.A., Franke J.E. (1989). Emission Factors for Trichloroethylene Vapor Degreasers, Am Ind Hyg Assoc J, 50(9): 496-500.

Paper II

Acute respiratory syndrome after inhalation of waterproofing sprays: a posteriori exposure-response assessment in 102 cases.

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Keywords:

airborne particles, exposure assessment, fluororesins, respiratory diseases, waterproofing spray

Abstract

Waterproofing agents are widely used to protect leather and textiles in both domestic and occupational activities. An outbreak of acute respiratory syndrome following exposure to waterproofing sprays occurred during the winter 2002-2003 in Switzerland. About 180 cases were reported by the Swiss Toxicological Information Centre between October 2002 and March 2003, whereas less than 10 cases per year had been recorded previously. The reported cases involved 3 brands of sprays containing a common waterproofing mixture, which underwent a formulation change in the months preceding the outbreak.

A retrospective analysis was undertaken in collaboration with the Swiss Toxicological Information Centre and the Swiss Registries for Interstitial and Orphan Lung Diseases to clarify the circumstances and possible causes of the observed health effects. Individual exposure data were generated with questionnaires and experimental emission measurements. The collected data was used to conduct numeric simulation for 102 cases of exposure. A classical two-zone model was used to assess the aerosol dispersion in the near and far-field during spraying. The resulting assessed dose and exposure levels obtained were spread on large scales, of several orders of magnitude. No dose-response relationship was found between exposure indicators and health effects indicators (perceived severity and clinical indicators). Weak relationships were found between unspecific inflammatory response indicators (Leukocytes, C-reactive protein) and the maximal exposure concentration. The results obtained disclose a high inter-individual response variability, and suggest that some indirect mechanism(s) predominates in the respiratory disease occurrence. Furthermore, no threshold could be found to define a safe level of exposure. These findings suggest that the improvement of environmental exposure conditions during spraying alone does not constitute a sufficient measure to prevent future outbreaks of waterproofing spray toxicity. More

efficient preventive measures are needed prior to the marketing and distribution of new waterproofing agents.

Introduction

Fluorinated polymers are widely used in a number of technologies requiring low surface energy, such as coating surface applications. The high electronegativity of fluorine strongly affects the molecules physical and chemical properties (1). Amongst other effects, the presence of fluorine tends to reduce surface tension and enhance thermal and chemical stability. Fluoro-acrylate polymers, which exhibit a high stability and durability, are increasingly used in coating. Diluted into solvents of low polarity, the polymers may be used to coat various surfaces either in liquid or aerosol application (spraying).

There is strong evidence that inhalation of waterproofing spray can lead, in certain circumstances, to respiratory disorders. Outbreaks of respiratory failure following the use of waterproofing sprays occurred in Germany between 1979 and 1983 (2,3), and in the United States, Canada and Japan in 1992-1993 (4,5,6). A recent case was also reported in Japan (7). Each outbreak closely followed the marketing of a product, which underwent a formulation change of the solvent (to eliminate ozone-depleting solvents) and the fluorinated polymer (to increase solubility in the new solvent). Clinical and experimental findings of previous studies suggest that the new formulation may have played a central role in pathogenesis because of the direct pulmonary toxicity of the new fluorinated resins or a possible increase in the amount of respirable fluororesin particles emitted (8,9). The mechanisms of pulmonary toxicity of waterproofing sprays are not yet well understood. Short-term management of previous outbreaks was mainly based on the removal of incriminated products from the market, but this strategy did not prevent new outbreaks to occur later with similar waterproofing agents. Instead, the periodical recurrence of toxicity outbreaks suggests that safety issues in the development of coating mixtures have so far followed a trial-and-error process, rather than a long-term anticipatory and preventive strategy.

A new outbreak of respiratory illness due to waterproofing sprays occurred recently in Switzerland (10, 11). More than 180 cases were reported between October 2002 and March 2003, whereas 10 cases per year had been observed in the previous years. Although various commercial products were involved, they had a common waterproofing agent: a mixture of fluorinated acrylate polymer and isoparaffinic hydrocarbons, which underwent a formulation change shortly prior to the outbreak. The same waterproofing agent appeared to be involved in a simultaneous outbreak reported in the Netherlands (12) and in a fatal case reported from France (13). A fatal case occurred also in the UK (14) at about the same period and under similar conditions.

Most of the incidents observed in Switzerland occurred after domestic activities, following the application of leather and textiles waterproofing sprays. Three occupational cases following the use of a stain-repellent resin on stone-tiled walls and floors were also reported (15). The exposure conditions of these three cases were investigated in a previous study (16). Emission measurements and simulations indicated that (1) significant aerosol and solvent concentrations may occur during waterproofing, and that (2) the amounts of solvent and particles in the workers' breathing zone were lower with the new resin formulation. This last result strongly suggests that the respiratory illness is related to the fluorinated polymer itself rather than an increase of the exposure level to solvents and particles.

The toxic mechanism involved is unclear and several hypotheses can be suggested. On the one hand, the polymer particles may directly exert their waterproofing effect on the alveolar surface, thereby increasing alveolar surface tension, counteracting the effect of surfactant, and leading to alveolar collapse and impairment in gas exchange as previously suggested (17). This hypothesis is somehow supported by the polymer stability and the absence of a polymerisation reaction during the formation of the coating layer (evaporation only). On the other hand, an indirect mechanism requiring a metabolic activation with or without interaction with other factors (i.e solvents, smoking) may also take place. Previous examples of such interactions have been reported in the case of polytetrafluoroethylen (Teflon) for instance (18).

Although the commercial products involved in the Swiss outbreak have been withdrawn from the market, waterproofing agents remain widely used. Moreover, new polymers and product formulations are regularly developed and marketed. The periodical recurrence of respiratory disease observed with these products is therefore a long-term concern for both public and occupational health. Understanding the conditions under which the illness occurs is of high interest to better prevent and control future outbreaks.

The Institute of Occupational Health Sciences (IST), the Swiss Toxicological Information Centre, and the Swiss Registries for Interstitial and Orphan Lung Diseases undertook a joint study of the 2003 Swiss outbreak. Exposure conditions and health effects were investigated in a retrospective way through questionnaires, emission measurements and numeric simulation. The main objectives were to characterise the exposure conditions during spraying and the possible relationship between exposure and observed health effects, in order to clarify the causes of the outbreak and formulate preventive recommendations.

Methods

Questionnaires

The Swiss Toxicological Information Centre and the Swiss Registries for Interstitial and Orphan Lung Diseases have systematically investigated the reported cases through questionnaires. Each exposed individual received a questionnaire covering the exposure conditions and the perceived intensity of the respiratory reaction (patient's questionnaire). The questionnaire asked for the type of waterproofing agent used (commercial name), the spraying activity (approximate spraying time, approximate amount of product used, items sprayed), the exposure environment (exposure location, room dimensions, open windows and doors, time spent in the same room after spraying) and perceived health effects (symptoms, time before occurrence, time before medical care, duration). Additional questions regarding potential contributing or confounding factors, such as smoking habits, were also included in the questionnaire.

Data of clinical findings were collected from patients who underwent medical examination and diagnostic procedures. Patients were asked to send the medical documents in their possession (laboratory results reports, chest X-ray), and questionnaires were sent to their physicians (physician's questionnaire). Common clinical parameters were extracted from these questionnaires and documents. They included severity parameters on admission (dyspnoea levels, respiratory rate, symptoms observed, need for supplemental oxygen) as well as objective clinical parameters (C-reactive protein, white blood cells (WBC) and arterial PO2 levels). These parameters, if available, were used as severity indicators of health effects. The clinical features of the pulmonary toxicity syndrome as well as the control of the outbreak by Public Health authorities will be described in detail in forthcoming papers.

Three subjective indicators of exposure effects have been considered in this study: the delay before medical care (DELAY), the perceived symptoms (SCORE) and the dyspnoea score (DYSP). The delay before medical care depends strongly on the severity of perceived effects from the patient's point of view. The more serious the patient believes the situation is, the more likely he will ask for urgent medical assistance. The symptoms reported by the patients were categorized according to the affected system: general (fever, shivers or myalgias), respiratory (cough or dyspnea), neurologic (giddiness, headache, or loss of consciousness), digestive (nausea, vomiting or abdominal pain) and Eyes/Ear-Nose-Throat (ENT) (burning eyes or throat). An arbitrary index of disease severity was used, one point was attributed to each affected system (i.e. a system for which one or more symptoms were present) and the number of systems affected was added to produce a symptom score. Thus a score of one indicates that symptoms were reported in only one system, while a score of five indicates that symptoms were present in all systems. The New York Heart Association dyspnoea score is a widely used medical rating of the severity of dyspnea ranging from I (shortness of breath on heavy exertion) to IV (shortness of breath at rest). The score used is the one established at the first medical examination.

Emission rate during spraying

The amount of respirable particles emitted during spraying must be known in order to assess aerosol exposure. An estimate based on a theoretical approach is quite complex in the case of volatile aerosol emissions because key parameters, such as the diameter of droplets and their velocity, become time dependent. Moreover, the initial size distribution of the particles is strongly dependant on the physico-chemical properties of the product and the discharge conditions (pressure, nozzle size). Because of this, the use of theoretical models, such as the one proposed by Flynn (19) to predict transfer efficiency from compressed air spray guns during painting, is limited. The spray cans used in our study may indeed differ significantly from air sprays guns.

An experimental approach, based on the measurement of the overspray, was therefore used. The experiment was similar to the one used to assess the transfer efficiency of the nebulizer-spray proposed by Tan and al. (20). The spraying was performed in a 7.9 m^3 booth with a constant descending laminar airflow (Figure 1). The air renewal of the booth was of 9.7 per hour. During spraying, the large particles impacted on ground surface while the smaller particles, constituting the overspray mist, escaped through the perforated floor plate. Overspray aerosol concentrations C(t) were measured in the exhaust duct at a downstream distance of about 5 meters. It is assumed that, at this point, the volatile compounds of the particles have been evaporated during the transport process (16). Aerosol concentrations and distribution were measured with a light-dispersion based device: a Grimm Dust Monitor (model 1.102, Labortechnik GmbH, Ainring, Germany).

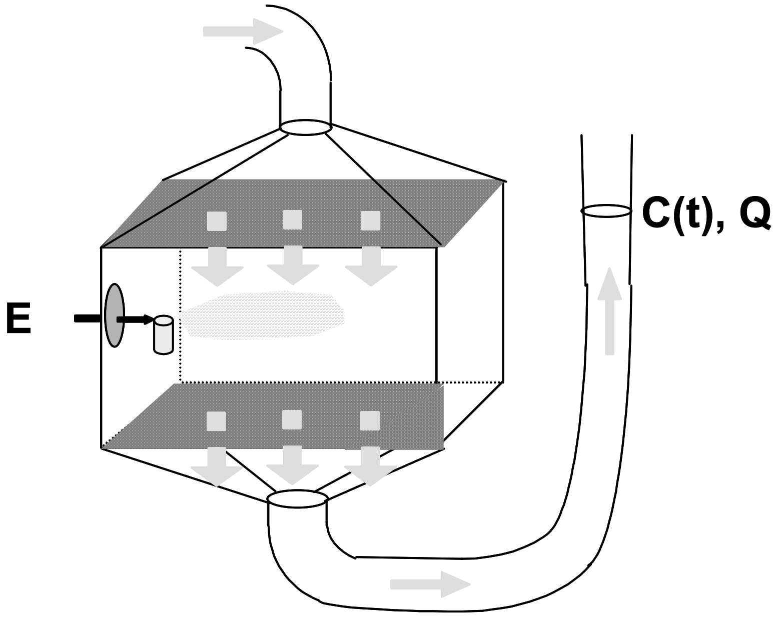


Figure 1. Schematic view of the ventilated booth.

The booth was separated from the laboratory by airtight doors, and a slight depression (10 Pa) was maintained in it to avoid any leakage during the experiment. An airtight glove system allowed the experimenter to use the spray from outside. As shown in Figure 2, the spray was introduced into the booth using repetitive short emission pulses. This “discontinuous emission” procedure was intended to avoid a significant temperature drop of the spray cans, which decreases the emission rate. It is also advantageous because it lengthens the possible duration of the experiment per spray can. As shown in Figure 2, the instantaneous emission rate E_i may easily be deduced from the cycle time (t_1) and emission time (t_2)

$$E_i = \frac{t_1}{t_2} \cdot E \qquad \text{(Equation 1).}$$

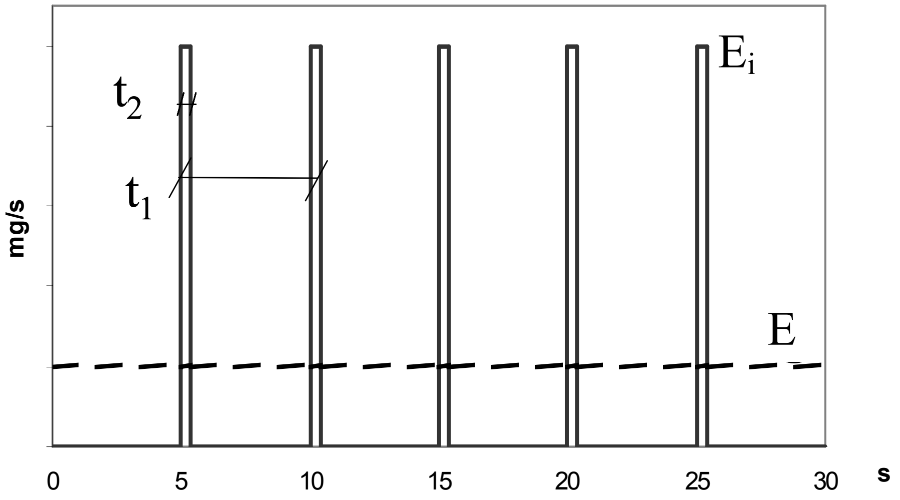


Figure 2. Effective and measured spray emission.

As very few of the original cans were available, preliminary experiments were therefore performed with commercial waterproofing sprays currently available on the market. These tests aimed to define the measurement protocol and set up the experimental parameters. A 5 seconds cycle time (t_1) was chosen. Each 5 seconds a short spray pulse was emitted into the booth. The experiment was recorded on a digital camera (DCR-TRV7E, Sony Corporation, Japan) and analysed in slow motion replay. The average pulse duration obtained (emissions duration, t_2) was 0.42 seconds.

Using these parameters, a steady aerosol concentration may be obtained within the booth in about 10 minutes before emptying out a spray can. When the concentration in the exhaust duct reaches a constant value (steady state), the amount of overspray emitted may easily be deduced from a mass balance equation:

$$E = C_{duct} \cdot Q_{duct} \qquad \text{(Equation 2)}$$

The preliminary experiments were also used to validate the aerosol measurement method. Results obtained from the Grimm Dust Monitor were compared with those of a Personal Data

Ram (PDR, global concentration) and of an Andersen impactor (particle distribution). The average variations for fine particles (<10μm) were of 12.6 % for the PDR and 8.9 % for the Andersen. These differences are not relevant in comparison with the uncertainties of other simulation parameters (such as the spraying time), which were established on the basis of patient's questionnaires. Moreover, they may easily be explained by the slight difference in the working ranges between the measuring devices.

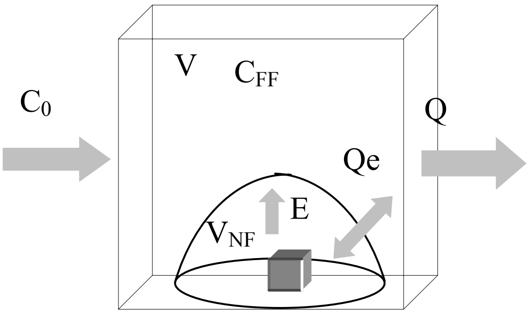
Modelling of exposure concentrations.

As this study focused on pulmonary alveolar-level effects, our concern regarding particulate matter was limited to respirable aerosols (<10 μm). Due to their limited mass and size, fine particles are not affected significantly by the gravitation and aerodynamic forces shortly after their emission and thus, behave in a similar way to gases with regard to their transportation and dispersion. Classical gas dispersion models can therefore be used to assess the respirable aerosols concentrations in the breathing zone at the time of exposure.

The well-known Two-Compartment Model (Figure 3) was used in this study (21). The choice of this compartmental model has been based on practical considerations. On the one hand, only models based on simple parameters, accessible through questionnaires or literature, can be used in such retrospective study. On the other hand, the simplest compartmental model, the Well-Mixed Room Model, which considers a uniform concentration through the room, may severely underestimate the exposure near the source (22). The Two-Compartment Model considers two ideally mixed dispersion volumes: the near-field zone (NF), containing the emission source including the individuals' breathing zone, and the far-field zone (FF) representing the remaining part of the room. Near and far-field zones are interconnected by an inter-compartment flow (Qe), which ensures the air and pollutant circulation inside the room. The model used considers air renewal in both near and far-field, although variations due to local geometrical effects, such as the spray orientation can not be taken into account. The evolution of the pollutant concentration into the two compartments is given in the following equations.

$$V_{NF} \cdot dC_{NF} = (E + Q_e \cdot C_{FF} - Q_e \cdot C_{NF}) \cdot dt \qquad \text{(equation 3)}$$

$$V_{FF} \cdot dC_{FF} = (Q_e \cdot C_{NF} - [Q + Q_e] \cdot C_{FF}) \cdot dt \qquad \text{(equation 4)}$$



compartment Volume V [m^3]
incoming conc. C_o [mg/m^3]
Far-field conc. C_{FF} [mg/m^3]
Near-Field conc. C_{NF} [mg/m^3]
ventilation flow Q [m^3 /s]
emission coefficient E [g/s]
near-field volume V_{NF} [m^3]
inter-compartment flow: Qe [m^3 /s]

Figure 3. Schematic view of the two-compartment model surrounding a punctual emission source (the grey cube).

Data from questionnaires

Patient’s questionnaires were returned for 105 cases (return rate 52 %). 3 of them, in which mandatory data was missing or inaccurate, were discarded. The exposure conditions and/or clinical data reported in the 102 remaining cases were analysed.

Products

The products involved were mostly commercial spray cans intended for domestic or light occupational waterproofing activities. RapiAquaStop (Werner & Mertz GmbH, Mainz, Germany) was the most frequently involved spray (46% of cases). The two other sprays reported were K2R (K2R Produkte GmbH, Gottmadingen, Germany) and RapiIntemp

(Werner & Mertz GmbH, Mainz, Germany) in respectively 27% and 12% of cases. A combination of several products was used in the remaining cases. In two cases the product name was not remembered or not known. One occupational exposure occurred with Patina-Fala (PATINA-FALA Beizmittel GmbH, Haar, Germany), a liquid stain-repellent mixture, when coated with a manual trigger spray. This specific case has been addressed in a previous study (16). The four involved products underwent a formulation change in both solvents and polymer prior to the incidents. A common waterproofing agent was present in all of them: a mixture of fluorinated acrylate polymer and isoparaffinic hydrocarbons.

Exposure conditions

The exposures took place in an outdoor environment surprisingly often, 14 % occurred in open-air and 32 % in a partially open area such as a terrace or a balcony. Indoor environments were reported in 54 % of the cases. Ventilation (either natural or forced) was present in most of them (92%). No ventilation (no open door, no open window) was reported in only 8 % of the indoor cases.

The average volume of the rooms in which spraying took place was 49 m^3 (ranged between a minimum of 5.7 m^3 and a maximum of 250 m^3, in the case of a garage). 80% of the exposures took place in rooms of less than 75 m^3. The spraying times ranged from a few seconds to 90 minutes, while the residence time (time spent in the same room after the spraying activity) ranged from 0 to 12 hours. 80% of the exposure times were shorter than 20 minutes and 80% of the residence times were shorter than 25 minutes. The distribution of reported spraying duration and total exposure duration (spraying time + residence time) are shown in Figure 4. The exact duration is difficult to assess retrospectively and a significant uncertainty is to be expected with these two parameters. This uncertainty is however mitigated by the wide range of values reported, which falls within several orders of magnitude.

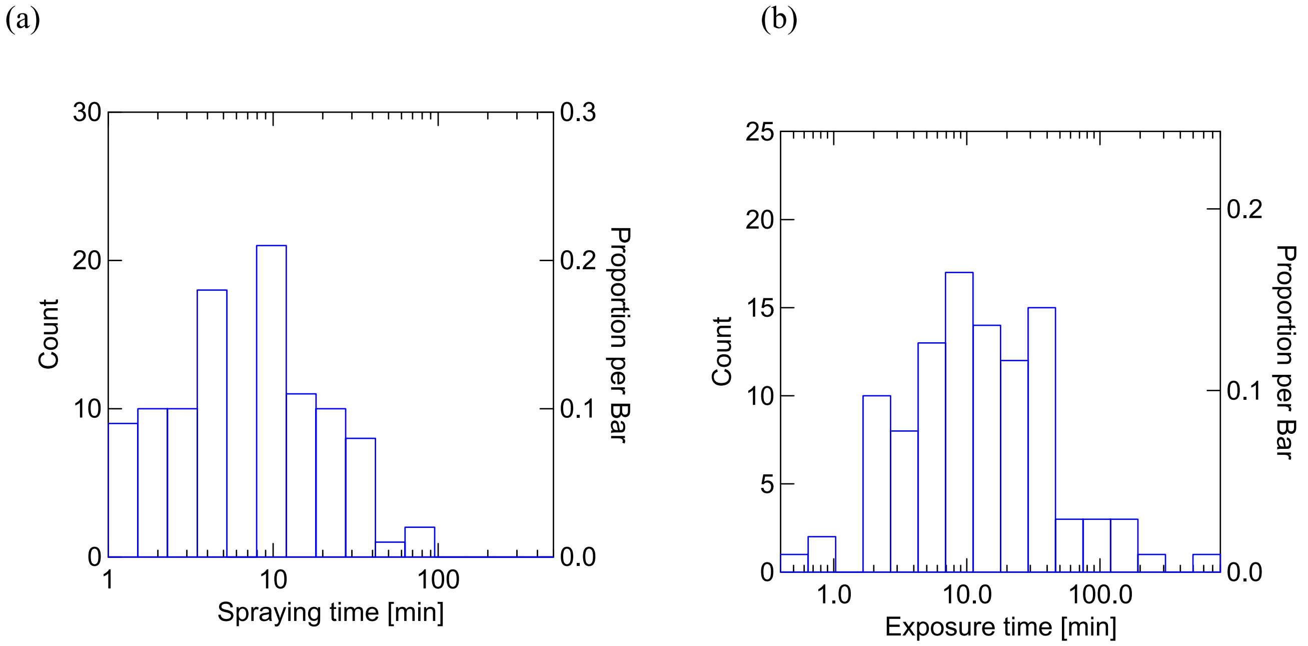


Figure 4. Distribution of the reported exposure time: (a) spraying time, (b) total exposure time in the reported cases. The number of corresponding cases is given by y-axis (count)

Effects

Nearly all exposed individuals reported respiratory symptoms such as cough or dyspnoea (98 % of cases). 22% had digestive troubles, such as nausea, vomiting or abdominal pain. 37% experienced general symptoms like fever, shivers or myalgias. 40% had neurological troubles such as giddiness, headache or loss of consciousness. Eyes or throat burning were reported in 20% of cases.

For 20 % of the exposed individuals, the symptoms were serious enough to require emergency hospital admission. Another 32% received ambulatory medical care, either from their regular physician or a hospital facility. The remaining 48% did not request medical attention.

The medical units carried out various diagnostic procedures. Three of them, which were frequently performed, were of particular interest in this study (each of them was performed in about 25-30 % of the cases). Two non-specific markers of inflammatory response were considered: the white blood cell count (WBC) and the serum C-reactive protein (CRP) concentration. The arterial partial oxygen pressure (PaO_2), reflecting pulmonary gas exchange, was also considered a marker of lung damage and impaired respiratory function. When diagnostic procedures were repeated several times for the same patient, the clinical value considered and discussed here below corresponds to the extreme observed (max for WBC and CRP, min for PaO_2). The white blood cell count (WBC) ranged between a minimum of 6.0 G/l and a maximum of 26.6 G/l with an average of 15.4 G/l (normal values 4-9 G/l). The CRP concentrations ranged between a minimum of 3 mg/dl and a maximum of 264 mg/dl with an average of 59 mg/dl (normal values <5 mg/dl). The PaO_2 while breathing room air ranged between 38 and 102 mmHg, with an average of 66 mmHg (normal values >80 mmHg).

47% of the involved individuals were active smokers, 25% were former smokers, and 28% had never smoked. Amongst the 64 cases in which a clinical assessment was available, 23% had a history of allergy, and 14% had a history of asthma or chronic obstructive pulmonary disease (COPD).

Experimental data

Emission rate during spraying

The average aerosol concentrations measured were 1770 $\mu g/m^3$ for RapiAquaStop and 2390 $\mu g/m^3$ for K2R. Considering an exhaust flow of 0.021 m^3/s (Q duct), the amount of overspray emitted (E) may easily be obtained using Equation 2. The total mass emitted was measured by gravimetry. The spraying cans were weighted before and after emission experiment. An instantaneous overspray emission of 0.19 mg/s, corresponding to 0.073 % of the total mass of the emitted product, was found for RapiAquaStop. An instantaneous overspray emission of 0.25 mg/s, corresponding to 0.124 % of the total mass of the emitted product, was found for

K2R. Typical particles size distribution for K2R and RapiAquaStop are shown in Figure 5 (distributions are expressed here in mass fraction and not in particles count). Particle size distributions for both products are similar and little differences were found between the toxic products and the apparently non-toxic products marketed afterwards. Differences in overspray emission rates were found between toxic and non-toxic products, although they tend to diverge. The fraction of overspray in the emitted product was higher for RapiAquaStop (about 0.15%) and lower for K2R (about 0.01%). No can of RapiIntemp, the third waterproofing spray, was available. As RapiIntemp and RapiAquaStop are comparable products delivered in similar cans, it has been assumed that their emission characteristics were similar.

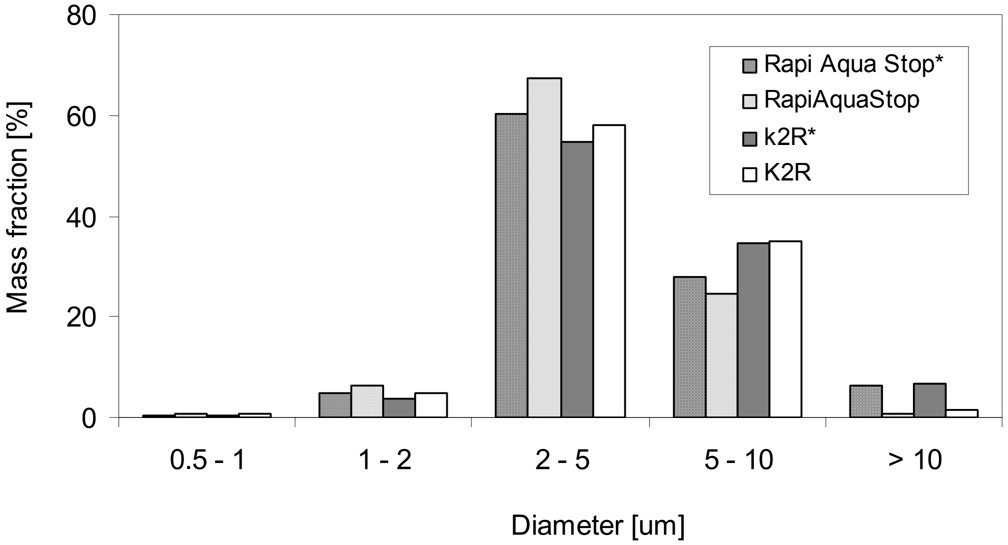


Figure 5. Example of particle size distribution obtained during spraying tests (*products involved in the toxicity outbreak as compared to similar non-toxic products)

In practice, the mean emission rate is lower than the instantaneous emission rate as the spray is not activated permanently. It has been estimated that, during textile or leather waterproofing activities, the spray was activated about 50% of the time. A mean emission rate corresponding to 50% of the instantaneous emission measured has therefore been considered in this study. A different ratio was used for 35% of the cases, where the reported spraying time was too high when compared to the amount of product available. When the spray had been obviously used less than 50% of time, the mean emission rate was adjusted according to a simple mass balance relationship (mean emission rate = amount of product used . percentage of overspray / reported spraying time).

Model implementation

The dispersions were modelled through numerical simulations using Ithink (version 7.0.2, HPS High Performance Systems, Inc., Hanover, NH). The spray emission rates measured experimentally were introduced into the two-zone model. The spraying conditions described in the questionnaires were used to set the various parameters required in the two-zone model. The room volume, spraying time and residence time were depicted by quantitative parameters in the questionnaires and could therefore be used as such in the numeric simulation. Parameters related to the ventilation conditions (air renewal) and inter-compartment exchanges were assessed on the basis of qualitative information about the number of openings in the room (windows or doors) and their connected spaces (outdoor connection or connection with another room)(23). The conditions reported were categorized in a reduced number of ventilation scenarios following the rules given in Table 1.

Two exposure times were considered to assess the breathed dose. The spraying time, during which the person was exposed to a near-field concentration, and the residence time, during which the exposure level was of a far-field concentration. A typical example of concentration and dose profile obtained from simulation is presented in Figure 6. In the case of outdoor exposures, the far-field volume was considered as infinite and the exposure during residence time was negligible.

Ventilation conditions	Qe (m^3/s)	Air Renewal (1/h)
indoor without ventilation	0.14	1
indoor with ventilation	0.20	2
location open on the outside	0.26	3
outdoor	0.32	-

Table 1. Implementation values for simulation.

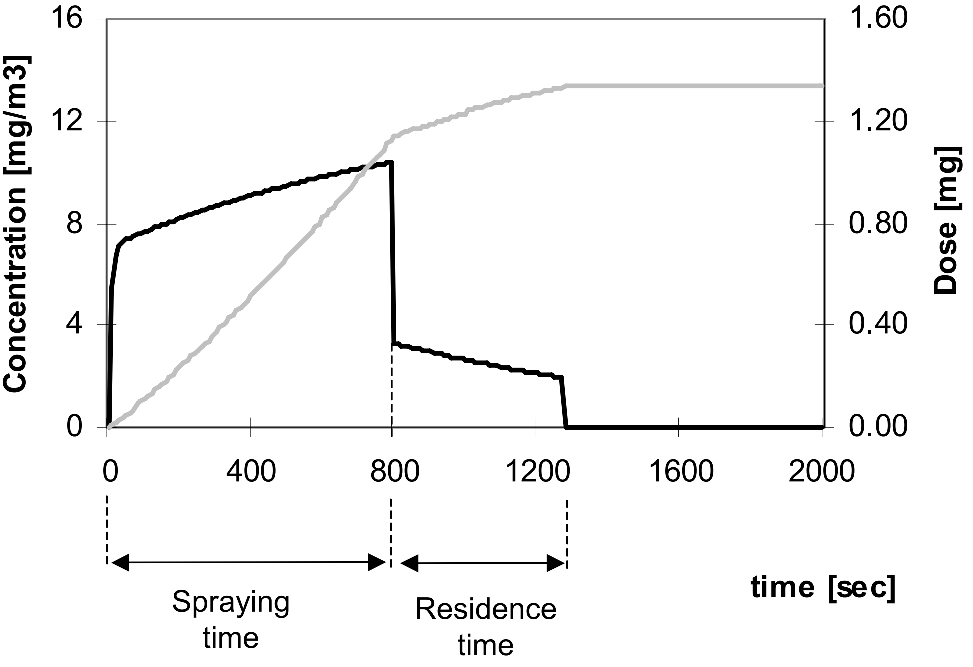


Figure 6. Typical concentration and dose profile

Results and discussion

Exposure assessment

An overview of the results obtained using the two-zone model is shown in Figure 7. The maximal concentrations assessed ranges from 0.003 mg/m3 to 35.98 mg/m3 (mean value 4.21

mg/m^3) while the estimated doses range from 0.2.10-5 mg to 11.27 mg (mean value 0.657 mg). The two distributions are of approximately lognormal shapes, with a score of surprisingly low values. In a general sense both assessed doses and concentrations exhibit wide ranges of values. The array of values is particularly large for the estimated dose, where five orders of magnitude separate the upper and lower limits. This scattering mostly results from the variety of spraying and residence times reported in the questionnaires.

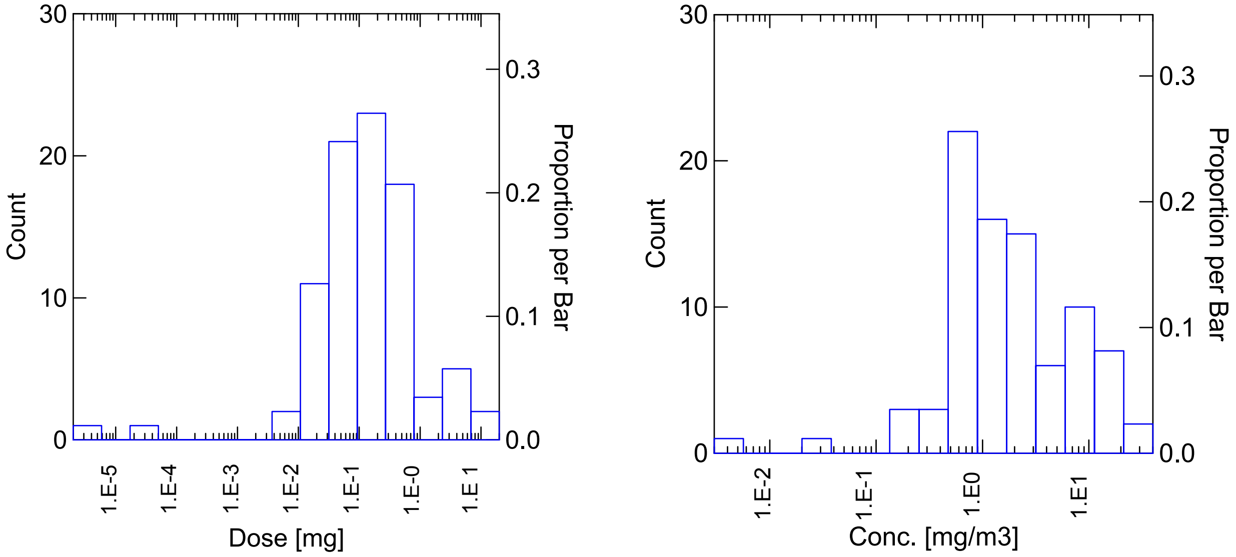


Figure 7. Assessed doses and maximal concentrations expressed in [mg] and [mg/m^3] of respirable aerosols

Because of the trivial exposure model considered and the conservative assumptions made, only a limited confidence should be given to the absolute numbers. Still, their relative ranking is of utmost interest. The exposure levels obtained indicate that the respirable mists from the waterproofing sprays have a very low OEL (Observable Effect Level). Adverse effects may obviously occur even at exposure or dose levels corresponding to well ventilated spaces, or very short exposure times. Considering the products involved are widely marketed and only a small fraction of users reported troubles, these results suggest that high response variability exists between exposed individuals.

This variability may be caused by individual factors amongst the spray users such as physiological, or metabolic differences. It should also be noted that the reported effects are presumably not of allergic nature. Another cause of variability is the presence of external factors related to exposure conditions. A typical example of this is the case of exposure to Teflon fumes (24), where the presence of the toxic product is triggered by a heat source. However, in this study, no heat source in the vicinity of the spraying activity was reported and smoking during or shortly after spraying was reported in only 10 out of the 102 cases.

Exposure vs. perceived effects

Subjective indicators of exposure effects have been compared to exposure levels for possible correlations. These comparisons are summarized in Table 2. No significant relationship was found with the dose or the maximal concentration obtained during the retrospective assessment. These results suggest that factors other than exposure to overspray mist play a determining role in the occurrence of adverse health effects. The relationship between the parameters of basic exposure conditions (amount of product, spraying time) and the perceived effects are poor. A statistically significant correlation was found between the perceived symptoms (SCORE) and these parameters, although calculation of the regression coefficients (0.017 for spraying time and 0.001 for amount of product) indicated that the contribution of exposure conditions on symptoms occurrence was limited. Besides, the perceived effect indicators should be considered carefully because they rely heavily on subjective perception.

Exposure vs. objective clinical effects

The objective clinical indicators collected in the physician's questionnaires were compared to the assessed exposure indicators and exposure conditions. Clinical objective indicators are expressed as continuous variables, which can be more conveniently compared to the continuous exposure variables. The drawback is that such clinical investigations have been conducted only for a fraction of cases (about one third), probably the most severe ones, which requested medical attention. A summary of the results obtained is given in Table 3.

Table II: Perceived severity vs. exposure conditions (DELAY= delay before medical care, DYSP= dyspnea score, SCORE= symptom score).

Spearman Correlation Coefficients (Prob>|r| under H0: Rho=0)

	Dose	**Cmax**	**Spraying Time**	**Amount of Product**
DELAY	-0.151 (0.328)	-0.175 (0.255)	-0.103 (0.475)	0.107 (0.476)
DYSP.	0.175 (0.373)	0.261 (0.179)	0.139 (0.448)	0.037 (0.848)
SCORE	0.216 (0.059)	0.159 (0.168)	**0.255 (0.014)**	**0.288 (0.009)**

Table III: Correlations between exposure conditions and clinical indicators (WBC=white blood cell count; CRP= C-reactive protein, PaO_2 = partial oxygen pressure).

Correlation Coefficients (Prob>|r| under H0: Rho=0)

	Dose	**Cmax**	**Spraying Time**	**Amount of Product**
WBC*	0.328 (0.102)	**0.404 (0.040)**	0.079 (0.696)	-0.162 (0.439)
CRP*	0.075 (0.699)	**0.375 (0.045)**	-0.140 (0.445)	-0.017 (0.928)
PaO_2**	0.021 (0.927)	0.018 (0.938)	**0.440 (0.031)**	0.389 (0.074)

* = Spearman * *= Pearson

No significant correlation was found between any of the clinical indicators and the assessed doses, which seems to exclude any direct dose-response relationships. These results are supported by the lack of correlation between the clinical indicators and the amount of product used, which is also an indicator, although quite rough, of the potential dose. The fact that the same tendencies were observed for predicted values (concentration, dose) and basic parameters (amount of spray used) is also comforting when considering the possible influence of the model lack of sensitivity on the results obtained.

The relationships found for the maximal concentration and the spraying time are less obvious and must be considered in a more detailed way. Weak but significantly positive correlations were found between the non-specific inflammatory markers WBC and CRP and the maximal exposure concentrations Cmax. The detailed results are presented in Table III and Figure 8.

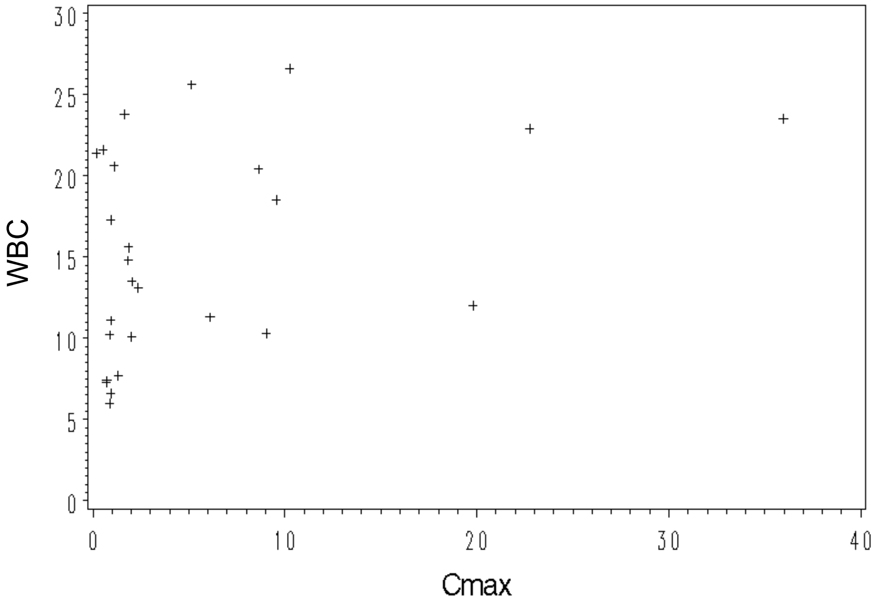


Figure 8. Relationship between Cmax and indirect inflammatory markers - WBC

They show that WBC levels tended to be directly correlated with Cmax (WBC = 13.6846 + 0.2926 Cmax, R2 = 0.15, Pearson = 0.0533, Spearman = 0.0404), although no similar trend could be observed for the C-reactive protein levels. A significant correlation was also found between the spraying time and the pulmonary gas exchange marker PaO_2 (Table III).

Surprisingly, the relationship was positive; longer spraying times were correlated with higher PaO_2 (Figure 9), that is, better pulmonary gas exchange, whereas the opposite would have been expected. Since the spraying time plays a major role in exposure, this unexpected relationship further suggests that no straightforward mechanism exists between the observed health effects and the exposure levels to respirable particles. This lack of direct relationship is also apparent when considering the lack of correlation between dose vs. PaO_2 levels (Table III). The PaO_2 levels appear to be highly variable, particularly in the lowest dose range.

At low doses, the dose-PaO_2 response is clearly indiscriminate, with a high variability in the PaO_2 levels obtained.

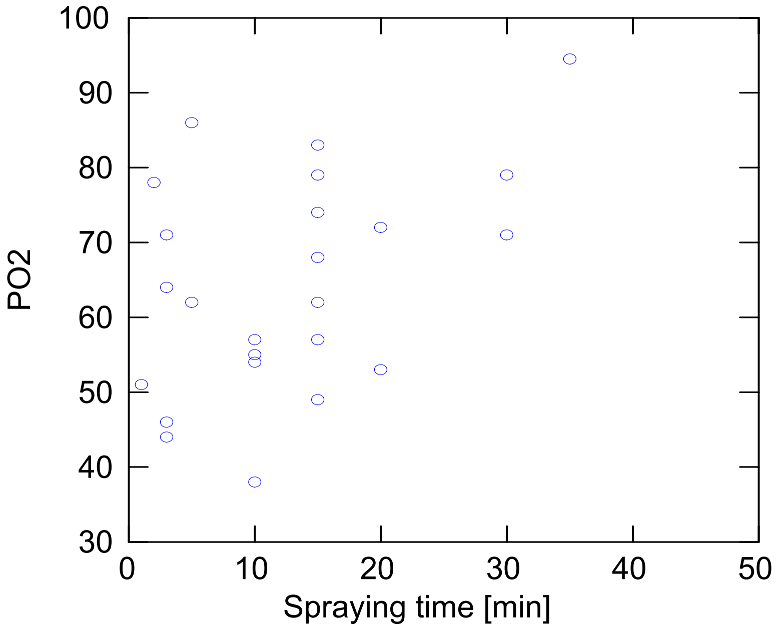


Figure 9. Relationship between PO_2 and spraying time.

Subcategories regarding smoking history, allergy, and asthma or COPD were investigated, to determine whether individual susceptibility could explain the occurrence of toxicity features at very low exposure levels. No statistically significant differences were found within these subgroups concerning Cmax, Dose, and spraying time (Wilcoxon-Mann-Whitney Test) (Table IV). It must however be mentioned that the number of cases with objective clinical indicators is reduced, and it is therefore difficult to get clear evidence or to analyse subcategories in a consistent way. This is particularly true when considering subcategories related to the exposure environment, for which limitations of the two-compartment model used to assess doses and concentrations may play a significant role. Compartmental models are known to give rough estimates of real exposure conditions. When these models are used to make relative comparisons between exposures occurring in the same kind of environment, this drawback is mitigated. However, more model limitations are to be expected when comparing exposure conditions of varied nature (i.e outdoor v. indoor).

Table IV : Comparisons Between Subgroups for Smoking, Asthma, COPD, and Allergies

	Mean	**p-value**
Dose		
Smoking		
Yes	0.51	0.37
No	0.80	
Allergy		
Yes	0.26	0.74
No	0.55	
Asthma, COPD		
Yes	0.12	0.32
No	0.50	
Cmax		
Smoking		
Yes	4.39	0.3
No	4.65	
Allergy		
Yes	2.65	0.79
No	4.53	
Asthma, COPD		
Yes	1.75	0.07
No	4.25	
Spraying time		
Smoking		
Yes	11.26	0.49
No	12.55	
Allergy		
Yes	9.87	0.84
No	12.07	
Asthma, COPD		
Yes	12.78	0.45
No	10.81	

Conclusions & recommendations

The acute respiratory syndrome associated with the 2002-2003 Swiss outbreak occurred in a wide array of exposure conditions, ranging from short to extensive spraying and in poorly ventilated rooms to open spaces. The resulting assessed dose and exposure levels obtained were spread on large scales of several orders of magnitude. The lack of dose-response correlation with both perceived severity and clinical indicators suggests that 1) it is not possible to define a threshold dose below which the incriminated sprays could be safely used, and 2) some indirect or complex mechanism(s) predominated in the occurrence of the respiratory disease. The occurrence of adverse effects is driven by other factors than the sole amount of respirable particles, such as: metabolic differences, interaction between particles and other chemicals agent (e.g. residues from the solvent) or even the presence of nanoparticles. The solvent alone could be ruled out as the cause of toxicity because the particles reaching the alveoli are essentially made of non-volatile material (16). It must be pointed out that neither environmental factors (heat source due to smoking), nor individual susceptibility (such as a pre-existing lung disease, allergy or smoking) were found to explain this high response variability.

For these reasons, and because of the vast array of spraying situations observed, it is unlikely that a simple improvement of the exposure conditions may have prevented the occurrence of the toxicity outbreak. Thus, enforcing the compliance with the basic safety measures, such as spraying in a well-ventilated space, is obviously not sufficient in this case. Besides, commercial products intended for domestic applications must be usable without respiratory protective equipment. A more efficient prevention should have taken place prior to the product marketing and distribution. It is interesting to note that the product toxicity has been tested according to German standards prior to marketing. To our knowledge, the effects of 4-5 μm aerosols droplets were tested on rats at a high exposure concentration. However, tests conducted in such a narrow range may not have appropriately reflected the possible human health effects at the pulmonary alveolar level. It is well established that the morphological differences between rats and human affect both inhalation and deposition patterns. Moreover, retention and clearance patterns have also shown to be species-dependant (25,26). Smaller particle size (around 0.1 μm) would have been more appropriate to assess alveolar toxicity. Finally, alveolar inflammation and impairment of gas exchange could have taken place in rats having inhaled the product, but remained undetected if only animal survival was considered as an outcome, and if appropriate analyses of lung function and inflammation were not performed.

Additionally, the preventive strategy should take into account the full range of particle size, which could be generated by various pressurization devices. Hence, the same waterproofing agent can be marketed in various mixtures and conditioning for a broad range of applications. A change in the product physico-chemical properties or in the spraying can design (especially the nebulization system) may have an important impact on the distribution of particle size.

In summary, we believe that new outbreaks of waterproofing spray toxicity may occur if a particular combination of fluororesin and triggering factors (solvents, nebulization system) appears in a marketed product. The potential toxicity of such a product is likely to remain undetected in the pre-marketing phase if new preventive strategies are not applied. Although they may reduce the inhaled dose, written warnings on product packages are probably insufficient to prevent the toxicity because of the apparent lack of a safe threshold dose. We therefore suggest that: 1) new waterproofing agents should be bench-tested in the final mixture in which they are intended to be marketed, 2) a wide range of distribution of particle size should be considered for testing in order to encompass interspecies differences as well as the various conditioning in which the product is intended to be marketed, and 3) animal toxicity experiments should assess sensitive markers of pulmonary function and inflammation.

Acknowledgements

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List of symbols

C	Mass concentration $[g/m^3]$ $[mg/m^3]$ or $[\mu g/m^3]$
	C0 incoming conc., CFF far-field conc., CNF near-field conc.
E	Emission rate [g/s] [mg/s] or [μg/s]
Q	Volumic flow $[m^3/s]$
	Qe inter-compartment flow
t	time [s]
V	Volume $[m^3]$

References

1. Grainger, D., and C. Stewart: Fluorinated coating and Films: Motivation and significance. In: Fluorinated Surfaces, Coatings and Films, D.G. Caster and D.W. Grainger, eds., pp. 1-14. (ACS symposium series; 787). American Chemical Society, Washington (2001).
2. Muller-Esch, G., E. Brunk, H. Djonlagic, et al.: Pulmonary Effect of Inhaling Leather Impregnation Sprays. Dtsch. Med. Wochenschr. 107(18): 692-695 (1982).
3. Okonek, S., H.J. Reinecke, W. Fabricius, and K. Preussner: Poisoning with Leather-Impregnation Sprays. A Retrospective Analysis of 224 Cases of Poisoning. Dtsch. Med. Wochenschr. 108(49): 1863-1867 (1983).
4. Tanino, M., K. Kamishima, K. Miyamoto, et al.: Acute Respiratory Failure Caused by Inhalation of Waterproofing Spray Fumes. Nihon Kokyuki Gakkai Zasshi 37(12): 983-986 (1999).

5. Smilkstein, M.J., B.T. Burton, W. Keene, et al.: Acute Respiratory Illness Linked to Use of Aerosol Leather Conditioner. Morb. Mortal Wkly. Rep. 41(52-53): 965-967 (1993).
6. Burkhart, K.K., A. Britt, G. Petrini, et al.: Pulmonary Toxicity Following Exposure to an Aerosolized Leather Protector. J. Toxicol. Clin. Toxicol. 34(1): 21-24 (1996).
7. Tagawa, A., K. Ikehara, T. Tsuburai, et al.: Acute lung injury caused by inhalation of waterproofing spray. Nihon Kokyuki Gakkai Zasshi 41(2):123-6 (2003).
8. Yamashita, M., and J. Tanaka: Pulmonary Collapse and Pneumonia due to Inhalation of a Waterproofing Aerosol in Female CD-1 mice. J. Toxicol. Clin. Toxicol. 33(16): 631-637 (1995).
9. Hubbs, A.F., V. Castranova, J.Y.C. Ma, et al.: Acute Lung Injury Induced by a Commercial Leather Conditioner. Toxicol. Appl. Pharmacol. 143: 37-46 (1997).
10. "Sprays imperméabilisants pour le cuir et les textiles: fréquence inhabituelle de troubles respiratoires. "[online] available at http://www.bag.admin.ch/chemikal/gesund/f/impraeg.htm (accessed January 28, 2005).
11. Kupferschmidt, H., E. Namer, R. Lazor, et al.: Acute respiratory toxicity after use of waterproofing textile and leather sprays: patient and exposure data [abstract]. Swiss Med Wkly. 134 (20): 50 (2004).
12. Bonte, F., A. Rudolphus, K.Y. Tan, and J.G. Aerts: Severe respiratory symptoms following the use of waterproofing sprays. Ned Tijdschr Geneeskd 147(24):1185-8 (2003).
13. Testud, F.: Toxicité des imperméabilisants pour le cuir et les tissus. VIGItox, journal du centre antipoison de Lyon. 24 :1-2 (2004).

14. Tizzard, Z., and J. Edwards: "Acute respiratory effects following use of waterproofing sprays: Some UK experience" Thorax Online, 25 Oct 2004. http://thorax.bmjjournals.com/cgi/eletters/59/6/541-a#331 (accessed February 14, 2005).

15. Lazor-Blanchet, C., S. Rusca, D. Vernez, et al.: Acute Pulmonary Toxicity Following Occupational Exposure to a Floor Stain-protector. Int. Arch. Occup. Environ. Health 77 (11): 244-248 (2004).

16. Vernez, D., P.-O. Droz, C. Lazor-Blanchet, and S. Jacques: Characterizing Emission and Breathing-zone Concentrations Following exposure cases to fluororesin-based waterproofing spray mists. J. of Occup. and Environ. Hyg. 1(9):582-592 (2004).

17. Yamashita, M., and J. Tanaka: Pulmonary Collapse and pneumonia due to inhalation of waterproofing aerosols in female CD-1 mice. Clin. Toxicol. 33: 631-7 (1995).

18. Rask-Andersen, A.: Inhalation Fever. In: Occupational and Environmental Respiratory Disease, pp. 243-258. P. Harber, M. B. Schenker and J. R. Balmes. Ed. Mosby-Year Book, Inc. St. Louis, Missouri (1996).

19. Flynn, M.R., B.L. Gatano, J.L. McKernan, et al.: Modelling Breathing-zone Concentrations of Airborne Contaminants Generated During Compressed Air Spray Painting. Ann. Occup. Hyg. 43(1): 67-76 (1999).

20. Tan, Y.M., and M.R. Flynn: Methods for Estimating the Transfer Efficiency of a Compressed air Spray Gun. Appl. Occup. Environ. Hyg. 17(1): 39-46 (2002).

21. Keil, C.B. Ed.: Mathematical Models for Estimating Occupational Exposure to Chemicals. American Industrial Hygiene Association (AIHA), Fairfax (2000).

22. Cherrie, J.W.: The Effect of Room Size and General Ventilation on the Relationship Between Near and Far-Field Concentrations. Appl. Occup. Environ. Hyg. 14(8): 539-546 (1999).

23. Keil, C.B.: A Tiered Approach to Deterministic Models for Indoor Air Exposures. Appl. Occup. Environ. Hyg. 15 (1): 145-155 (2000).

24. Albrecht, W.N., and C.J. Bryant: Polymer-fume fever associated with smoking and use of a mold-release spray containing PTFE. J. Occup. Med. 29: 817-9 (1987).

25. Snipes, M.B., R.O. McClellan, J.L. Mauderly, R.K.Wolff: Retention patterns for inhaled particles in the lung : comparisons between laboratory animals and humans for chronic exposure. Health Phys. 57(1): 69-78 (1989).

26. Asgharian, B., R. Wood, and R.B. Schlesinger: Empirical modeling of particles deposition in the alveolar region of the lungs: A basis for interspecies extrapolation. Fundam. and appl. Toxicol. 27: 232-238 (1995).

Paper III

Exposure models in Switzerland. An overview of the present situation

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exposure model, toxicokinetic model, database, questionnaire, methoxypropanol,

Abstract

A survey was undertaken among Swiss occupational health and safety specialists in 2004 to identify uses, difficulties and possible developments of exposure models. Occupational hygienists (121), but also occupational physicians (169) and safety specialists (95), were surveyed with an in depth questionnaire. Results obtained indicate that models are not used very much in practice in Switzerland, and are reserved to research groups focusing on specific topics. However various determinants of exposure are often considered important by professionals (emission rate, work activity), and in some cases recorded and used (room parameters, operator activity). These parameters cannot be directly included in present models. Nevertheless, more than half of the occupational hygienists think that it is important to develop quantitative exposure models.

Looking at research institutions, there is however a big interest in the use of models to solve problems which are difficult to address with direct measurements; i.e. retrospective exposure assessment for specific clinical cases, and prospective evaluation for new situations or estimation of the effect of selected parameters. In a recent study about cases of acute pulmonary toxicity following water proofing spray exposure, exposure models have been used to reconstruct exposure of a group of patients. Other ongoing studies include: (1) looking at combining expert judgment of exposure levels and direct measurements, including empirical and physical models, (2) improving the description of the near field dispersion, (3) making models based on parameters more accessible to practionners, (4) developing tools to describe emission.

Finally in the context of exposure prediction, it is also important to report about ongoing efforts in the area of exposure databases. Such a measurement database exists now in

Switzerland since 1991. It does not at present cover all measurements taken in Switzerland, but it can however serve experts in their prediction of exposure in the absence of direct measurement. The (small) database is accessible via internet (http://www.iurst.ch/ist-bin/ist_nuisances_db.pl).

Zusammenfassung

Eine im Jahr 2004 unter schweizerischen Gesundheits- und Sicherheitsspezialisten durchgeführte Umfrage hatte zum Ziel, Gebrauch, Schwierigkeiten sowie Entwicklungsmöglichkeiten von Expositionsmodellen zu erfassen. Insgesamt 121 Arbeitshygieniker, 169 Arbeitsmediziner und 95 Sicherheitsspezialisten beantworteten einen detaillierten Fragebogen. Vorläufige Resultate weisen darauf hin, dass Modell selten im Alltag zur Anwendung kommen, sondern mehrheitlich von Forschungsgruppen für spezifische Fragestellungen benutzt werden.

Die Expositionsdeterminanten Arbeitsaktivität und Emissionsrate wurden von den Spezialisten als die wichtigsten beeinfussenden Parameter betrachtet. Zudem wurden Anwenderaktivität und Raumparameter aufgrund ihrer einfachen Verfügbarkeit regelmässig aufgezeichnet.

Diese Parameter können in heutigen Modellen nicht direkt mit einbezogen werden. Über die Hälfte der Arbeitshygieniker war der Meinung, dass es wichtig sei, quantitative Expositionsmodelle zu entwickeln.

Bei Forschungsinstituten bestand eine grosses Interesse an Modellen, um Fragen zu lösen, welche mit direkten Messungen schwierig zu beantworten sind, wie etwa retrospektive Expositionsbestimmungen für spezielle klinische Fälle, prospektive Bewertung neuer Situationen oder zur Abschätzung des Einflusses ausgewählter Parameter. In einer kürzlich durchgeführten Studie über Fälle von akuter Lungenschädigung nach der Verwendung von Imprägnierspray wurden Expositionsmodelle verwendet, um die Exposition einer Gruppe von Patienten zu rekonstruieren. Andere aktuelle Studien betreffen: (1) die Kombination von Expertenurteilen und direkten Messungen unter Berücksichtigung empirischer und physikalischer Modelle, (2) eine verbesserte Beschreibung der Nahfeldverteilung, (3) die

Definition von Modellen, die auf für Praktiker besser zugänglichen Parametern beruhen, (4) die Entwicklung von Methoden zur Beschreibung von Emissionen.

Im Zusammenhang mit der Expositionsvorhersage sollten noch die Anstrengungen im Bereich der Expositionsdatenbanken erwähnt werden. Seit 1991 besteht in der Schweiz eine solche Datenbank. Obschon sie nicht alle in der Schweiz durchgeführten Messungen abdeckt, kann sie dennoch Experten dabei helfen, Expositionen in der Abwesenheit von direkten Messungen abzuschätzen. Auf die (noch kleine) Datenbank kann mittels Internet zugegriffen werden (http://www.iurst.ch/ist-bin/ist_nuisances).

Introduction

This presentation intends to cover several aspects of exposure modelling in Switzerland. It will first concentrate on a recent survey among Swiss occupational health specialists about their involvement in exposure modelling activities. Some specific research and development will then be presented. Finally future developments in Switzerland will be discussed.

Exposure models among Swiss occupational hygienists

A survey was undertaken among Swiss occupational health and safety specialists in 2004 to identify uses, difficulties and possible developments of exposure models (1) . A questionnaire was prepared to cover the following points:

- OHS background and activities
- exposure assessment techniques used
- modalities used for expert judgments
- identification of the main exposure determinants used in expert judgments
- identification of the models used by practitioners

The questionnaire was addressed by mail to 121 occupational hygienists, members of the Swiss Occupational Hygiene Society. A shorter questionnaire was also sent to a sample of occupational physicians (169) and safety specialists (95). Results obtained from occupational hygienists are reported here.

The survey among occupational hygienists had a response rate of 64%. Most occupational hygienists surveyed in Switzerland have a training in chemistry and/or are involved in some form in the chemical industry. They are therefore clearly interested and concerned by chemical exposure modelling. Considering their occupational duties, Figure 1 indicates that exposure assessment is not in most cases their dominant activity, either because they are responsible for other domains (environment, quality, production…), or because other occupational hygiene activities are also important (management, training, …). In fact less then 10% report doing exposure assessment daily.

When asked about the way they perceive the reliability or the efficiency of different exposure assessment methods presented to them, hygienists favour clearly measurements (long or short term air sampling, biological monitoring). Exposure modelling comes last with an average score of 2.0 on scale of 6. Even expert judgment via rapid site visits is judged more reliable with an average score of 2.8. As a consequence exposure models are not used very much by practitioners. It comes at the last rank of exposure methods proposed by the questionnaire. When asked for reasons, occupational hygienists declare in 40% of the cases that models are difficult to apply in specific practical cases, that it is too time consuming to use them (22%), that they are not accurate/precise enough (22%). For 16% of them, they do not know enough about models to apply them (Figure 2).

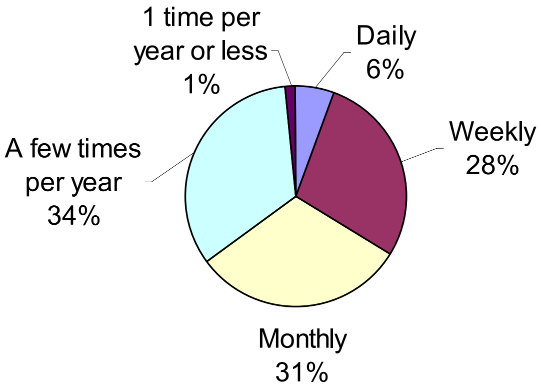


Figure 1: Frequency of exposure assessment activities among occupational hygienists

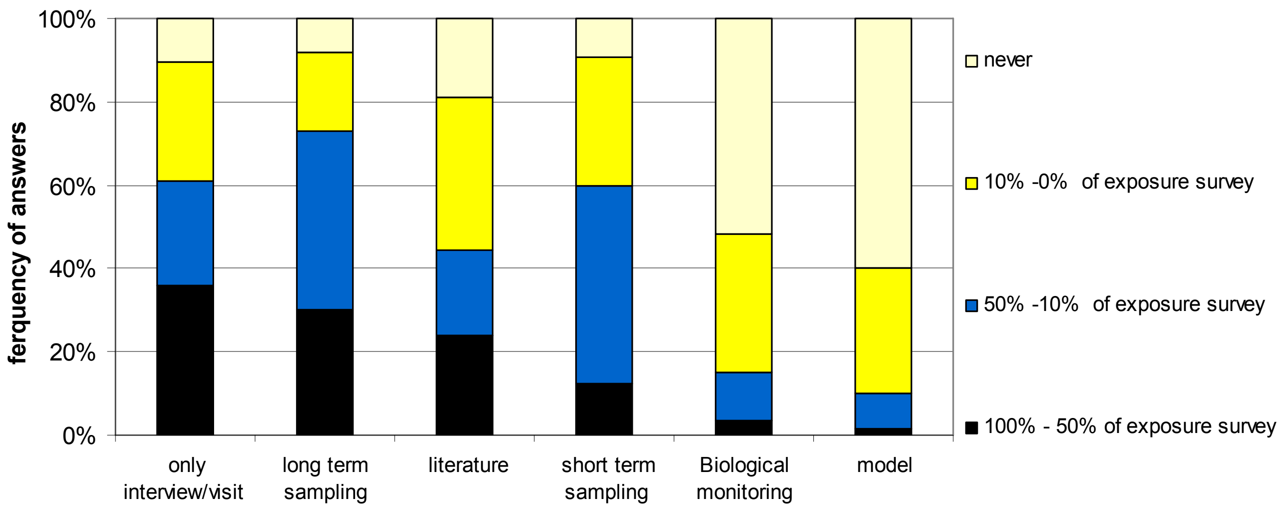


Figure 2: Frequency of use of the different exposure assessment techniques

One of the key parameters in the use of exposure models is the emission rate of the pollutant. Most of the surveyed persons applying models (coming from research institutions) responded that they use mass balance, which in fact is only applicable to a limited number of cases. Other approaches consisted in measuring the contaminant in the exhaust ventilation, using literature data or applying specific emission models. For the contaminant dispersion in the workplace, one- and two-compartment models, as well as EASE model are the most widely used tools.

In any case overall very few hygienists know and use exposure models. This is probably linked to the fact that they do not really know about these techniques. But an other reason is the perceived low efficiency and reliability of the existing models. Generally they consider that models are either too simple to trust the results they produce or too complicated and sophisticated to be applied simply in specific practical situations. This is clearly the case of computer fluid dynamic modelling, which is considered to be very inefficient (too many parameters thus too complicated and time consuming to use) and reliable (it takes into account a wide range of physical phenomenon and produced detailed data). .

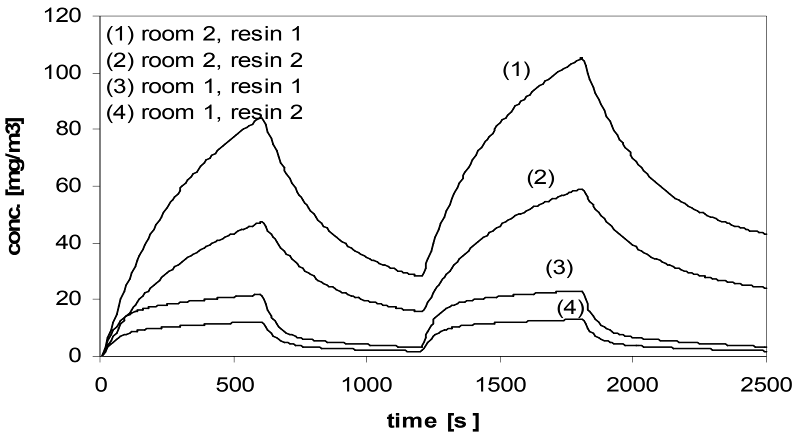
As a consequence, almost 70% of hygienists declare that exposure models should be improved to integrate factors more easily accessible to practitioners. 50% consider also that near field local phenomena are important for operator estimation and that they should be described in more details. Finally they recommend that models for emission estimation should be developed.

Examples of models in a research institution

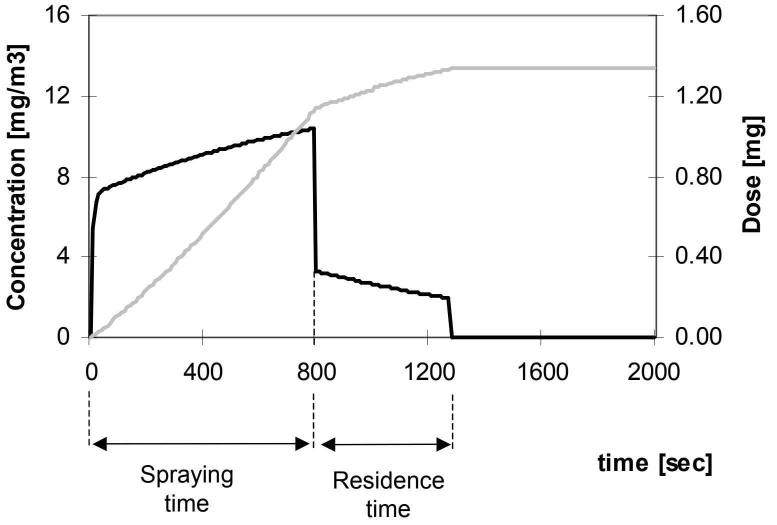
Despite this low overall usage of exposure models by practitioners in Switzerland, there is an interest in research institution to apply and develop new techniques.

Waterproofing sprays

As an example exposure models have recently been applied to a specific outbreak of lung diseases among people using waterproofing sprays in various situations (1). This occurred after a change in the formulation of the commercial products. It was decided to retrospectively estimate the exposures to both products for each medical case. An experimental set up was designed to enable an estimation of emission rates to be made under various conditions (product, spray nozzle, work rate…). Questionnaires were also sent to exposed individuals in order to assess the exposure environmental conditions during spraying (room size, spraying time, ventilation..). Measured emission rates were then fed into a two-compartment exposure model (Figure 3 a). This allowed (1) the prediction of exposure indices for each medical case to perform an epidemiological approach of the outbreak, and (2) the analysis of different scenarios through sensitivity analysis (Figure 3 b).



(a) exposure profiles for different scenarios



(b) typical individual exposure profile and dose calculation

Figure 3: Typical concentration and dose profile obtained from simulation.

Propylene glycol monomethyl ether (PGME)

Another example is represented by the case of propylene glycol monomethyl ether (PGME) exposure in various occupations including water-based products. A study was undertaken in Switzerland to predict potential exposures in various workplaces. These were identified based on the Swiss chemical register database (4). The activities were then simplified to 2 types of emission processes: evaporation from a surface, and evaporation during liquid transfer. These were simulated experimentally in an exposure chamber to measure "standard" emission rates. Using a two-compartment model, both operators' and bystanders' exposures were predicted for the various identified activities. This made it possible to predict ranges of potential exposures, and also to compare different activities as far as their risks are concerned (Figure 4, Figure 5). Exposures predicted by the model can be compared to average measured exposures reported in several studies (Figure 6) (5). Operator exposures ranged from less then 10 ppm up to more then 400 ppm, PGME's TLV being 100 ppm.

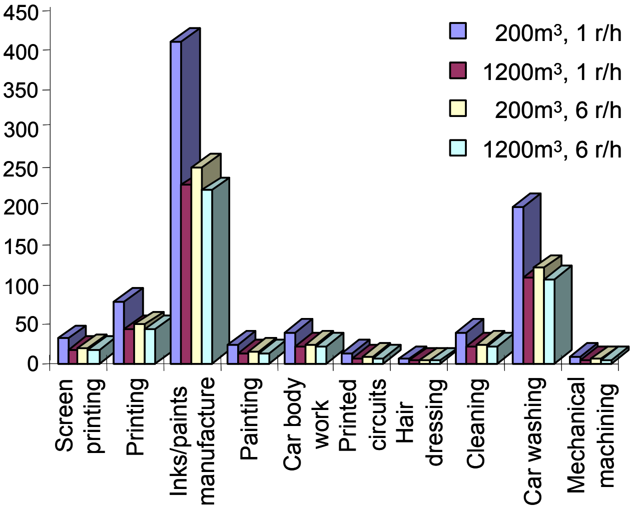


Figure 4 : Operator exposure to PGME

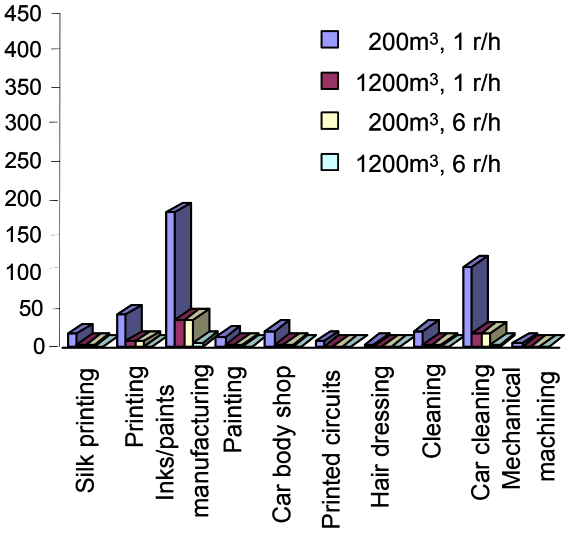


Figure 5: Indirect exposure to PG

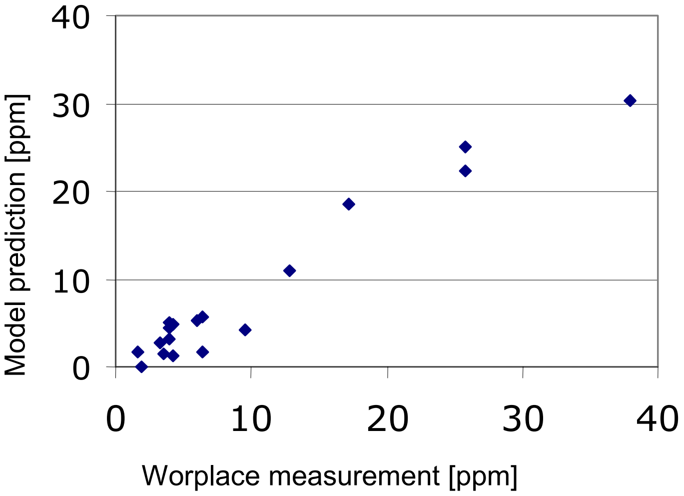


Figure 6 : Model predictions using experimental emission rates compared to field measurements in various workplaces, such as printing, ink manufacture, cleaning operations

Toxicokinetic modelling

In the same study toxicokinetic modelling allowed prediction of equivalent biological levels for the identified occupational activities, concerning specifically methoxypropanol and its metabolite methoxyprópronic acid in urine (Table 1) (5). This could serve as exposure indicators or the estimate corresponding risks using dose-response relationships. There is in Switzerland a significant interest and experience in different types of toxicokinetic modelling applied to occupational health (6).

	Air exposure	Urinary levels	
Reference level Activity	***100 ppm***	***PGME***	***2-MPA***
		300 µmol/l	***130 µmol/l***
Printing	43-80	135-250	40-75
inks/paint mfg	220-410	690-1280	210-390
car washing	106-199	330-620	100-190

Table 1: PGME and 2-MPA biological levels simulated with a toxicokinetic model associated to exposure concentrations for some activities.

Exposure registry

Lastly it is worthwhile to report on developments concerning an exposure data base in Switzerland. This could also serve as a modelling tool for exposure assessment in new, or unmeasured situations. Since 1991 a register of exposure measurements is maintained at the Institute of Occupational Health Sciences in Lausanne. It contains now close to 10’000 measurements associated with information on the economic sector, the profession, the conditions of measurement and analysis (7). These can be used to describe new exposure situations in similar cases. At the beginning considered as the Institute’s database, it is now open to anyone in Switzerland who wants to share his results. It is accessible via internet and can thus be consulted with free access by Swiss occupational hygienists.

Discussion

We can anticipate several developments in the near future in Switzerland. Two projects have started in our Institute concerning exposure modelling. On the one hand a doctoral thesis is currently carried out to improve existing modelling techniques, mainly looking at the emission side for various types of pollutants, and at the conditions in the near field region which are determinant for the operator's exposure.

On the other hand, we are currently discussing possibilities to combine exposure measurements with information on the exposure determinants in a Bayesian framework to improve strategies in exposure assessment. In a first step exposure determinants will be collected retrospectively for existing measurement. These will then be used in conjunction with future measurements to improve our decision making.

In conclusion, exposure models are not used very much in practice in Switzerland, and their application in practice is limited to a few motivated occupational hygienists. Looking at research institutions, there is however a big interest in the use of models to solve problems which are difficult to address with direct measurements; ie retrospective exposure assessment for specific clinical cases, and prospective evaluation for new situations or estimation of the effect of selected parameters.

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References

1. Bruzzi R., Vernez D., P.-O. Droz: Beliefs and practices in the assessment of workplace pollutants. Submitted for publication in Sozial - und Präventivmedizin (December 2004).
2. Vernez, D., P.-O. Droz, C. Lazor-Blanchet, and S. Jacques: Characterizing Emission and Breathing-zone Concentrations Following exposure cases to fluororesin-based waterproofing spray mists. J. of Occup. and Environ. Hyg. 1(9):582-592 (2004).
3. Vernez D., Bruzzi R., Kupferschmidt H., De Batz A., Droz P.-O., Lazor R.: acute respiratory syndrome after inhalation of waterproofing sprays : a posteriori exposure-response assessment in 102 cases. Submitted for publication in J. of Occup. and Environ. Hyg. (February 2005).
4. Dentan A., Devanthéry A., de Peyer J.E., Droz P.-O.: Propylene glycol monomenthyl ether (PGME) occupational exposure 2. Identification of products containing PGME, their importance and their use in Switzerland. Int. Arch. Occup. Environ. Health 73: 349-351 (2000).
5. Devanthéry A., Toxico-cinétique et surveillance biologique du propylène glycol monométhyl éther. Etude sur des volontaires. EPFL thèse no 2764 (20003)
6. Pierrehumbert G., Droz P.-O., Tardif R., Charest-Terdif G., Truchon G.: Impact of human variability on the biological monitoring of exposure to toluene, phenol, lead and mercury: II. Compartmental based toxicokinetic modelling. Toxicology letters 134: 165-175 (2002).

7. Vernez D., Charrière N., Droz P.-O., Favre O.: Une base de données des mesures d’exposition en milieu professionnel. Revue de santé et sécurité au travail IZA, 6 (2002).

Paper IV

Pertinence of a two-zone model for occupational exposure assessment

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Keywords:

exposure modelling, airflow pattern, exposure assessment, near field, experimental measurements

Abstract

Several authors have proposed different types of exposure models. The two-zone model is often preferred and applied, both for retrospective and prospective exposure estimations. The purpose of this paper is to scrutinize the notion of near field and its definition as well as the pertinence of the two-zone model, through theoretical aspects, experimental investigations, statistical analysis and computational fluid dynamic simulations.

Theoretical considerations based on mass diffusion and convection in a ventilated room show that concentrations in the two zones correspond to average concentrations. As a consequence a local sensor cannot directly measure these concentrations. Simultaneous measurements were performed in a 10 m^3-ventilated booth under different ventilation conditions. 64 measurement points were observed simultaneously. Two irregular shape fields of "similar" concentrations were observed within the room. These geometrical patterns appeared to be strongly influenced by ventilation rates. This segregation was confirmed by a statistical analysis (Kernel density function, Silverman test). Based on two non-spherical compartments, we were able to prove a good agreement between the experimental measurements and the predictions of the two-zone model.

These results indicate that, (1) from a statistical point of view, the compartmental theory makes sense and (2) simple geometrical shapes, such as the half-sphere commonly used in the two-zone model, are not suitable to depict near field zones.

Introduction

Estimating exposure is a crucial aspect in occupational health studies. A physical model may represent a transparent method for exposure assessment, both retrospective (1) and prospective. However, when applying such a deterministic model in exposure assessments, it is of critical importance to understand the influence of exposure determinants such as pollutant generation rate and transport. In certain cases, the complexity of these estimations could be a serious obstacle to using exposure models. Indeed, a recent survey among Swiss occupational hygienist (2,3) demonstrated that only limited confidence was given to exposure models, because of the difficulties in integrating accessible exposure determinants in term of model parameters. A thorough understanding of the models' theoretical grounds, their strengths and weaknesses, is also crucial in order to select the appropriate model for each specific circumstance. Keil and Murphy (4) described a tiered approach for model selection, considering the input available and the complexity needed to have results with an acceptable degree of uncertainty.

One of the most important differences between current physical models is the definition of pollutant dispersion in the room. Contaminant dispersion phenomena within rooms are influenced by several variables such as room geometry, thermal effects, direction of main airflow, presence of a worker and even by arms movements (5-9). Experiments have also been carried out to understand the influence on breathing zone concentrations of the worker's position and the source of contaminant with respect to the airflow directions (10,11).

Dispersion phenomena may be very complex. In practice, however, relatively coarse models based on a wide range of assumptions and/or simplifications may be used (12). The Ideal Mixed Model, for instance, relies on the concept of mass conservation and of a complete instantaneous mixing throughout a single workplace volume (13). This model does not provide information about the spatial dispersion of air contaminants but may nevertheless represent a practical approach in some particular exposure and ventilation conditions.

Alternatively, multi-compartmental model may be used to account for non-homogeneous situations. Multi-compartmental models split the room in a series of conceptual well-mixing zones, connected with a volumetric flow rate across each boundaries (14,15). The concept can be extended to as many compartments as a specialist may judge necessary, but the complexity of the model increases with the number of zones selected. Furthermore, accuracy is compromised by the difficulty in quantifying the exchange rate for each compartment (16).

Other authors (17,18) cut down the problem by focusing on a classical industrial situation where the volume concerned with exposure could be divided in just two conceptual compartments (two-zone model), one near the source (near field) containing the worker's breathing zone when working near the source, and the other represented by the remaining volume (far field). Different studies (19), undertaken to compare personal sampling with general sampling, have shown that personal exposures are generally higher than general exposures. This effect was also demonstrated by Furtaw et al. (20) who employed a two-compartment model, called the source-proximate effect (SPE) model, to fit data from measured concentrations at various distances from the source. Still, Cherrie (21) reviewed data about personal and area concentrations for 40 different working situations and found that 80% of the personal measurements exceeded the respective environmental measurements. This model, simplifying spatial variability of concentration into just two compartments, may represent a useful tool in the occupational hygiene practice, which tends indeed to focus exposure assessment on two kinds of situations, individual and ambient exposures (other workers within the same room).

However, some questions arise about the pertinence of dividing the exposure zone into two compartments. In case of a directional source or in presence of a reverse flow region, a widespread concentration gradient may occur and the assumption of an ideal mixed space near the source could represent a bad approximation. Some experiments (22) were carried out to establish the influence of worker's presence on the contaminant dispersion in the near field. These studies demonstrated that a reverse flow zone, produced in front of a worker, might cause high contaminant concentrations in the breathing zone.

Another current drawback of this model is the need to develop criteria for compartment definition, in terms of near field extension and shape. Some authors suggested various practical configurations of a near field. For instance, for the near field volume, Keil (23) conceptualized a hemisphere with a radius equal to the distance between the source and the human receptor. Another configuration is offered by Nicas (17), who divided the room in an upper ventilated zone and a lower zone of occupancy. This case implies a particular ventilation scenario, where both the supplied ventilation air and the room air exhaust systems are near the ceiling. Hemeon, quoted in Burton (24), discussed various geometries for the near field, depending on the particular work operation involved. An example is found in Nicas et al. (25) who employed a compartment with a rectangular base of the same area as the wash basin used (the emission source), while the height coincided with the vertical distance between the wash basin and the breathing level of the worker.

The purpose of this paper is to examine the notion of the near field definition and the pertinence of the two-zone model through theoretical considerations, practical experiences, statistical analysis and computational fluid dynamic simulations.

Theoretical considerations

In order to better understand the theoretical bases of a two-zone model, equations of a two-compartment model were derived from the more general advection diffusion equation. Thus, the classic equation for mass diffusion and convection were applied for the two hypothetical compartments of the two-zone model.

A simplified scenario was considered for this mathematical development: a ventilated room (Figure 1), with entry and exit airflows (ventilation rate Q [m^3/s]), and an emission source S(t) [mg/s] releasing a passive scalar (not affecting the velocity field).

The concentration at the entry is supposed to be null. An important assumption is that the flow dynamic boundary conditions are steady state. This room can be divided through a free surface area (FSA), in two parts, representing the two fields of a two-boxes model, near field (NF) and far field (FF).

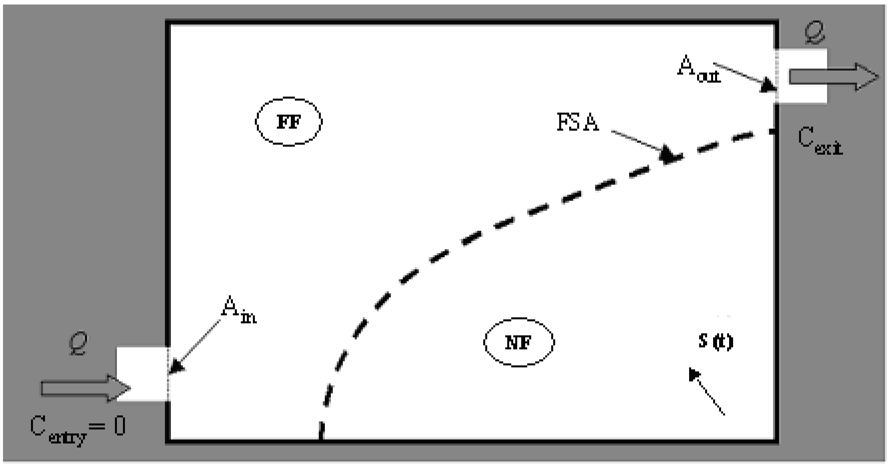

Figure 1 Room with near field (NF) and far field (FF).

Under the previously described assumptions, the local equation for mass diffusion and transport describing the pollutant concentration for each point M of the volume at any time t, may be written as:

$$\frac{\partial C}{\partial t}(M,t)+\vec{u}\ (M)\ .\ \vec{\nabla}C(M,t)=\vec{\nabla}.\left(D_{\mathrm{eff}}(M)\ \vec{\nabla}C(M,t)\right)+\frac{S\ (t)}{V_{\boldsymbol{source}}}\chi_{\boldsymbol{source}}(M) \qquad [1]$$

where C (M, t), is the pollutant concentration for a point M at time t, u (M) is the air velocity at point M. The source is uniformly distributed over a domain Wsource whose volume is Vsource and χ_{source} is a characteristic function defined by

$$\begin{cases}\chi_{\mathrm{source}}(M)=1 & \text{if} \quad M\in\Omega_{\mathrm{source}}\\ \chi_{\mathrm{source}}(M)=0 & \text{if} \quad M\notin\Omega_{\mathrm{source}}\end{cases}$$

D_{eff} is the effective diffusivity, which is the sum of the pollutant molecular diffusivity and of the turbulent diffusivity. Let us now integrate equation [1] for the FF compartment

$$\frac{\partial}{\partial t}\int_{V_{\mathrm{FF}}} C(M,t)\ \boldsymbol{d}V=\int_{V_{\mathrm{FF}}}\left(-\vec{u}\ (M)\ .\ \vec{\nabla}C(M,t)+\vec{\nabla}.\left(D_{\mathrm{eff}}(M)\ \vec{\nabla}C(M,t)\right)\right)\boldsymbol{d}V \qquad [2]$$

Applying the property $-\vec{u}\ .\ \vec{\nabla}C=\vec{\nabla}\ .\ (C\ \vec{u})-C\ \vec{\nabla}.\ \vec{u}$, assuming an incompressible fluid (air + pollutant) ($\vec{\nabla}.\vec{u}=0$), defining a diffusive flow as $\vec{j}=-\ D_{eff}(M)\ \vec{\nabla}C(M,t)$, and using the divergence-flux theorem, the following expressions can be obtained

$$\int_{V_{\mathrm{FF}}}\vec{\nabla}.\vec{j}\ \boldsymbol{d}V=\int_{A_{\mathrm{FF}}}\vec{j}\ .\ \vec{n}_{\mathrm{FF}\boldsymbol{ext}}\ \boldsymbol{d}A$$

$$\int_{V_{FF}} \vec{\nabla}.(C\,\vec{u})\,dV = \int_{A_{FF}} C\,\vec{u}\;.\;\vec{n}_{FF\boldsymbol{ext}}\,dA \qquad [3]$$

where $\vec{n}_{FF\boldsymbol{ext}}$ is the unit normal vector pointing outwards the far field domain. AFF is the surface of the far field, which is the reunion of four surfaces: the interface between the two compartments (FSA), the entry section (A_{in}) and the exit section (A_{out}) and finally the interface with the solid walls. This allows rewriting previous equation [2] under the following form:

$$\frac{\partial}{\partial t}\int_{V_{FF}} C\,\mathrm{d}V = \int_{A_{FF}} (-C\,\vec{u} - \vec{j})\;.\;\vec{n}_{FF\mathrm{ext}}\,\mathrm{d}A = \int_{A_{\mathrm{in}}\,\mathrm{U}\,A_{\mathrm{out}}\,\mathrm{U}\,A_{FSA}} (C\vec{u} + \vec{j})\;.\;\vec{n}_{FF\mathrm{int}}\,\mathrm{d}A \qquad [4]$$

where $\vec{n}_{FF\boldsymbol{int}} = -\vec{n}_{FF\boldsymbol{ext}}$

As a first approximation, the diffusive component $\vec{j}$ of the pollutant flow going through the section of entry can be neglected. The pollutant flows going through the exit and FSA section can be expressed by:

$$\int_{A_{\boldsymbol{out}}} (C\,\vec{u} + \vec{j})\;.\;\vec{n}_{\boldsymbol{FFint}}\,dA \approx -\,Q\,C_{\boldsymbol{exit}} \qquad [5]$$

$$\int_{A_{FSA}} (C\,\vec{u} + \vec{j})\;.\;\vec{n}_{FF\boldsymbol{int}}\,dA = q_{N\rightarrow F} \qquad [6]$$

In equation [5] we have supposed a uniform concentration trough the exit section and neglected diffusive flow. In equation [6], $q_{N\rightarrow F}$ represents the flow of pollutant from NF to FF (mg/s). The average concentrations of pollutant for the two compartments NF and FF can now be calculated as:

$$C_{FF}\ (t) = \frac{1}{V_{FF}} \int_{V_{FF}} C\ (M,\ t)\ \boldsymbol{d}V$$

$$C_{NF}\ (t) = \frac{1}{V_{NF}} \int_{V_{NF}} C\ (M,\ t)\ \boldsymbol{d}V \qquad [7]$$

Combining expressions [5], [6] and [7] we can rewrite equation [4] :

$$V_{FF} \frac{\boldsymbol{d} C_{FF}}{\boldsymbol{d} t} = q_{N \to F} - Q\ C_{\boldsymbol{exit}}\ (t) \qquad [8]$$

Integrating equation [1] for the NF compartment gives in a similar way:

$$V_{NF} \frac{\boldsymbol{d} C_{NF}}{\boldsymbol{d} t} = -\ q_{N \to F} + S\ (t) \qquad [9]$$

In order to integrate these two last equations, the pollutant flow between the two compartments, $q_{N \to F}$, as well as the exit concentration $C_{exit}(t)$ must be described as a function of the source and its history between time 0 and t. Using a Laplace transform, the concentration and the source can be described as follows:

$$\begin{aligned} \overline{C}\,(M,p) &= L\left[C\,(M,t)\right] = \int_{t=0}^{\infty} C\,(M,t)\ \exp(-p\,t)\,\mathrm{d}t \\ \overline{S}\,(p) &= L\left[S\,(t)\right] = \int_{t=0}^{\infty} S\,(t)\ \exp(-p\,t)\,\mathrm{d}t \end{aligned} \qquad [10]$$

$$L\left[\frac{\partial C}{\partial t}\right] = p\ L\left[C(M,t)\right] - C(M,\ t{=}0) = p\ \overline{C}(M,p) \qquad [11]$$

applying the differentiation property of Laplace transform [11], under the hypothesis of a null initial concentration C(M, t=0), equation [1] becomes:

$$p\ \overline{C}(M,p) + \vec{u}\ (M)\ .\ \vec{\nabla}\overline{C}(M,p) - \vec{\nabla}\ .\ \left(D_{eff}(M)\ \vec{\nabla}\overline{C}(M,p)\right) = \frac{\overline{S}\ (p)}{V_{\text{source}}}\chi_{\text{source}}(M) \qquad [12]$$

Equation [12] is a partial derivative equation in space that does not have any time dependency (stationary equation). It is linear with constant coefficients, and its solution, proportional to emission $\overline{\mathrm{S}}(\mathrm{p})$, may be written under the following form:

$$\overline{C}(M,p) = \overline{Z}\ (M,\ p)\ \overline{S}(p) \qquad [13]$$

where function $\overline{\mathrm{Z}}\,(\mathrm{M},\,\mathrm{p})$ is the Laplace transform of the Green's function Z(M,t) of the problem, integrated over the source volume. This function depends on the coefficients of equation [12], i.e. the velocity and turbulent diffusivity fields, as well as on the boundary conditions for the mass transfer at the walls and in the sections of entry and exit. Performing the inverse transform of the equation [13], we obtain a convolution product in time-space:

$$\mathrm{C(M,t)} = \int_0^{\mathrm{t}} \mathrm{Z\ (M,t-t')\ S\ (t')}\ \boldsymbol{d}\mathrm{t'} \qquad [14]$$

This shows that the concentration in a point of the room at time t depends on the history of the source. In Laplace domain all concentrations and flows are proportional to $\overline{S}$, which allows to write:

$$\overline{\mathrm{C}}_{\boldsymbol{exit}}\ (\mathrm{p}) = \overline{\mathrm{K}}_{\boldsymbol{exit}}\ (\mathrm{p})\ \overline{\mathrm{S}}\ (\mathrm{p})$$

$$\overline{C}_{FF}\ (p) = \overline{K}_{FF}\,(p)\,\overline{S}\ (p) \quad ; \quad \overline{C}_{NF}\ (p) = \overline{K}_{NF}\,(p)\,\overline{S}(p) \quad ; \quad \overline{q}_{N\rightarrow F}\,(p) = \overline{K}_q\ (p)\,\overline{S}(p)$$

$$\Rightarrow \qquad \overline{C}_{\text{exit}}(p) = (\overline{K}_{\text{exit}} / \overline{K}_{FF})\, \overline{C}_{FF} = \overline{k}(p)\, \overline{C}_{FF}(p) \qquad [15]$$

$$\Rightarrow \qquad \overline{q}_{N\to F}(p) = \frac{\overline{K}_q}{\overline{K}_{NF} - \overline{K}_{FF}} (\overline{C}_{NF} - \overline{C}_{FF}) = \overline{\beta}\ (\overline{C}_{NF} - \overline{C}_{FF}) \qquad [16]$$

At this point we may introduce three time-constants: t_{source}, the characteristic of the source, t_{conv} (= V_{total}/Q), the characteristic convection time, and finally, t_{diff} (= $V^{2/3}/D_{\text{eff}}$), the characteristic diffusion time.

Assuming very slow temporal variations of the source, i.e. t_{source} significantly higher than t_{diff}, and t_{conv}, we can assume that a kind of ‘slipping mode’ is achieved (no time lag between excitation and response in a system dynamics model). In that case, $\overline{k}$ and $\overline{\beta}$ may be replaced by their long time asymptotic values, k_0 and β_0, (for Laplace parameter p tending towards zero) and equations [15] and [16] may be rewritten as

$$\overline{C}_{exit} = k_0\ \overline{C}_{FF} \qquad \Rightarrow \qquad C_{exit}(t) = k_0\ C_{FF}\ (t) \qquad [17]$$

$$\overline{q}_{N\to F} = \beta_0\ (\overline{C}_{NF} - \overline{C}_{FF}) \qquad \Rightarrow \qquad q_{N\to F} = \beta_0\ (C_{NF} - C_{FF}) \qquad [18]$$

If the far field FF is perfectly mixed, then $k_0 = 1$. Finally, we may write the long time version of equations [8] and [9] in the following form:

$$\begin{aligned} V_{FF} \frac{d C_{FF}}{d t} &= \beta_0\ C_{NF} - (\beta_0 + k_0\ Q)\ C_{FF} \\ V_{NF} \frac{d C_{NF}}{d t} &= -\beta_0\ (C_{NF} - C_{FF}) + S(t) \end{aligned} \qquad [19]$$

These last coupled equations correspond to the two-zone model mass balance equations, which at steady state become as follows (with a constant emission rate, S (t)=S):

$$C_{NF} = \frac{S}{k_0 Q} + \frac{S}{\beta_0} \qquad C_{FF} = \frac{S}{k_0 Q} \qquad [20]$$

The previous theoretical considerations may lead to several comments. Firstly the concentration C_{NF} and C_{FF} represent spatial average concentrations: two single captors cannot directly record them, except if each compartment is perfectly mixed. Moreover C_{NF} and C_{FF} depend on the definition of the compartments, as well as on the value of the air flow rate between the two compartments. It is interesting to observe that in order to obtain these equations no hypothesis has been made on the shape of the near field volume. So these mass balance equations are applicable for whatever near-field volume we wish, under the conditions that it includes the emission without the presence of the entry and exit airflows sections on its boundary. Equations [20] are generally written in the classical two-zone model with coefficient k_0 equal to one, reducing the spatial variability in concentration to only two ideally mixed zones.

Material and methods

Experimental Setup

Laboratory experiments were performed in order to evaluate the existence of compartments and their geometrical shapes under specific ventilation conditions. Direct reading instruments were used in order to obtain simultaneous measurements of the concentrations in several points of an experimental room.

Measurements were performed in a 10 m^3 ventilated booth under controlled emission conditions.

Figure 2 shows a schematic view of the experimental setup. It consisted of a $10m^3$ experimental chamber with air inlet and exhaust openings located on the same wall. The total cross sections were respectively of 0.0325 m^2 for the inlet air and 0.0251 m^2 for the exhaust air. Airflow through the air entry and exit openings were monitored to calculate air exchanges per hour. The air velocities on cross-sectional area of the openings were measured in multiple points using an anemometer (TSI VelociCalc Plus., St Paul, MN, USA) before and after each experience, in order to confirm a stable ventilation rate during all experiments. Experiments were performed under two different ventilation conditions representing 17 and 10 [h^{-1}] air exchanges per hour.

Emission (S) was positioned at the floor level. A constant emission of 1.69 mg/s was achieved with a peristaltic pump injecting ethanol on a hotplate causing instantaneous evaporation. A hotplate-like heating device was conceived to allow constant and instantaneous evaporation without affecting temperature gradient in the vicinity. Temperature around the source was measured to confirm that no convection was induced by a potential temperature gradient. The emission rate was assessed gravimetrically through mass balance.

Emission was released long enough to reach steady state conditions (at least 16 and 28 minutes respectively for 17 and 10 [h^{-1}]) and maintain them for at least 5 minutes. These condition also insured slipping mode conditions since the values of Peclet numbers were > 1 for each ventilation condition (with D_{eff}=0.0025 m^2/s).

Concentrations were measured at 64 locations in the room using direct reading semiconductor sensors (Figaro 822, Figaro Engineering Inc., Japan) and analyzed using a dedicated solution for data acquisition (LabVIEW, National Instrument Corp., Austin, Tx, USA). This sensor was selected considering its high sensitivity to organic solvent vapors, such as ethanol, its simplicity, and its low cost. Measurements were performed at four heights (0.4, 0.8, 1.2 and 1.6 m) with 16 sampling points for each height arranged in a 0.4m spaced grid (Figure 2 b). This particular sampling scheme was chosen in order to provide a fair representation of the pollutant spread in the experimental chamber. Due to the limited number of available sensors, sequential runs of eight measurements were performed in similar ventilation conditions (reproductive conditions) to obtain 64 measurements points. Two replicate runs were made for each set of measurements in order to evaluate the reproducibility of results. Captors were calibrated before each simulation, using freshly prepared static standards. Concentrations were recorded at a frequency of one measure every 10 second for each captor.

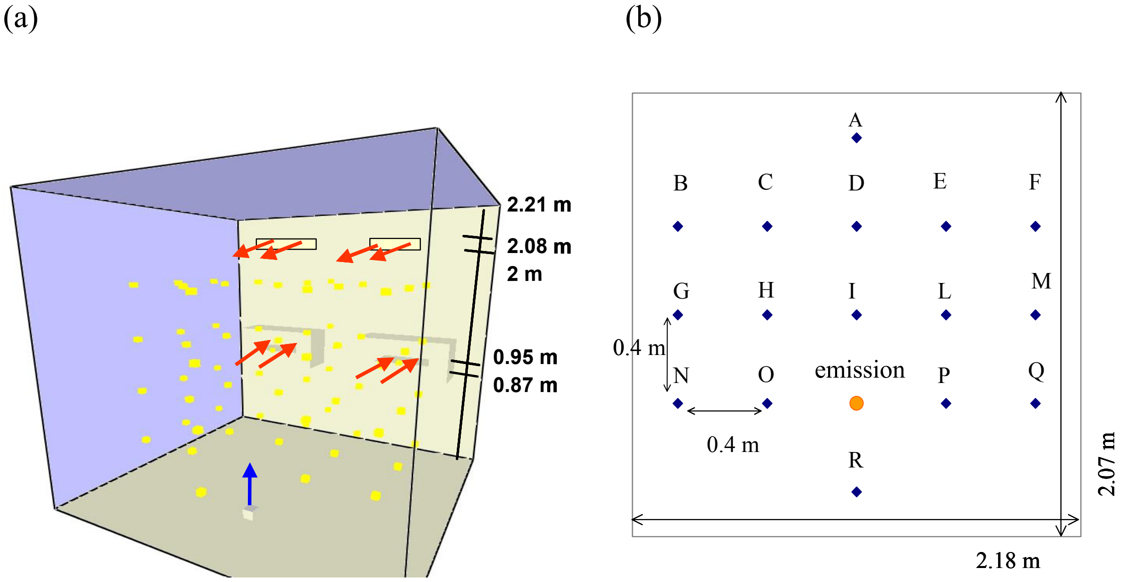

Figure 2 Experimental set-up: a) experimental chamber, b) measurement locations at one height.

Numerical Analysis

Experimental measurements were analyzed in order to study the pertinence of the compartment theory and to verify whether a two-compartment hypothesis was adequate.

Steady state concentrations averaged over at least 5 minutes were used to describe the spatial distribution of the pollutant. The steady state concentrations estimated by a two-compartment model (equations [20]) were compared to experimental results. Different values of β, the inter-compartment flow rate, have been selected over a range of recommended value (23).

In a first step of our analysis, we have considered the classical definition of near field shaped as a hemisphere spread around the source. Thus, measured near field concentrations were obtained as spatial averaged concentration in a hemisphere around the source for different radii. The mixing factor coefficient, k_0, and the inter-compartment flow rate β, have also been calculated by applying the two-zone model to our measures.

In a second step the existence of compartments was explored without specifying their shape. The spatial distributions of the concentration were observed, and the possibility of a multimodal distribution evaluated using Kernel density functions (26) (see equation [21]).

As an alternative approach to histograms, Kernel density estimation represents data distributions, without the typical drawbacks associated with histograms such as the dependence of their density shape and location on width of the interval chosen (scale shift problem). Given a series of n observations (measured) x_i, the Kernel estimator of the density of xi may be obtained using the following equation:

$$\hat{f}_k(x) = \frac{1}{nh}\sum_{i=1}^{n} k\left(\frac{x - x_i}{h}\right) \qquad [21]$$

where k() is the standard normal density function.

The bandwidth parameter, h, determines the degree of smoothing, and may influence the form of the distribution. In our application we used a recommended value (27): the optimal bandwidth that minimizes the Asymptotic Mean Integrated Squared Error. It is defined as:

$$h = 0.9 * \min(s, \frac{IQR}{1.34}) * n^{-0.2} \qquad [22]$$

where s is the standard deviation for xi observations, IQR is the interquartile range and n the number of data points.

CFD Simulations

The spatial and temporal variations of measured steady state concentrations for two ventilation conditions were also compared to numerical simulation results, using Fire Dynamics Simulator (FDS, (Version 4), a free Computer Fluid Dynamic numerical code developed by NIST (National Institute of Standards and Technology). This model, designed specifically for fire-driven fluid flow, may also be applied to simulate other fluid flow not involving fire or thermal processes (28). It is based on the fundamental equations of mass, momentum and energy, solved numerically over a finely-spaced grid. Equations and numerical algorithm are described in a Technical Reference Guide (29).

FDS outputs (airflows, pollutant concentrations) were visualized through Smokeview, a software tool permitting to display time dependent tracer flow, animate contour slice and surface of computed variable (30,30).

The grid size was chosen to give a resolution sufficient to describe the physical dimensions of the smallest details of chamber and emission device, namely 10 cm. This is a critical parameter to obtain reliable results.

Results and discussion

Experimental Results versus Two-zone model

Table I shows the time-averaged steady state concentrations measured at the 64 points for each ventilation condition. For the case of 17 air exchanges two replicate sets of measurements were done, to check the reproducibility. An averaged difference of 6 % suggested a fairly reproducible experimental set-up.

Table I: Measured concentration [mg/m^3] in the experimental chamber, respectively for 17 and 10 h^{-1}. The measured points attributed to the near field according to the Kernel analysis are indicated in bold.

Position Height [m]	A	B	C	D	E	F	G	H	I	L	M	N	O	P	Q	R
0.4	**49.4**	15.0	18.7	**60.9**	35.8	20.4	14.8	18.0	24.2	32.2	15.0	16.2	17.0	23.3	20.6	19.8
0.8	**53.1**	18.4	17.5	25.5	**53.4**	17.9	17.8	22.2	25.0	22.5	17.9	17.6	19.8	27.4	25.7	22.7
1.2	**45.7**	20.6	21.1	30.8	33.2	36.9	20.9	27.0	32.8	26.8	21.8	23.4	25.6	35.8	34.1	27.8
1.6	**47.1**	22.9	21.9	27.6	23.7	24.1	22.6	24.0	28.7	23.7	19.6	22.4	25.1	33.2	31.7	25.2

Position Height [m]	A	B	C	D	E	F	G	H	I	L	M	N	O	P	Q	R
0.4	30.2	**70.5**	**83.0**	23.0	**75.4**	**68.0**	**69.3**	**77.0**	24.4	**66.4**	**62.7**	27.3	28.2	24.8	24.8	24.0
0.8	33.7	**70.1**	**65.0**	30.2	**103.3**	**80.2**	**56.9**	**66.4**	**56.0**	**77.5**	**67.7**	27.5	25.9	46.9	25.5	31.0
1.2	49.6	**90.9**	**79.8**	**64.9**	**115.0**	**81.8**	**71.2**	**90.7**	**94.0**	**95.0**	**73.7**	30.8	28.8	39.3	29.5	34.5
1.6	**80.9**	**87.2**	**96.4**	**66.3**	**147.2**	**92.6**	**77.2**	**102.9**	38.7	**113.5**	**83.2**	35.3	32.4	33.4	35.7	39.3

Firstly, the steady state concentrations of the two compartments model were compared with measurements. To estimate the near field concentrations, according to equation [20], the following values of β were taken into account (23): 0.096, 0.188 and 0.368 m^3/s. We also assumed an ideal mixing in the far field ($k_0=1$). Application of these literature values to equation [20] allows the calculation of the NF and FF concentrations of the model (C_{NF} model and C_{FF} model).

Then, the measured near field concentrations were averaged over different hemisphere around the source. The considered hemisphere radii ranged between the minimum radii points, 0.6 m, until a maximum radius, 1.4 m, which corresponds to a hemisphere volume (5.7 m^3) approximately equal to half the chamber volume. The averaged concentrations corresponded to measurement points inside (NF) or outside (FF) the hemisphere for each of its radii.

As Table II shows, the measured concentrations do not fit well with C_{NF} and C_{FF} calculated by the model. Moreover, the measured near field concentrations, averaged over a hemisphere around the source, are always lower than the measured far field concentrations averaged over the rest of the volume, and this for any radius. As a consequence, the inter-compartment flow rate, estimated by applying the two-box model to these concentrations, always takes negative values, which does not make sense.

That could mean that, if a near field compartment exists, for this experimental condition it does not have any hemispheric shape at all, as assumed here to calculate the average measured concentrations.

To investigate the existence of possible compartments, measured concentration distributions were observed through a Kernel density function. As illustrated in Figure 3, a multimodal distribution of concentrations for each ventilation condition can be observed. At least two distributions can be demonstrated statistically (Silverman's test of multimodality Silverman 1983), which may confirm the initial hypothesis about the existence of two zones of "similar" concentrations. Thus, it was possible to attribute these two zones to the two modes of concentration distribution. In particular we associated all the points presenting a concentration higher than 42 mg/m^3 and 50 mg/m^3 to a near field volume for 10 and 17 air exchanges par hour respectively. A sensitivity analysis was performed around these minimum values, and no significant differences leading to different conclusion were found.

It is interesting to note that the zones defined that way had no regular geometrical shape. Moreover, their geometry appeared to be strongly influenced by ventilation rates, as indicated by the changes in their shape and extension in relation to ventilation rate. Indeed, the new "near" field, for 17 ach, assumes an irregular shape (see the measurement points corresponding to values reported in bold figures in the first part of Table I). Figure 5 shows this irregular shape around the emission source for this airflow whereas, for 10 ach, the new near field assumes another configuration, as shown in Table 1.

With this new compartment partition, it was possible to recalculate the averaged measured concentrations for the new two fields and to compare them again with model outputs, as shown in Table III. This new partition allows a better fit with the model output, specially concerning the near field concentrations. The β parameters recalculated on the base of this new compartments partition take positive values.

These results indicate that from a statistical point of view, the compartmental theory makes sense for this experimental configuration: two distinct zones, described respectively by two different concentration distributions can be clearly detected here. Nevertheless simple geometrical shapes, such as the half-sphere commonly used in the two-zone model, are not suitable to describe the near field zone, since pollutant dispersion is strongly influenced by ventilation around the source.

Table II: Comparison between CNF and CFF calculated through the two-box model (at steady state condition for a given range of β parameters [0.096, 0.188 and 0.368 m^3/s]), and the spatial averaged concentrations measured in the experimental booth, for different near field radius.

	ethanol concentration [mg/m^3]									
	ventilation: 17 h^{-1}					ventilation: 10 h^{-1}				
radius $_{NF}$ [m]:	**0.6**	**0.8**	**1**	**1.2**	**1.4**	**0.6**	**0.8**	**1**	**1.2**	**1.4**
C_{NF} (avg measured conc.)	21.1	22.4	24.0	23.9	25.1	25.3	40.8	48.1	45.3	53.8
C_{NF} model [min-max]		**40.4**	**-**	**53.4**			**65.4**	**-**	**78.4**	
C_{FF} (avg measured conc.)	26.8	26.9	27.5	28.5	28.3	62.9	62.6	65.8	67.9	69.7
C_{FF} model			**35.8**					**60.8**		
k_0	*1.334*	*1.331*	*1.301*	*1.255*	*1.263*	*0.967*	*0.972*	*0.925*	*0.896*	*0.872*
β [m^3/s]	*-0.293*	*-0.377*	*-0.483*	*-0.362*	*-0.521*	*-0.045*	*-0.078*	*-0.096*	*-0.075*	*-0.106*

Another important observation concerns the temporal variability of measurements. Large temporal variations were observed for the measurements classified as near field points. As shown in Figure 4, a large variability with inter-quartile ranges of respectively 41 and 23 mg/m^3 were observed for points A and D. For the other data points at the same level, classified in the far field, the interquartile range had an average equal to 2.6 mg/m^3. The same tendency was observed for the other near field data points in the chamber.

So, in the presence of a complex flow pattern with directional effects, a new "near field" can be defined, for which the term "near" does not necessary mean close to the source. It rather means a "proximity effect", corresponding to a higher concentration value combined with a large variability, as defined by McBride (31).

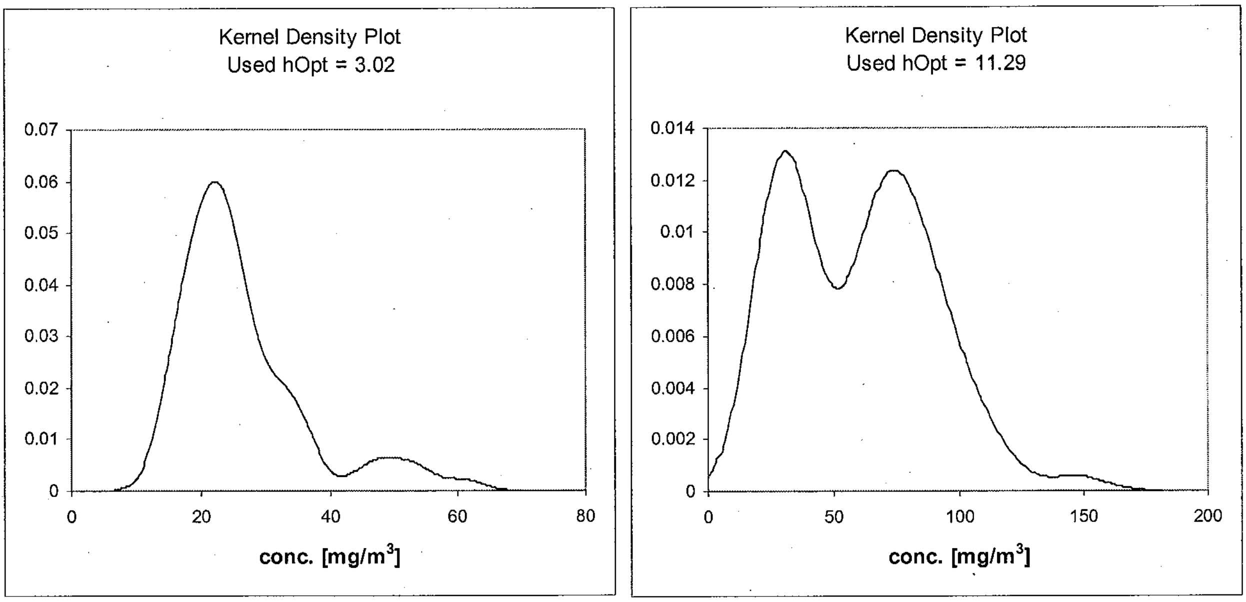

Figure 3 : Kernel density function for the ethanol concentrations measured during experience with respectively 17 (a) and 10 (b) air exchange per hour. The presence of at least two peaks has been confirmed by a Silverman's test of multimodality.

Table III: Comparison between C_{NF} and C_{FF} calculated through the two box model (at steady state condition, for a given range of β parameters [0.096, 0.188 and 0.368 m3/s] and for k0 = 1) and the spatial average of measured concentrations (C_{NF}^2 and C_{FF}^2) for the new compartment partition found out through a Kernel density function.

	ethanol concentration [mg/m^3]	
ventilation:	**17 h^{-1}**	**10 h^{-1}**
C_{NF}^2 (avg measured conc.)	51.6	81.4
C_{NF} model [min-max]	**40.4 - 53.4**	**65.4 - 78.4**
C_{FF}^2 (avg measured conc.)	23.9	31.9
C_{FF} model	**35.8**	**60.8**
k_0	*0.7*	*0.52*
β [m^3/s]	*0.06*	*0.03*

Experimental Results versus CFD Results

The measured concentrations, spatial distributions and temporal variations of concentration at steady state, were also compared with CFD results, for the case of 17 air exchanges per hour (Figure 5). Even if the agreement observed between experimental measurements and simulation data as shown in Figure 6 is not perfect, the general tendency of this two kinds of

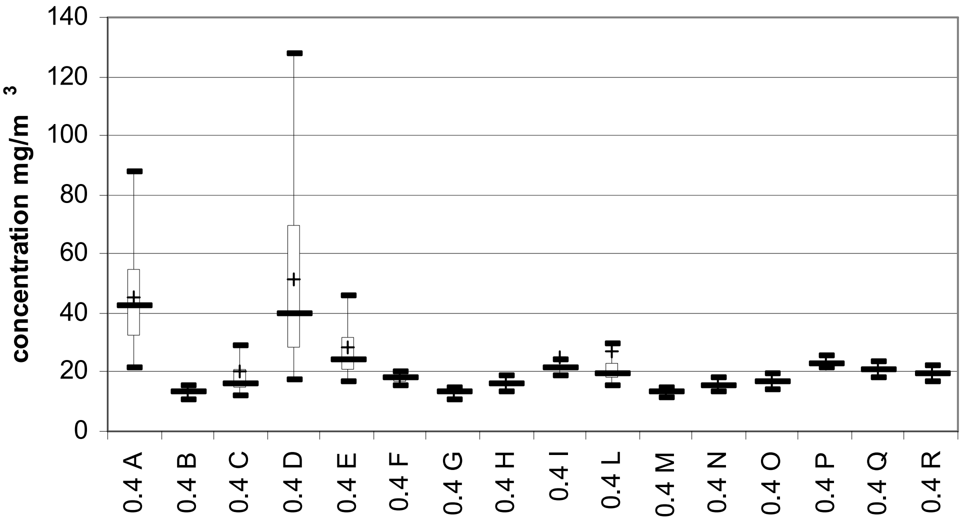

Figure 4: Boxplots of concentrations for the first level (0.4 m of high).

data demonstrate a positive correlation. Furthermore, the observation of the spatial distribution of the concentrations obtained by CFD confirmed our initial hypothesis about the existence of two compartments of irregular geometries. This is clearly apparent in Figure 5 where the concentration profiles are clearly not described by a hemispherical geometry. Still, we have also observed that the FDS simulated concentrations belonging to the "near field" presented, at steady state conditions, the same behaviour as the measured concentration with a high level of temporal fluctuations.

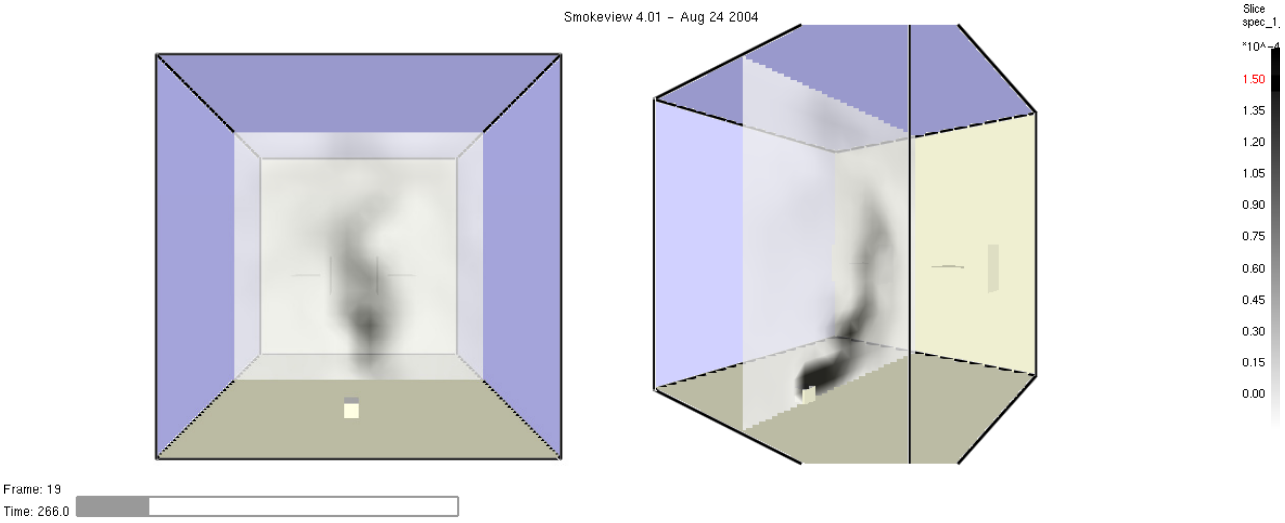

Figure 5: Snapshots of the steady state concentrations contour in two vertical planes, for the case of 17 h^{-1} [kg ethanol/kg air].

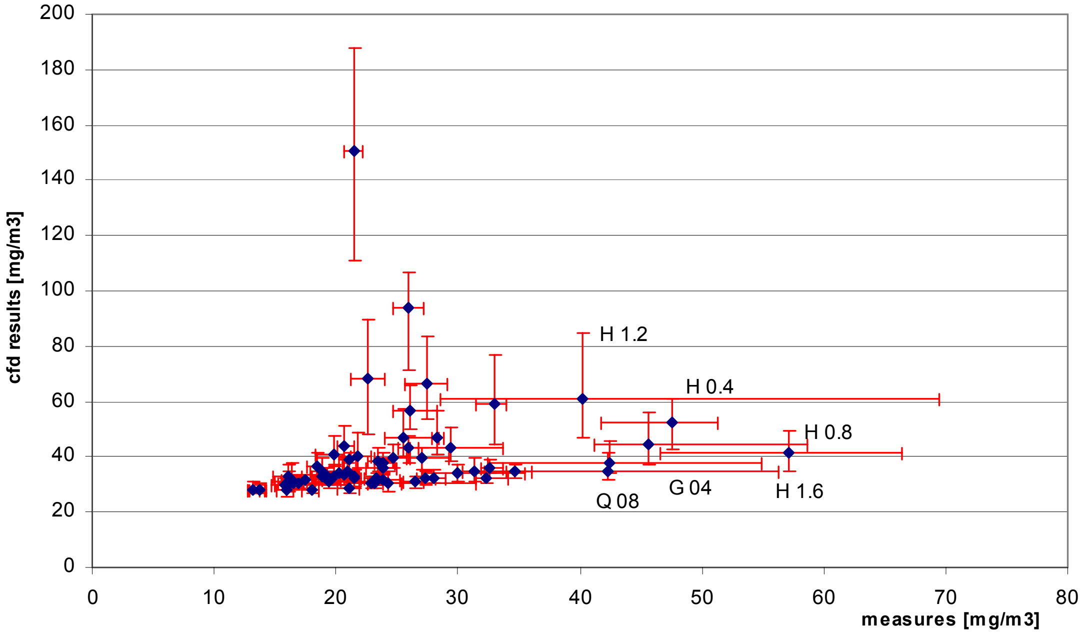

Figure 6: Measured versus CFD model concentration with the representation of inter-quartile range

Conclusions

Theoretical considerations and experimental investigations reported here are useful for a better appraisal of the concept of a two-zone model. Specifically, we have demonstrated that this simplified model can be obtained from the more general advection-diffusion equation. Both NF and FF concentrations have been shown to correspond to virtual average concentrations within the compartments and not to point concentrations that were directly observable.

On the other hand, the experimental measurements carried out were not in agreement with NF and FF concentrations estimated with a hemispherical two-zone model. A closer look at the data indicated nevertheless the existence of several compartments. Based on two non-spherical compartments, we were thus able to demonstrate a good agreement between the experimental data and the predictions of the two-zone model.

The application of CFD simulation also confirmed the existence of two irregular compartments, whose average concentrations were close to the measured data.

It should be noted that specific concentration distributions observed in this experience were associated to the particular configuration of the experimental booth; each situation has thus to be evaluated on a case-by-case basis.

However, for field practice, it is essential to realize that a 2-zone compartmental model can produce reliable results if the geometry of the NF compartment is defined with caution. The difficulty is thus now to decide which section of a workplace belongs to the NF zone. It has been clearly demonstrated that distance, although important, is not the only parameter to be considered. Further field measurements taken under different working conditions should be used to define the two zones. Research in this direction is in progress. Data (measurements and exposure determinants) collected in a related project (32) are currently analysed focusing on field attribution decision.

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References

1. Vernez, D., R. Bruzzi, H. Kupferschmidt, A. De Batz, P. O. Droz, and R. Lazor: Acute respiratory syndrome after inhalation of waterproofing sprays: A posteriori exposure-response assessment in 102 cases. J.Occup.Environ.Hyg. 2005).
2. Bruzzi, R., D. Vernez, P. O. Droz, and A. De Batz: Beliefs and practices in the assessment of workplace pollutants. Sozial- und Praventivmedizin 51:5-13 (2006).
3. Bruzzi, R., D. Vernez, P. E. Sottas, and P. O. Droz: Exposure models in Switzerland. An overview of the present situation. Gefahrstoffe Reinhaltung Der Luft 2005).
4. Keil, C. B. and R. Murphy: An application of exposure modeling in exposure assessments for a university chemistry teaching laboratory. J.Occup.Environ.Hyg. 3:99-106 (2006).
5. Whicker, J. J., P. T. Wasiolek, and R. A. Tavani: Influence of room geometry and ventilation rate on airflow and aerosol dispersion: Implications for worker protection. Health Physics 82:52-63 (2002).
6. Lee, E., C. E. Feigley, J. A. Khan, and J. R. Hussey: The effect of temperature differences on the distribution of an airborne contaminant in an experimental room. Annals of Occupational Hygiene. 50:527-537 (2006).
7. Brohus, H. and P. V. Nielsen: Personal Exposure in Displacement Ventilated Rooms. indoor air 6:157-167 (1996).
8. Welling, I., I. M. Andersson, G. Rosen, J. Raisanen, T. Mielo, K. Marttinen, and R. Niemela: Contaminant dispersion in the vicinity of a worker in a uniform velocity field. Annals of Occupational Hygiene 44:219-225 (1-5-2000).

9. Guffey, S. E., M. E. Flanagan, and G. Van Belle: Air sampling at the chest and ear as representative of the breathing zone. American Industrial Hygiene Association Journal 62:416-427 (2001).

10. Kim, T. and M. R. Flynn: The effect of contaminant source momentum on a worker's breathing zone concentration in a uniform freestream. American Industrial Hygiene Association Journal 53:757-766 (1992).

11. Ojima, J.: Effect of cross current due to worker's arm movement on local exhaust ventilation hood. J.UOEH 27:273-278 (2005).

12. Keil, C. B.: Mathematical Models for Estimating Occupational Exposure to Chemicals. AIHA Press, Farifax (2000).

13. Jayjock, M.A. Modeling Inhalation Exposure, chapter 16 in The Occupational Environment – Its Evaluation and Control, edited by S.R. DiNardi, American Industrial Hygiene Association, Fairfax, VA (1997).

14. Ozkaynak, H., P. B. Ryan, G. A. Allen, and W. A. Turner: Indoor air quality modeling: Compartmental approach with reactive chemistry. Environ Int 8:461-471 (1982).

15. Nazaroff, W. W. and G. R. Cass: Mathematical modeling of indoor aerosol dynamics. Environmental Science and Technology 23:157-166 (1989).

16. Heinsohn, R.: Industrial Ventilation: Engineering Principles, New York/ Chichester/ Brisbane/ Toronto/ Singapore: John Wiley & Sons, 1991.

17. Nicas, M.: Estimating exposure intensity in an imperfectly mixed room. Am.Ind.Hyg.Assoc.J. 57:542-550 (1996).

18. Cherrie, J. W. and T. Schneider: Validation of a new method for structured subjective assessment of past concentrations. Annals of Occupational Hygiene. 43:235-245 (1999).

19. Rodes, C. E., R. M. Kamens, and R. W. Wiener: The significance and characteristics of the personal activity cloud on exposure assessment measurements for indoor contaminants. Indoor Air 2:123-145 (1991).

20. Furtaw, E. J., M. D. Pandian, and J. V. Behar: An indoor personal air exposure model enhancement to account for proximity to emission sources. ISEE/ISEA Joint Conference 1994).

21. Cherrie, J. W.: The beginning of the science underpinning occupational hygiene. Annals of Occupational Hygiene. 47:179-185 (2003).

22. Flynn, M. R. and B. Ljungqvist: A review of wake effects on worker exposure. Annals of Occupational Hygiene. 39:211-221 (1995).

23. Keil, C. B.: A tiered approach to deterministic models for indoor air exposures. Appl.Occup.Environ.Hyg. 15:145-151 (2000).

24. Burton, D.: Hemeon's Plant and Process Ventilation, Burton, D. (eds.). Lewis Publishers, 1999.

25. Nicas, M., M. J. Plisko, and J. W. Spencer: Estimating benzene exposure at a solvent parts washer. J.Occup.Environ.Hyg. 3:284-291 (2006).

26. Hirschberg, J., L. Lu, and J. Lye: Descriptive Methods for Cross-Section Data. The Australian Economic Review 38:333-350 (2005).

27. Silverman, B. W.: Density estimation for statistics and data analysis, New York: Chapman & Hall., 1986.

28. Chang, C. H. and R. N. Meroney: Concentration and flow distributions in urban street canyons: wind tunnel and computational data. J.Wind Engin.and Indus.Aerody. 91:1141-1154 (2003).

29. McGrattan, K. B., Baum, H. R., Rehm, R. G., Hamins, A., and Forney, G. P.: Fire Dynamics Simulator: Technical Reference Guide, National Institute of Standards and Technology.NISTIR 6467 (eds.). 2000.

30. Forney, G. P. and McGrattan, K. B.: User's Guide for Smokeview Version 4. A Tool for Visualizing Fire Dynamics Simulation Data., National Institute of Standards and Technology (eds.). 2004.

31. McBride, S. J., A. R. Ferro, W. R. Ott, P. Switzer, and L. M. Hildemann: Investigations of the proximity effect for pollutants in the indoor environment. J.Exp.Anal.Environ.Epidem. 9:602-621 (1999).

32. Sottas, P. E., R. Bruzzi, and D. Vernez: A Bayesian framework for combining different occupational exposure assessment methods. Proceeding of IOHA Conference 2005).

Paper V

Adaptation of compartmental exposure models to include workplace information.

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Keywords:

Exposure determinants, compartmental models, exposure modelling, workplace assessments

Abstract

Despite being often the most preferred among the various physical models and being more frequently applied, the two-zone model may present some drawbacks regarding the physical compartments basis as well as the lack of criteria for determining some of the input parameters (i.e. the inter compartment airflow).

The purpose of this paper is to propose an alternative method that allows gathering information on field attribution of a given exposure point on the basis of observable determinants.

We asked a group of hygienists to retrospectively describe a series of exposure field situations for which concentrations data were available, according to predefined series of observable exposure determinants.

The one-zone and two-zone models were compared to the new model, a determined based model, built in order to organize in a structured manner all the selected determinants, allowing for a better description of near field dispersion.

A field attribution index was also calculated from the determinants based model in order to integrate additional information in a field attribution decision. The hypothetical improvements progressively achieved from the additional information have consequently been tested. Although results did not show a significant improvement in near field definition according to exposure determinants, we have proposed a generalized preliminary approach allowing for a rough quantitative estimation of exposure levels.

Introduction

Among the various approaches for exposure assessment, physical models are considered as an attractive tool for decision making in occupational hygiene, particularly in situations where air sampling is not feasible (Nicas and Jayjock, 2002;Keil and Murphy, 2006). Briefly, 'physical' or 'emission' models, provide exposure estimates based on data about the emission rate of contaminants and their dispersion in the workplace.

According to Cherrie, the two-zone model represents a good compromise between more elaborated but more data-demanding physical models (e.g. computational fluid dynamics) – and a rough one-zone model assuming perfect mixing in the whole workplace (Cherrie, 1999). The two-zone model conceptualises the exposure space into just two simple volumes of two different concentration values: the near field and the far field. The near field represents a volume close to the emission point while the far field is the remaining workplace area.

Recent studies have demonstrated the suitability of these models to predict solvent concentrations during laboratory simulation of metal parts washing (Nicas et al., 2006;Spencer and Plisko, 2007).

The simplification of the workplace volume into a near field and a far field is especially practical since it provides a way of estimating personal exposure levels (i.e. exposure of the worker at the emission point, in the near field) and general ambient exposure (ambient concentration in the workplace, the far field).

The two-zone model is, however, not without limitations. Firstly, the physical ground of compartments theory – how to define the near field volume and which kind of shape it assumes – is not obvious. Another drawback with this model is the lack of criteria or guidelines for determining the input parameters, such as the inter compartment airflow.

Literature (Nicas, 2003;Keil and Murphy, 2006) often recommend this model which has been used in a retrospective exposure assessment to identify, thanks to the near field concentration, the pollutant concentration level of a person standing close to the source (Vernez et al., 2006). Usually, for the near field volume, studies suggested a hemisphere of a fixed 1 m radius, including source and worker breathing zone, (Keil and Murphy, 2006;Spencer and Plisko, 2007). Variations of the near field geometry as a function of emission and ventilation configuration have also been proposed by Nicas (Nicas, 1996;Nicas et al., 2006).

Recently, based on multiple measurements made in a controlled emission chamber as well as a computational fluid dynamics model, Bruzzi et al. (Bruzzi et al., 2006b) observed that the volume qualified as the near field – because of higher associated concentrations – was not of regular geometrical shape and that geometry was strongly influenced by ventilation rates. Their results therefore argue against the use of rigid geometrical shapes, such as a half-sphere or a cube, to define near field zones. In the same study, the authors observed good agreement between measurements and model predictions when the near field was defined empirically, with no particular pre-defined geometry. Other studies have shown the inadequacy of ideal mixing assumption in a priori defined near field volume: air movements induced by the worker presence or directional convective flows are not included in the classical model (Furtaw et al., 1996;Welling et al., 2000).

Thus, in field practice, the challenge could be to decide which section of a workplace to associate to the NF concentration level. It has been clearly demonstrated that distance, although important, is not the only parameter to consider for a field attribution decision, and some other criteria are required. Further, the need to predict near source concentrations on the base of observable parameters was also stressed out in a recent survey among Swiss occupational hygienists (Bruzzi et al., 2006a).

In this paper we propose a method that allows estimation of near and far field concentrations of a two-zone model from a set of determinants easily and flexibly estimated by expert judgment, and permits to evaluate whether a particular situation should be evaluated using the near field or the far field concentration estimate.

Performance of the proposed method is evaluated using measurements collected in different workplaces, by hygienists of the Institute for occupational health sciences of Lausanne.

Exposure determinants

Exposure determinants selection

Firstly, a series of key exposure determinants have been selected, based on literature review and according to a recent study focused, inter alia, on an identification of contextual parameters observed during expert judgements (Bruzzi et al., 2006a).

Together with the widespread determinants, such as measurement position – expressed as distance from the emission source – workplace volume and general ventilation rate, we have combined additional parameters allowing for a better description of emission conditions and near-field dilution. Those are intrinsic emission determinants such as source velocity and source orientation, and air turbulence around the source.

Intrinsic factors, regarding emission phenomena, such as source orientation and source velocity, give us information on how the emitted pollutant will diffuse in the near field. Previous investigations (Brohus et al., 1996;Welling et al., 2000;Guffey et al., 2001;Hyun and Kleinstreuer, 2001) already demonstrated that the inclusion of emission orientation in exposure assessment models may reduce bias. Guerra (Guerra, 2005) took advantage of initial source velocity to define the near field extension: he represented the near field boundary as the limit between the area for which the pollutant flow velocity is governing and the rest of the room dominated by natural or forced convection.

Air turbulences near the source, due for instance to worker activity and movements around the source (Ojima, 2005), specify the degree of mixing of the released pollutant induced by external factors. Several authors have studied the influence of turbulence on pollutant dispersion (Mora et al., 2003;Jayaraman et al., 2006).

Determinants parameterisation

In a second step, we asked a group of hygienists to describe retrospectively, using the proposed determinants, a series of exposure field situations for which concentrations were available.

To facilitate the description of each determinant, a spreadsheet has been designed, allowing to record exact information when available, to estimate a range (when possible) or to give a qualitative information. Table I shows how we defined qualitative categories for workplace volumes, ventilations, initial emission velocities, turbulences directions and distances. These classes have been selected in accordance with experts' observations.

The emission rate expresses the total mass released (G) per unit time. No qualitative estimation is offered for this first parameter but a range of values is generally used to describe it. On the other hand, five qualitative categories are proposed to the experts, for workplace volume (V) and general ventilation (Q), if they are not confident enough to give an exact value, or a range.

Table I: Possible qualitative categories for the different exposure determinants

Workplace volume (V):		**m^3**
1	very small local	< 50
2	small local	50-500
3	standard local	500-2500
4	large local	2500-10000
5	very large local	> 10000

Ventilation (Q):		**m^3/h**
1	very small installation	< 50
2	small installation	50-500
3	standard installation	500-2500
4	large installation	2500-10000
5	very large installation	> 10000

Initial emission velocity (v):		**m/s**
1	slow	0 - 0.2
2	normal	0.2 - 0.5
3	high	> 0.5

Turbulence (τ):		**m/s**
1	weak	<0.05
2	standard	0.05 - 0.2
3	high	> 0.2

Direction (θ):		**degree**
1	omnidirectional	30
2	opposed to sampling point	90
3	direct to sampling point	0

Distance (d):		**m**
1	personal sampling	0,5

For the measurements location, three different options have been offered to specify distance from emission source (d): for fixed measurements, 1) to fill with an exact value or 2) with a range (both in meters unit), 3) otherwise a personal sampling option was proposed.

The source orientation (θ) that symbolizes the angle of the emission direction depended on a specific emission process or particular ventilation pattern in the near field, with respect to the sampling point location. To simplify we proposed three possible options: omnidirectional emission, source with a direction opposed to sampling point, and emission direct to sampling point. For each situation we have selected three possible angles (see Figure 1); for the situation of no preferential direction – the omnidirectional situation – we have nevertheless assumed a main vertical direction.

To describe the degree of turbulence near the source (τ), three qualitative levels have been specified: low, standard and high. For source velocity (v) three options are proposed: introduction of a single value, of a range or of a qualitative appreciation according to the categories illustrated in Table 1.

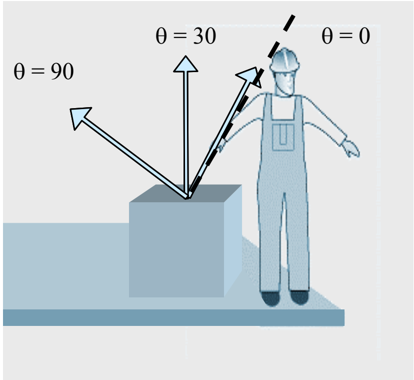

Figure 1: Outline of the three possible angles between worker and emission direction

Physical model developments

To take advantage from all the information gathered about exposure conditions, we needed to build up a model permitting to organize all previously selected determinants, in a structured approach. The aim was to calculate concentration levels combining emission estimations and determinants observations, according to different approaches.

We have considered the classical one-zone and two-zone models, a new model integrating all determinants and finally a modified two-zone model, as illustrated below.

Thus, we tested the hypothetical improvements progressively achieved through additional information, considering respectively ventilation condition, near field ventilation patterns, directivity turbulence and emission velocity.

For a better understanding, we may formulate the concentration levels C_i calculated by the different approaches i, as following:

$$C_i = G * F_i \qquad \text{with} \qquad F_i = f(determinants) \qquad [1]$$

G stands for the emission rate and F_i represents the dispersion factor, as a function of the various determinants considered respectively by the different approaches i.

One-zone model

Firstly, we have thus integrated information on general ventilation (Q), through a one-zone model, and the previous equation becomes simpler:

$$C_1 = \frac{G}{Q} \qquad \text{with} \qquad F_1 = \frac{1}{Q} \qquad [2]$$

Two-zone model

Secondly, for the case of the two-zone model, we obtain two concentration levels, respectively the far and near field concentrations:

$$C_{FF} = \frac{G}{Q} \qquad \text{distance} > 1\text{ m} \qquad F_{FF} = \frac{1}{Q} \qquad [3]$$

$$C_{NF} = \frac{G}{Q} + \frac{G}{\beta} \qquad \text{distance} < 1\text{ m}, \qquad \text{with} \qquad F_{NF} = (\frac{1}{G} + \frac{1}{\beta})$$

β represents the inter compartment air exchange. Hygienists are used to associating to near field concentration levels to the exposure that occurs at a distance lower then 1m, basing therefore their decision only on geometric considerations. However it could be interesting to profit from supplementary knowledge for an enhanced field attribution decision, as described below.

Determinant based model

The third approach considered is the one defined in this paper. This model attempts to combine all observable and available determinants to better represent exposure:

$$C_{det} = G^{*} F_{det} \qquad F_{det} = f(V, Q, d, \tau, v, \vartheta) \qquad [4]$$

The matter is now how to relate all previous determinants. We have considered two variants of general turbulent diffusion model (Franke and Wadden, 1987), described respectively by Roach (Roach, 1981;Lennert et al., 1997) and Scheff (Scheff et al., 1992;Mulhausen and Damiano, 1998). The classical diffusion model is based on Fick's law, and at the steady state condition becomes:

$$C_r = \frac{G}{4\pi D r} \qquad [5]$$

The concentration is directly proportional to emission (G), and inversely proportional to the distance (r) and eddy diffusion coefficient (D), following a spherical symmetry.

In the second equation, Roach integrated to the diffusion model the ventilation rate. He also added the concept that the stationary concentration in the air discharged at the periphery (distance equal to R) is equal to the equilibrium concentration of an ideally mixed model. This version is more suitable for indoor situations, for which boundaries may cause local accumulation.

$$C_r = \frac{G}{Q} + \frac{G}{4\pi D}(\frac{1}{r} - \frac{1}{R}) \qquad [6]$$

Both models are used for completely random dispersion, but it can also be modified to reflect the presence of advective flow in the room. Thus, the Gaussian Plume Dispersion model is based on a diffusion model that takes into account the direction of air currents (x the downwind distance from the source along the centerline of the plume and v the air velocity).

$$C_r = \frac{G}{4\pi Dr} e^{\left[\frac{-v}{2D}(r-x)\right]} \qquad [7]$$

Therefore, a resulting equation, taking advantage of previously adaptations, will integrate diffusion and advection. In this version, we have expressed R (distance from the periphery), taking into account the room volume, idealized as a hemisphere centered into a source location. Thus, if exposure occurs close to the source, concentration level will be rather influenced by eddy diffusion coefficient, orientation of principal local flow and source velocity. On the opposite, in the room boundary, concentration will converge to the ideal mixed concentration, and exposure will be more influenced by general ventilation than local conditions.

$$C_d = G[\frac{1}{Q} + \frac{1}{4\pi D}(\frac{1}{r} - \frac{1}{\sqrt[3]{\frac{3V}{2\pi}}})e^{[\frac{-v\cdot r}{2D}(1-\cos(\theta))]}] \qquad [8]$$

The simple observable parameters previously illustrated have been parameterised in order to include them in the different models. To model – from previous described observable determinants – the value of the inter compartment flow, beta (in the two-zone model) and the eddy diffusion coefficient (in the diffusion models), we took advantage of the observation regarding turbulence around the source and source velocity. A combination of the three qualitative categories of turbulence with the three qualitative categories of source velocity has been taken into account, to find out five classes. As shown in table II and III, a value of a beta coefficient or an eddy diffusion coefficient has been associated to each class, in accordance with range values proposed by Keil (Keil, 2000). If the emission source velocity was introduced as a range, we considered the category in which the average value was found.

Diffusion coeff. [m^2/s]

	source velocity		
Turbulence	1	2	3
1	0.002	0.004	0.005
2	0.004	0.005	0.008
3	0.005	0.008	0.010

Table II: The diffusion coefficients as a function of qualitative exposure determinants

Beta coeff. [m^3/s]

	source velocity		
Turbulence	1	2	3
1	0.05	0.09	0.17
2	0.09	0.17	0.29
3	0.17	0.29	0.5

Table III: The inter compartment flow rate (beta) as a function of qualitative exposure determinants

Modified two-zone model

Finally, the last approach to describe exposure was based on classical two-zone model predictions, for which field attribution was defined, not only by observing the distance from the source, but also by integrating additional determinants, on the base of the previous model outputs. A "field attribution index" was thus calculated, describing the relative distance of the new concentration estimation to the near and far field concentration levels (see Figure 2), as following:

$$field\_attribution\_index = \frac{C_{det} - C_{FF}}{C_{NF} - C_{FF}} \qquad [13]$$

Thus, field attribution decision, made on the basis of this index, will be considered as a preliminary part of a two-zone model application. For the following analyses we have chosen arbitrarily 0.5 as our discriminating value.

$$\begin{aligned} C_{FF} &= \frac{G}{Q} && \text{index} < 0.5 \\ C_{NF} &= G * \left(\frac{1}{Q} + \frac{1}{\beta}\right) && \text{index} > 0.5 \end{aligned} \qquad [14]$$

Therefore, different classical approaches, such as one zone model and two-zone model, were tested and compared to the new approach, proposed in this article, which integrates all determinants available. Moreover, on the base of the output of this last approach, we proposed a new discrimination between the two levels of a classical two-zone model.

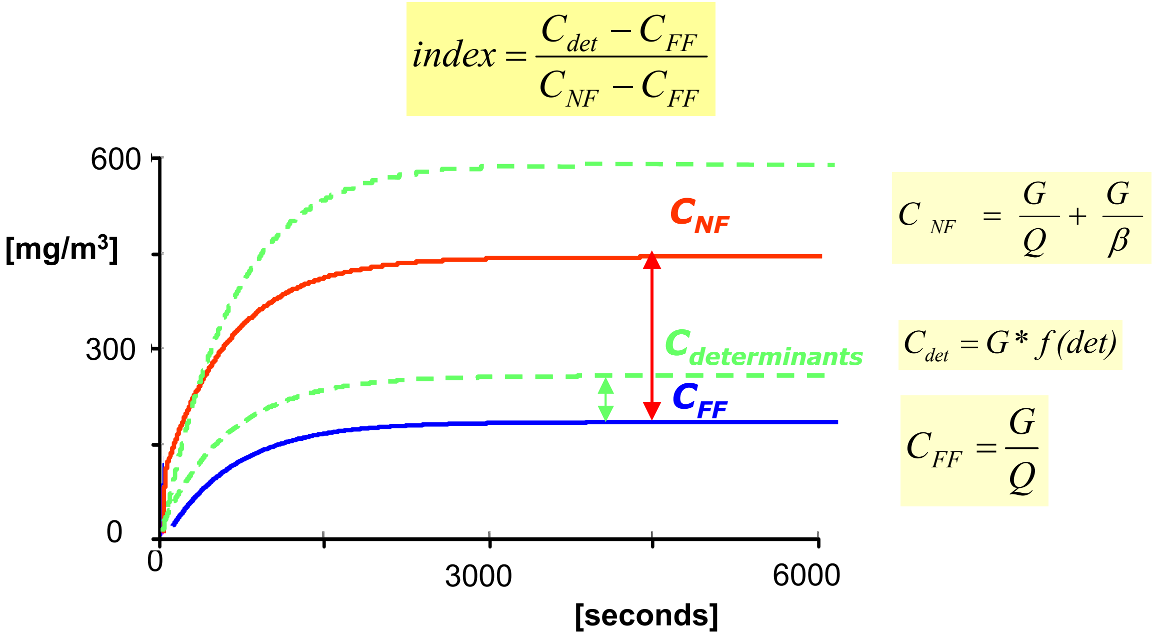

Figure 2: Field attribution index definition.

Application to occupational field situations

Data description

The previously described models have been partially evaluated using data collected at the Institute for Work and Health of Lausanne. These data come from 15 surveys and include exposure levels for 12 different substances, for a total of 144 measurements (see table IV).

As table IV shows, this data set consists mainly of two big categories, N,N-dimethylethylamine (DMEA) and isopropyl alcohol, which correspond to almost 60% of all measurements. These data were collected in a survey focused on the characterization of exposure to tertiary amines in iron foundries (Buser et al., 1991). All measurements, for these 2 chemicals and for the other contaminants were made using state-of-the art occupational hygiene sampling and analytical procedures (NMAM – NIOSH Manual of Analytical Methods). The data set represents both long term (8 hours) and short term (several minutes) measurements. Associated determinants were estimated to represent the values associated to these time frames.

To simplify data presentation, we have summarized results according to 4 groups: DMEA, isopropyl alcohol, other organic volatiles (COVs) and aerosols.

Considering all substances, the emission rates estimated ranged over a large scale, from 0.004 to 1054 mg/s. Still, for the same situation the uncertainty of expert estimation for the emission, measured as the ratio of the maximum and the minimum of the emission ranges, was about two orders of magnitude.

Substance	**Survey**	**n**	**Industry**
*Cutting Oil mist	A	10	Machine manufactory
DMEA	B	37	Foundry
**Acetonitrile	C	6	Biotechnology
**Benzene	D	3	Chemical
*Chromium	E	4	Clock-making
**Formaldehyde	F	5	Chemical
**Heptane	G	4	Medical manufactory
Isopropyl alcohol	H	46	Foundry
	I	1	Medical manufactory
	J	1	
**Perchloroethylene	K	6	Dry cleaning
*Lead	L	12	Metal manufactory
*Aerosols	M	3	
	N	3	Metal manufactory
**Ethanol	O	3	Pharmaceutical
Total		144	
**COVs		27	
*Aerosols		32	

Table IV: Summary of exposure cases for the different substances

Evaluation of the agreement between predictions and measurements

Agreement between the different model predictions and measured concentrations was evaluated using three different approaches. Firstly, the ratio of measured to predicted levels was calculated as reported by Spencer (Spencer and Plisko, 2007), and quartiles of this ratio were determined. Secondly, the Spearman correlation coefficient between measured and predicted levels was estimated, providing insight in the models' ability to order exposure estimates similarly to the measured concentrations. Thirdly, a linear regression was applied on the natural logarithm of the measured concentrations. Our models are of the form:

$$C = G * F \qquad [15]$$

After logtransformation, a multiple regression model can be fitted to the data using the following framework:

$$\ln(C) = a + b \times \ln(G) + c \times \ln(F) + \varepsilon \qquad [15]$$

a, b, and c are to be estimated, and ε is the error term.

For each model, the coefficient of multiple determination was calculated, and the parameters a, b, and c were estimated.

The field attribution index ranged from 0 (for the exposure "far" from the source) equal to an ideal mixed situation, to 9.1 (for the worst cases), with a median of 1.12.

According to this new discrimination, based on the field attribution index and not on worker's distance from the source, we found roughly 70% of cases fall in the same field depicted by a classical two-zone model, but roughly 16% of the exposures occurs at a distance more than 1 m fell again in the near field category (Table V).

Table V: Summarise of near and far field cases according to the classical two-zone model and the two-zone model modified respectively.

		Model two-zone according with new index	
		FF	NF
Classical two-zone model	FF	**29**	23
	NF	20	**72**

Table VI: Distribution of the ratio of measured to predicted levels, calculated according to different approaches for the all products' categories.

		25th	**Median**	**75th**
COVs	1 box	0.041	0.13	4.48
	Classical 2 box	0.017	0.044	0.57
	Determinant based	0.017	0.038	0.057
	2 box modified	0.016	0.03	0.56
Aerosols	1 box	0.024	0.076	1.3
	Classical 2 box	0.00046	0.017	0.058
	Determinant based	0.0097	0.034	0.079
	2 box modified	0.0005	0.014	0.063
Isopropyl alcohol	1 box	7.6	12.9	20.8
	Classical 2 box	0.21	6.15	12.2
	Determinant based	0.09	0.16	0.33
	2 box modified	0.02	0.04	0.14
DMEA	1 box	4.82	8.19	14.2
	Classical 2 box	0.3	0.47	1.02
	Determinant based	0.33	0.81	1.71
	2 box modified	0.127	0.39	0.7

Table VI shows the distribution of the ratio of measured to predicted levels calculated according to different approaches for the different categories of products.

Except few cases found in isopropyl alcohol and DMEA categories, all models overestimate exposures. For all substances we found that the two zones model modified – on the bases of all determinants – was the more conservative.

Table VII: Spearman correlation coefficient between measured and predicted levels for the different models (* p<0.05, ** p<0.005).

	G	**G^*F_{1box}**	**G^*F_{2box}**	**$G^*F_{det.}$**	**$G^*F_{2box\ modified}$**	**n**
Aerosols	**0.43 ***	**0.6 ****	**0.49 ***	**0.52 ****	**0.55 ****	**32**
COVs	**0.61 ****	**0.83 ****	**0.86 ****	**0.86 ****	**0.84 ****	**27**
Isopropyl alcohol	**-0.26**	**-0.22**	**0.17**	**0.41 ***	**0.05**	**46**
DMEA	**0.6 ****	**0.73 ****	**0.77 ****	**0.77 ****	**0.69 ****	**37**

With regard to only DMEA (see Figure 3), results confirm in general a better models performance, especially for the case of models integrating near field observations.

Table VII resumes Spearman's correlation coefficient between measured and predicted levels. A positive and significant correlation ($p < 0.005$) was found for the single categories of COVs and DMEA. Still for these categories, a slightly better correlation was found for the model integrating all the determinants available in this analysis. For the case of isopropyl alcohol a negative correlation was even found between the experts' emissions estimations and the measured concentrations. This partially explains further results for this specific substance.

Table VIII shows respectively the coefficient of multiple determinations, and the estimated parameters a, b, and c. The R^2 coefficients present relatively high values (>0.56), excluding the case of isopropyl alcohol for which we have already found negative Spearman's coefficients.

We may observe that the R^2 coefficients show a general weak increasing tendency as we integrate in the emission estimates the different Fi functions of each one-zone model, two-zone model, model based on determinants and the two-zone model according with the field attribution index (Figure 4). Only for isopropyl alcohol no correlation was found: the negative value of b coefficient depicted for this substance by almost all models, meaning an inverse proportionality with emission estimations, confirms the clear insufficiency of an adequate understanding of this exposure situation.

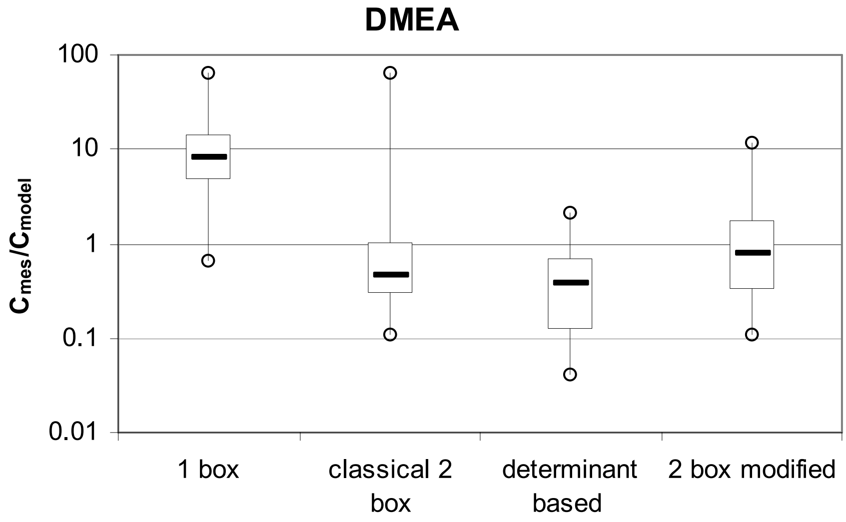

Figure 3: Box plot for the ratio of measured to predicted levels calculated according to different approaches for the DMEA

Table VIII : Coefficient R^2 of multiple determination, and the parameters a, b, and c, estimate according with the model Ln(C) = a + b*Ln (G)+c*Ln(F)

	$Ln(C_{mesured})$=	Ln(G)	$Ln(G)+ln(F_{1box})$	$Ln(G)+ ln(F_{2box})$	$Ln(G)+ ln(F_{det.})$	$Ln(G)+ ln(F_{2box\ modif.})$
R^2	Aerosols	0.52	0.58	0.58	0.64	0.61
	COVs	0.46	0.56	0.63	0.57	0.56
	Isopr. Alc.	0.03	0.03	0.15	0.2	0.06
	DMEA	0.63	0.8	0.73	0.89	0.84
a	Aerosols	-2.54	-1.95	-3.11	-2.76	-3.05
	COVs	2.19	1.36	-0.08	0.56	0.32
	Isopr. Alc.	6.33	6.3	5.75	6.07	4.8
	DMEA	-0.13	5.83	-0.52	-0.63	-1.41
b	Aerosols	0.42	0.36	0.42	0.51	0.44
	COVs	0.7	0.65	0.54	0.58	0.56
	Isopr. Alc.	-0.2	-0.21	-0.07	-0.16	0.04
	DMEA	1.15	1.04	1.16	1.28	1.35
c	Aerosols		0.6	0.34	0.45	0.32
	COVs		0.35	0.67	0.49	0.56
	Isopr. Alc.		-0.05	0.08	0.04	0.12
	DMEA		3.14	0.47	0.54	0.62

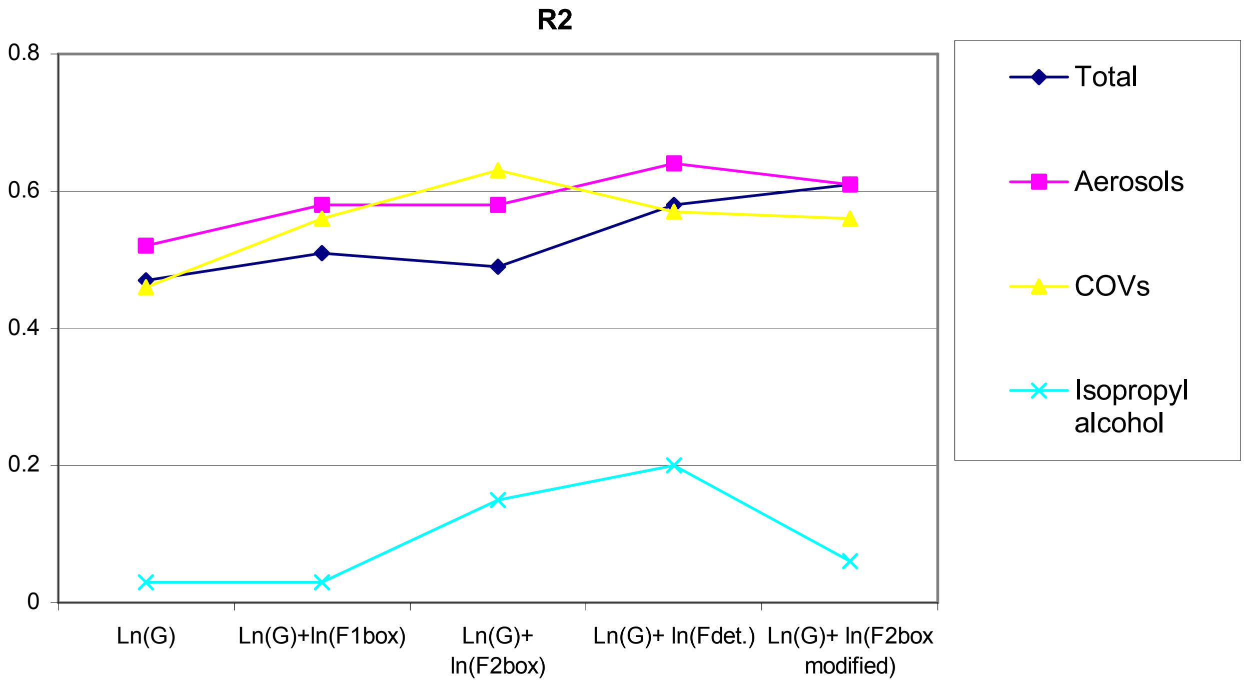

Figure 4 Distribution of R2 coefficients for the different models and substances

Discussion and conclusion

This paper proposes a new method to characterize worker exposure, allowing the integration of expert observations in risk assessment. Our aim was to provide hygienists with a tool able to associate the estimated near or far field concentration levels in a particular exposure situation, considering only a qualitative description of easy observable determinants.

The model ability to accurately predict the absolute value of concentration levels is not as good as we had expected. This is especially the case when we consider all substances as a single category. Looking at each single substance, there is a general tendency to overestimate exposure. However, for DMEA, models predictions are fairly comparable to measurements, especially in the case of determinants based model.

In this case, the comparison between the predicted and the measured values is similar to the output found in a recent study (Spencer and Plisko, 2007), in which however the assessed situations were test simulations, under controlled and known conditions (rather then retrospective field situations).

This significant gap between measurements and models estimates may be explained by the following considerations. Firstly, since input models are based on qualitative estimations of exposure determinants, a bias is introduced by subjective opinion of the experts participating in surveys. An uncertainty of evaluation of determinants was indeed often observed, especially concerning emission rate estimation, defined over a large range.

Some of the approximations we made in modeling also generated a bias. For instance, we have applied the modelled steady state concentrations, due to the missing information concerning the concentration evolution. Afterward, in personal exposure cases, we have assumed a fixed workers' distance to the emission source (0,5 m). These assumptions may partially explain the overestimation.

We have also to mention the complexity of basing models validation on measurements, with regard to their variability. Various cases, presenting different values of concentration measurements, were described by the same determinants.

Despite the limitations previously summarized, this approach represents a useful method for ranking different exposure situations. Actually, if we do not consider the isopropyl alcohol, we have observed through multi regression analyses an average $R^2 = 0,65$, with a general tendency to increase if progressively including determinants.

This application represents a preliminary illustration of how this kind of approach, based on exposure determinants, may support hygienists in an exposure assessment. On the base of these results, a model calibration could be required to better represent exposure levels. More, further investigations will be useful to enhance the performance of this model.

We thus highly recommend all practitioners involved in a measurements survey, to collect, during their assessments, next to concentration levels, all possible information which allows a more comprehensive description of worker exposure.

In conclusion, the model described in this paper is a useful alternative to expert judgments rather than a method competing with measurements. This approach does not pretend to completely replace measurement, since results did not show a significant improvement in near field definition according to exposure determinants. We have, however, proposed an original method, meant to integrate observable determinants to risk assessments.

Indeed, there is a general increase of the expert judgment in practice, and risk assessment is thus often based on specialists' own experience. However generalizations of this experience are relatively challenging, which makes difficult for hygienists to apply them to new situations. We suggest a selection of exposure determinants having an influence on exposure levels, and at the same time we propose a generalized preliminary approach allowing a rough quantitative estimation of exposure levels.

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Reference

1. Brohus,H., Knudsen,H.N., Nielsen,P.V., Clausen,G., and Fanger,P.O., 1996. Perceived air quality in a displacement ventilated room. Proceedings of Indoor Air '96, 1, 811-816.
2. Bruzzi,R., Vernez,D., Droz,P.O., and De Batz,A., 2006a. Beliefs and practices in the assessment of workplace pollutants. Sozial- und Praventivmedizin, 51, 5-13.
3. Bruzzi,R., Vernez,D., Droz,P.O., and De Batz,A., 2006b. Exposure models: current practices and on-going development. In: Renewing a century of commitment to a healthy, safe and productive working life : 28 th International Congress on Occupational Health, Milan, Italy, June 11-16, 2006 : book of abstracts. - Milan : ICOH (Scientific Committee on Education and Training in Occupational Health, 2006. - P. 190).
4. Buser,S., Nicole,C., and Droz,P.O., 1991. Exposition aux amines tertiaires dans les fonderies. Travail et santé, 7, 33-37.
5. Cherrie,J.W., 1999. The effect of room size and general ventilation on the relationship between near and far-field concentrations. Applied Occupational and Environmental Hygiene, 14, 539-546.
6. Franke,J.E. and Wadden,R.A., 1987. Indoor contaminant emission rates characterized by source activity factors. Environmental Science and Technology, 21, 45-51.
7. Furtaw,J., Pandlan,M.D., Nelson,D.R., and Behar,J.V., 1996. Modeling Indoor Air Concentrations Near Emission Sources in Imperfectly Mixed Rooms. Journal of the Air and Waste Management Association, 46, 861-868.

8. Guerra,D., 2005. Modélisation du transfert d'un aerocontaminant dans un local ventilé en champ proche d'une source d'émission accidentelle. Thèse. Laboratoire de Génie Chimique, Département Procédés et Systèmes, Toulouse..

9. Guffey,S.E., Flanagan,M.E., and Van Belle,G., 2001. Air sampling at the chest and ear as representative of the breathing zone. American Industrial Hygiene Association Journal, 62, 416-427.

10. Hyun,S. and Kleinstreuer,C., 2001. Numerical simulation of mixed convection heat and mass transfer in a human inhalation test chamber. International Journal of Heat and Mass Transfer, 44, 2247-2260.

11. Jayaraman,B., Finlayson,E.U., Sohn,M.D., Thatcher,T.L., Price,P.N., Wood,E.E., Sextro,R.G., and Gadgil,A.J., 2006. Tracer gas transport under mixed convection conditions in an experimental atrium: Comparison between experiments and CFD predictions. Atmospheric Environment, 40, 5236-5250.

12. Keil,C.B., 2000. A tiered approach to deterministic models for indoor air exposures. Applied Occupational and Environmental Hygiene, 15, 145-151.

13. Keil,C.B. and Murphy,R., 2006. An application of exposure modeling in exposure assessments for a university chemistry teaching laboratory. Journal of Occupational and Environmental Hygiene, 3, 99-106.

14. Lennert,A., Nielsen,F., and Breum,N.O., 1997. Evaluation of evaporation and concentration distribution models--a test chamber study. Ann.Occup.Hyg., 41, 625-641.

15. Mora,L., Gadgil,A.J., and Wurtz,E., 2003. Comparing zonal and CFD model predictions of isothermal indoor airflows to experimental data. Indoor Air, 13, 77-85.

16. Mulhausen,J.R. and Damiano,J., 1998. A Strategy for Assessing and Managing Occupational Exposures.

17. Nicas,M., 2003. Using mathematical models to estimate exposure to workplace air contaminants. Chemical Health and Safety, 10, 14-21.

18. Nicas,M., 1996. Estimating exposure intensity in an imperfectly mixed room. American Industrial Hygiene Association Journal, 57, 542-550.

19. Nicas,M. and Jayjock,M., 2002. Uncertainty in exposure estimates made by modeling versus monitoring. American Industrial Hygiene Association Journal, 63, 275-283.

20. Nicas,M., Plisko,M.J., and Spencer,J.W., 2006. Estimating benzene exposure at a solvent parts washer. Journal of Occupational and Environmental Hygiene, 3, 284-291.

21. Ojima,J., 2005. Effect of cross current due to worker's arm movement on local exhaust ventilation hood. Journal of UOEH, 27, 273-278.

22. Roach,S.A., 1981. On the role of turbulent diffusion in ventilation. Annals of Occupational Hygiene, 24, 105-132.

23. Scheff,P.A., Friedman,R.L., Franke,J.E., Conroy,L.M., and Wadden,R.A., 1992. Source activity modeling of Freon« emissions from open-top vapor degreasers. Applied Occupational and Environmental Hygiene, 7, 127-134.

24. Spencer,J.W. and Plisko,M.J., 2007. A comparison study using a mathematical model and actual exposure monitoring for estimating solvent exposures during the disassembly of metal parts. J.Occup Environ.Hyg, 4, 253-259.

25. Vernez,D., Bruzzi,R., Kupferschmidt,H., De-Batz,A., Droz,P., and Lazor,R., 2006. Acute respiratory syndrome after inhalation of waterproofing sprays: A posteriori exposure-response assessment in 102 cases. Journal of Occupational and Environmental Hygiene, 3, 250-261.

26. Welling,I., Andersson,I.M., Rosen,G., nen,J., Mielo,T., Marttinen,K., and Niemela,R., 2000. Contaminant dispersion in the vicinity of a worker in a uniform velocity field. Annals of Occupational Hygiene, 44, 219-225.

Discussion

The aim of this project was to develop a model that would take into account easily observable exposure determinants in order to represent occupational exposure to chemicals while integrating the variability of workplace conditions.

A survey among Swiss occupational hygienists was undertaken in order to provide a better insight into hygienist's practices. We found that relatively few occupational hygienists have extensive knowledge of or experience in physical models for exposure assessments. This is due to their perception of models as overly sophisticated tools, not easy applicable to real situations, with little accuracy and reliability. For this reason, they think that the most beneficial improvements of exposure models would be to include input parameters which are easily accessible by field investigations.

Alternatively, long-term sampling has been recognized as the best method in risk assessment. However this method also has its weaknesses. We identified for instance the inability to represent exposure variability within and between workers, generally higher costs, and the technical complexity. Due to environmental variability, a measurement is only representative of a short period of time, at one location and for a specific worker's activity. As such, the elaborated information is not exploitable otherwise.

Indeed, models may enrich the practitioners' toolbox for exposure assessment. In fact, allowing for simulation of unlimited exposure situations, under different assumptions, these models may help hygienists in the understanding and interpretation of data derived from sampling activities. For this reason, we strongly recommend that hygienists regularly collect all the available information ancillary to a measurement, in order to associate all the relevant determinants to a single concentration value. This will ensure proper interpretation of a measurement and allow for later exploitation, such as modelling.

In this context, we have suggested a set of exposure determinants to be combined with measurements data in a exposure databank. This databank was created in Switzerland in 1991 by the Institute of Work and Health of Lausanne in order to support experts in assessing new exposure situations in cases to those documented in the databank.

Moreover, models permit exposure estimations whenever measurements are not possible or not available, such as for epidemiological investigations or for decisions regarding the selection of controls.

For instance, in a recent breakdown of acute pulmonary toxicity following exposure to waterproofing spray, we have implemented a simple model to reconstruct exposure of a group of exposed individuals. Although confidence in model output was limited due to the conservative assumptions made and the simplicity of the model, the procedure permitted a very interesting relative ranking of exposure conditions. Through this analysis, we were indeed able to deduce certain conclusions. High response variability occurred between exposed individuals, and therefore, for this kind of product, it was not possible to define a threshold dose below which adverse effects appear. Thus, in this case, we demonstrated how a simple model may solve problems which are difficult or impracticable to address with field measurements.

Indeed, to depict meaningful outcomes from modeling application, a thorough understanding of the prefixed goals as well as of the data available is essential. In fact, the variety of existing models really allows for a tiered approach in model selection. On one hand, for an epidemiological study, as we have shown, a rough model allowed us to rank the different exposure situations. On the other hand, for experimental experiences, more sophisticated models, such as computational fluid dynamic models (CFD), may provide, under well-known specific conditions, a suitable tool to represent concentration distributions.

In this research, we also had the opportunity to test the CFD model's performance to predict concentration spatial distributions and temporal variations for several measurements points spread inside an experimental chamber. Even if we achieved reasonable predictions, this approach requires a large computational capability and its field application is limited by the necessity of detailed knowledge on multiple parameters. Thus, even if promising, we would suggest that such a tool is not of realistic and practical application.

Nevertheless, on the opposed site of occupational hygiene science, "expert judgments" are found. According to our survey, because of the lack of knowledge on model application and difficulties with measurements, occupational exposure assessments are increasingly based on expert judgments. However, despite being widely used, hygienists themselves qualify this method as the less reliable and efficient approach, just before modelling technique. Expert judgment is based on the knowledge coming from the hygienists' experience acquired in previous assessments. A generalization and formalization of this mental exercise is however rarely found in the literature. We therefore acknowledged the need to organize such information derived from individual observations. More generally, in accordance with the current trends in occupational hygiene and the legal requirements (i.e. MSST, REACH regulation), the need for new tools to assess exposures was also openly declared by the majority of the hygienists investigated.

Another point of interest which emerged from our survey, regards the hygienists' suggestions about requirements for improving models performance. Indeed, they believe that the most valuable improvement of exposure models would be the integration into a single model of more accessible input parameters, to take local dispersion behaviour into account, and to include emission source. Thus, it was just with these suggestions in mind that we developed our research. For a near field exposure representation, according to the outcomes of our simulation experience, we suggest the use of the two-zone compartmental model, but with certain caution. Indeed for field practice, it is important to remember that the two-zone model can generate reliable results only if the geometry of the near field compartment is defined with regard to specific local conditions.

Hence, following these findings, we attempted to depict an alternative representation of near field volume only on the basis of those determinants, which in practice are observed during an expert judgement. Even if, when considering our results with regards to absolute values of measurement levels, we suggest further investigation and validation, we have nevertheless proposed an innovative approach compared to the traditional ones (measurements or expert judgement). Indeed, we suggest a method to structure the information which is normally handled by hygienists through a mental process and on which an expert judgement is generally based. The clear advantage of this approach is that, by allowing for more objective and traceable exposure estimation, it renders possible comparison of different exposure conditions. However, certain limitations still remain in relation to the difficulty to appreciate some determinants such as the emission rate.

The quantification of emission rates represents a non negligible obstacle in the application of all exposure models. If, on one hand, the estimation of this parameter is especially difficult, on the other hand, its accuracy is known to strongly affect the overall assessment performance.

We have found that the emission rate characterization through different models is usually limited due to the large number of possible mixtures, which may generate pollutants, as well as the various emission conditions. In addition, according to the survey's results, hygienists tend to favour practical approaches to estimate emissions, such as a mass balance or field measurements.

In this regard, we intend to carry out field experiences to test the ability of a procedure involving tracer gas, which as traditionally been used industrial hygiene to assess ventilation patterns. Preliminary experiences have shown a strong potential, warranting further investigation.

The European new strategy for managing chemical risks, REACH, (Registration, Evaluation and Authorization of CHemicals), which aims at increasing the transparency in risk assessment, also represents an incentive to continue our research in order to provide more adequate tools to chemical industries and hygienists.

Perspectives

We recommend the following actions to be performed:

- To record systematically together with measurements all determinants having a possible impact on exposure levels.
- To build an inventory for the emission rate values.
- To provide occupational health specialists with a tool, including the present research results, allowing for an immediate estimation of exposure, based on the observation of determinants.
- To develop a more sophisticated tool, permitting, through statistical consideration, the integration of model outputs with the hygienists past experiences as well as with the present measurements.
- To transfer the knowledge gained from such research to occupational health specialists

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References

1. Baldwin,P.E.J. and Maynard,A.D., 1998. A survey of wind speeds in indoor workplaces. *Annals of Occupational Hygiene*, **42**, 303-313.

2. Batterman,S., Jia,C., Hatzivasilis,G., and Godwin,C., 2006. Simultaneous measurement of ventilation using tracer gas techniques and VOC concentrations in homes, garages and vehicles. *J.Environ.Monit.*, **8**, 249-256.

3. Baughman,A.V., Gadgil,A.J., and Nazaroff,W.W., 1994. Mixing of a point source pollutant by natural convection flow within a room. *Indoor Air*, **4**, 114-122.

4. Bemer,D., Dessagne,J.M., and Aubertin,G., 1999. Evaluation of the emission rate from a gaseous source: development of a method using a helium tracer. *American Industrial Hygiene Association Journal*, **60**, 354-362.

5. Bemer,D., Lecler,M.T., Regnier,R., Hecht,G., and Gerber,J.M., 2002. Measuring the emission rate of an aerosol source placed in a ventilated room using a tracer gas: influence of particle wall deposition. *Annals of Occupational Hygiene*, **46**, 347-354.

6. Benke,G., Sim,M., Fritschi,L., Aldred,G., Forbes,A., and Kauppinen,T., 2001. comparison of occupational exposure using three different methods: hygiene panel, job exposure matrix (JEM), and Self Reports. *Applied Occupational and Environmental Hygiene*, **16**, 84-91.

7. Bennett,J.S., Feigley,C.E., Khan,J., and Hosni,M.H., 2000. Comparison of mathematical models for exposure assessment with computational fluid dynamic simulation. *Applied Occupational and Environmental Hygiene*, **15**, 131-144.

8. Bennett,J.S., Feigley,C.E., Khan,J., and Hosni,M.H., 2003. comparison of emission models with computational fluid dynamic simulation and a proposed improved model. *American Industrial Hygiene Association Journal*, **64**, 739-754.

9. Brohus,H., Knudsen,H.N., Nielsen,P.V., Clausen,G., and Fanger,P.O., 1996. Perceived air quality in a displacement ventilated room. In *Proceedings of Indoor Air '96*, Tokyo: Institute of Public Health, **1**, 811-816.

10. Bruzzi,R., 2006. Modèle d'exposition au CO [Abstract]. *Archives des maladies professionnelles et de l'environnement*, **67**, 78.

11. Buringh,E. and Lanting,R., 1991. Exposure variability in the workplace: its implications for the assessment of compliance. *American Industrial Hygiene Association Journal.*, **52**, 6-13.

12. Burton,D., 1999. Hemeon's Plant and Process Ventilation. Boca Raton, Lewis.

13. Chang,C.H., 2006. Computational fluid dynamics simulation of concentration distributions from a point source in the urban street canyons. *Journal of Aerospace Engineering*, **19**, 80-86.

14. Chang,T.J., Hsieh,Y.F., and Kao,H.M., 2006. Numerical investigation of airflow pattern and particulate matter transport in naturally ventilated multi-room buildings. *Indoor Air*, **16**, 136-152.

15. Chern,M.J. and Ma,C.H., 2007. Numerical investigation and recommendations for push-pull ventilation systems. *Journal of Occupational and Environmental Hygiene*, **4**, 184-197.

16. Cherrie,J.W., 2003. The beginning of the science underpinning occupational hygiene. *Annals of Occupational Hygiene*, **47**, 179-185.

17. Cherrie,J.W., 1999. The effect of room size and general ventilation on the relationship between near and far-field concentrations. *Applied Occupational and Environmental Hygiene*, **14**, 539-546.

18. Cherrie,J.W. and Hughson,G.W., 2005. The validity of the EASE expert system for inhalation exposures. *Annals of Occupational Hygiene*, **49**, 125-134.

19. Cherrie,J.W. and Schneider,T., 1999. Validation of a new method for structured subjective assessment of past concentrations. *Annals of Occupational Hygiene*, **43**, 235-245.

20. Cherrie,J.W., Tickner,J., and Friar,J., 2003. *Evaluation and Further Development of the EASE Model 2.0*. Sudbury: HSE Books. Research Report/HSE; 136.

21. Conroy,L.M., Wadden,R.A., Scheff,P.A., Franke,J.E., and Keil,C.B., 1995. Workplace emission factors for hexavalent chromium plating. *Applied Occupational and Environmental Hygiene*, **10**, 620-627.

22. Cooper,D.W. and Horowitz,M., 1986. Exposures from indoor powder releases: Models and experiments. *American Industrial Hygiene Association Journal*, **47**, 214-218.

23. Creely,K.S., Tickner,J., Soutar,A.J., Hughson,G.W., Pryde,D.E., Warren,N.D., Rae,R., Money,C., Phillips,A., and Cherrie,J.W., 2005. Evaluation and further development of EASE model 2.0. *Annals of Occupational Hygiene*, **49**, 135-145.

24. de Cock,J., Kromhout,H., Heederik,D., and Burema,J., 1996. Experts' subjective assessment of pesticide exposure in fruit growing. *Scandinavian Journal Work Environmental Health*, **22**, 425-432.

25. Dosemeci,M., Stewart,P.A., and Blair,A., 1990. Three proposals for retrospective, semiquantitative exposure assessments and their comparison with the other assessment methods. *Applied Occupational and Environmental Hygiene*, **5**, 52-59.

26. Drescher,A.C., Lobascio,C., Gadgil,A.J., and Nazaroff,W.W., 1995. Mixing of a point-source indoor pollutant by forced convection. *Indoor Air*, **5**, 204-214.

27. Drivas,P.J., 1996. Modeling indoor air exposure from short-term point source releases. *Indoor Air*, **6**, 271-277.

28. Fehrenbacher,M.C. and Hummel,A.A., 1996. Evaluation of the mass balance model used by the Environmental Protection Agency for estimating inhalation exposure to new chemical substances. *American Industrial Hygiene Association Journal*, **57**, 526-536.

29. Feigley,C.E., Bennett,J.S., Khan,J., and Lee,E., 2002a. Performance of deterministic workplace exposure assessment models for various contaminant source, air inlet, and exhaust locations. *American Industrial Hygiene Association Journal*, **63**, 402-412.

30. Feigley,C.E., Bennett,J.S., Lee,E., and Khan,J., 2002b. Improving the use of mixing factors for dilution ventilation design. *Applied Occupational and Environmental Hygiene*, **17**, 333-343.

31. Finlayson,E.U., Gadgil,A.J., Thatcher,T.L., and Sextro,R.G., 2004. Pollutant dispersion in a large indoor space. Part 2: Computational fluid dynamics predictions and comparison with a scale model experiment for isothermal flow. *Indoor Air*, **14**, 272-283.

32. Flynn,M.R., Gatano,B.L., McKernan,J.L., Dunn,K.H., Blazicko,B.A., and Carlton,G.N., 1999. Modeling breathing-zone concentrations of airborne contaminants generated during compressed air spray painting. *Annals of Occupational Hygiene*, **43**, 67-76.

33. Flynn,M.R. and Ljungqvist,B., 1995. A review of wake effects on worker exposure. *Annals of Occupational Hygiene*, **39**, 211-221.

34. Franke,J.E. and Wadden,R.A., 1987. Indoor contaminant emission rates characterized by source activity factors. *Environmental Science and Technology*, **21**, 45-51.

35. Furtaw,J., Pandlan,M.D., Nelson,D.R., and Behar,J.V., 1996. Modeling Indoor Air Concentrations Near Emission Sources in Imperfectly Mixed Rooms. *Journal of the Air and Waste Management Association*, **46**, 861-868.

36. Gadgil,A.J., Lobscheid,C., Abadie,M.O., and Finlayson,E.U., 2003. Indoor pollutant mixing time in an isothermal closed room: An investigation using CFD. *Atmospheric Environment*, **37**, 5577-5586.

37. Gmehling,J., Weidlich,U., Lehmann,E., and Fröhlich,N., 1989. Verfahren zur berechnung von luftkonzentrationen bei freisetzung con stoffen aus flüssigen produktgemischen. *Staub - Reinhaltung der Luft*, **49**, 295-299.

38. Guffey,S.E., Flanagan,M.E., and Van Belle,G., 2001. Air sampling at the chest and ear as representative of the breathing zone. *American Industrial Hygiene Association Journal*, **62**, 416-427.

39. Guo,Z., 2002. Review of indoor emission source models. Part 2. Parameter estimation. *Environmental Pollution*, **120**, 551-564.

40. Haberlin,G.M. and Heinsohn,R.J., 1993. Predicting solvent concentrations from coating the inside of bulk storage tanks. *American Industrial Hygiene Association Journal*, **54**, 1-9.

41. He,G., Yang,X., and Srebric,J., 2005a. Removal of contaminants released from room surfaces by displacement and mixing ventilation: modeling and validation. *Indoor Air*, **15**, 367-380.

42. He,Y., Xing,Y.B., Ni,B., Jiang,Y., Suo,J.J., Zhong,G.L., Chang,Q., and Jiang,B., 2005b. [Hypothesis on generating and tracer gas study regarding transmission of severe acute respiratory syndrome through ventilation system in a general hospital]. *Zhonghua Liu Xing.Bing.Xue.Za Zhi.*, **26**, 33-35.

43. Heinsohn,R., 1991. Industrial Ventilation: Engineering Principles. New York: J. Wiley.

44. Heung,W., Yun,M.J., Chang,D.P., Green,P.G., and Halm,C., 2007. Emissions of chromium (VI) from arc welding. *Journal of the Air & Waste Management Association* , **57**, 252-260.

45. Hewett,P., Logan,P., Mulhausen,J., Ramachandran,G., and Banerjee,S., 2006. rating exposure control using bayesian decision analysis. *Journal of Occupational and Environmental Hygiene*, **3**, 568-581.

46. Ishizu,Y., 1980. General equation for the estimation of indoor pollution. *Environmental Science and Technology*, **14**, 1254-1257.

47. Jayaraman,B., Kristoffersen,A.H., Finlayson,E.U., and Gadgil,A.J., 2006. CFD investigation of room ventilation for improved operation of a downdraft table: novel concepts. *Journal of Occupational and Environmental Hygiene*, **3**, 583-591.

48. Jayjock,M.A., 1994. Back pressure modeling of indoor air concentrations from volatilizing sources. *American Industrial Hygiene Association Journal*, **55**, 230-235.

49. Jayjock,M.A., 1997b. Modelling inhalation exposure. In: *The Occupational Environment - Its Evaluation and Control*, DiNardi S.R. ed Fairfax: A.I.H.A, pp. 69-74..

50. Jayjock,M.A., 2005. Exposure modelling research - First things first. *The Synergist*, **16**, no2, 34.

52. Jayjock,M.A., 1988. Assessment of inhalation exposure potential from vapors in the workplace. *American Industrial Hygiene Association Journal*, **49**, 380-385.

53. Karplus,W., 1983. The Spectrum of Mathenatical Models. *Perspectives in Computing*, **2**, 4-13.

54. Kauppinen,T.P., Pannett,B., Marlow,D.A., and Kogevinas,M., 1994. Retrospective assessment of exposure through modeling in a study on cancer risks among workers exposed to phenoxy herbicides, chlorophenols and dioxins. *Scandinavian Journal of Work, Environment and Health*, **20**, 262-271.

55. Keil,C.B., 2000b. A tiered approach to deterministic models for indoor air exposures. *Applied Occupational and Environmental Hygiene*, **15**, 145-151.

56. Keil,C.B.,ed 2000a. *Mathematical Models for Estimating Occupational Exposure to Chemicals*. Fairfax: A.I.H.A.

57. Keil,C.B., kbar-Khanzadeh,F., and Konecny,K.A., 2001. Characterizing formaldehyde emission rates in a gross anatomy laboratory. *Applied Occupational and Environmental Hygiene*, **16**, 967-972.

58. Keil,C.B. and Murphy,R., 2006. An application of exposure modeling in exposure assessments for a university chemistry teaching laboratory. *Journal of Occupational and Environmental Hygiene*, **3**, 99-106.

59. Keil,C.B. and Nicas,M., 2003. Predicting room vapor concentrations due to spills of organic solvents. *American Industrial Hygiene Association Journal*, **64**, 445-454.

60. Keil,C.B., Wadden,R.A., Scheff,P.A., Franke,J.E., and Conroy,L.M., 1997. Determination of multiple source volatile organic compound emission factors in offset printing shops. *Applied Occupational and Environmental Hygiene*, **12**, 111-121.

61. Kim,T. and Flynn,M.R., 1991. Airflow pattern around a worker in a uniform freestream. *American Industrial Hygiene Association Journal*, **52**, 287-296.

62. Kromhout,H., Oostendorp,Y., Heederik,D., and Boleij,J.S., 1987. Agreement between qualitative exposure estimates and quantitative exposure measurements. *American Journal of Industrial Medicine*, **12**, 551-562.

63. Kura,B., Kambham,K., Sangameswaran,S., and Potana,S., 2006. Atmospheric particulate emissions from dry abrasive blasting using coal slag. *Journal of the Air & Waste Management Association*, **56**, 1205-1215.

64. Lacey,S.E., Conroy,L., Schoonover,T., Franke,J., Hedeker,D., and Forst,L., 2006. Dust emission rates from food processing. *Annals of Agricultural and Environmental Medicine*, **13**, 251-257.

65. Lanning,J.S., Boundy,M.G., and Leith,D., 1995. Validating a model for the prediction of dust generation. *Particulate Science and Technology*, **13**, 105-116.

66. Lee,E., Feigley,C.E., and Khan,J., 2002. An investigation of air inlet velocity in simulating the dispersion of indoor contaminants via computational fluid dynamics. *Annals of Occupational Hygiene*, **46**, 701-712.

67. Lee,E., Feigley,C.E., Khan,J.A., and Hussey,J.R., 2006. The effect of temperature differences on the distribution of an airborne contaminant in an experimental room. *Annals of Occupational Hygiene*, **50**, 527-537.

68. Lennert,A., Nielsen,F., and Breum,N.O., 1997. Evaluation of evaporation and concentration distribution models--a test chamber study. *Annals of Occupational Hygiene*, **41**, 625-641.

69. Mannetje,A., Fevotte,J., Fletcher,T., Brennan,P., Legoza,J., Szeremi,M., Paldy,A., Brzeznicki,S., Gromiec,J., Ruxanda-Artenie,C., Stanescu-Dumitru,R., Ivanov,N., Shterengorz,R., Hettychova,L., Krizanova,D., Cassidy,A., Van Tongeren,M., and

Boffetta,P., 2003. Assessing exposure misclassification by expert assessment in multicenter occupational studies. *Epidemiology*, **14**, 585-592.

70. Matthiessen,R.C., 1986. Estimating chemical exposure levels in the workplace. *Chemical Engineering Progress*, **82**, 30-34.

71. Mora,L., Gadgil,A.J., and Wurtz,E., 2003. Comparing zonal and CFD model predictions of isothermal indoor airflows to experimental data. *Indoor Air*, **13**, 77-85.

72. Mulhausen,J.R. and Damiano,J., ed 1998. *A Strategy for Assessing and Managing Occupational Exposures*. 2nd ed Fairfax: A.I.A.H.

73. Nagaraj,A. and Sattler,M.L., 2005. Correlating emissions with time and temperature to predict worst-case emissions from open liquid area sources. *Journal of the Air and Waste Management Association*, **55**, 1077-1084.

74. Nazaroff,W.W. and Cass,G.R., 1989. Mathematical modeling of indoor aerosol dynamics. *Environmental Science and Technology*, **23**, 157-166.

75. Nicas,M., 2003a. Estimating methyl bromide exposure due to offgassing from fumigated commodities. *Applied Occupational and Environmental Hygiene*, **18**, 200-210.

76. Nicas,M., 1996. Estimating exposure intensity in an imperfectly mixed room. *American Industrial Hygiene Association Journal*, **57**, 542-550.

77. Nicas,M., 2003b. Using mathematical models to estimate exposure to workplace air contaminants. *Chemical Health and Safety*, **10**, 14-21.

78. Nicas,M. and Jayjock,M., 2002. Uncertainty in exposure estimates made by modeling versus monitoring. *American Industrial Hygiene Association Journal*, **63**, 275-283.

79. Nicas,M., Plisko,M.J., and Spencer,J.W., 2006. Estimating benzene exposure at a solvent parts washer. *Journal of Occupational and Environmental Hygiene*, **3**, 284-291.

80. Ohmichi,K., Komiyama,M., Matsuno,Y., Takanashi,Y., Miyamoto,H., Kadota,T., Maekawa,M., Toyama,Y., Tatsugi,Y., Kohno,T., Ohmichi,M., and Mori,C., 2006. Formaldehyde exposure in a gross anatomy laboratory--personal exposure level is higher than indoor concentration. *Environmental Science and Pollution Research – International,* **13**, 120-124.

81. Ojima,J., 2005. Effect of cross current due to worker's arm movement on local exhaust ventilation hood. *Journal of University of Occupational and Environmental Health*, **27**, 273-278.

82. Olcerst,R., 1999. 'Use in Well-Ventilated Area'? *American Industrial Hygiene Association Journal*, **60**, 377-383.

83. Ozkaynak,H., Ryan,P.B., Allen,G.A., and Turner,W.A., 1982. Indoor air quality modeling: compartmental approach with reactive chemistry. *Environment International*, **8**, 461-471.

84. Perrier,F., Richon,P., and Sabroux,J.C., 2005. Modelling the effect of air exchange on 222Rn and its progeny concentration in a tunnel atmosphere. *Science of the Total Environment*, **350**, 136-150.

85. Plinke,M.A.E., Leith,D., Goodman,R.G., and Loffler,F., 1994. particle separation mechanisms in flow of granular material. *Particulate Science and Technology*, **12**, 71-87.

86. Qian,H., Li,Y., Nielsen,P.V., Hyldgaard,C.E., Wong,T.W., and Chwang,A.T.Y., 2006. Dispersion of exhaled droplet nuclei in a two-bed hospital ward with three different ventilation systems. *Indoor Air*, **16**, 111-128.

87. Raisanen,J., Niemela,R., and Rosenberg,C., 2001. Tetrachloroethylene emissions and exposure in dry cleaning. Journal of the *Air & Waste Management Association*, **51**, 1671-1675.

88. Ramachandran G,V.JH., 1999. A Bayesian approach to retrospective exposure assessment. *Applied Occupational and Environmental Hygiene*, **14**, 547-557.

89. Ramachandran,G., Banerjee,S., and Vincent,J.H., 2003. Expert judgment and occupational hygiene: Application to aerosol speciation in the nickel primary production industry. *Annals of Occupational Hygiene*, **47**, 461-475.

90. Repace,J.L. and Lowrey,A.H., 1980. Indoor air pollution, tobacco smoke, and public health. *Science*, **208**, 464-472.

91. Roach,S.A., 1981. On the role of turbulent diffusion in ventilation. *Annals of Occupational Hygiene*, **24**, 105-132.

92. Rodes,C.E., Kamens,R.M., and Wiener,R.W., 1991. The significance and characteristics of the personal activity cloud on exposure assessment measurements for indoor contaminants. *Indoor Air*, **2**, 123-145.

93. Rong,C.Y., WaiYuan,T., and Mathew,R.M., 1990. A deterministic mathematical model for quantitative estimation of historical exposure. *American Industrial Hygiene Association Journal*, **51**, 194-201.

94. Säämänen,A. and Skrifvars,M., 2002. The effect of spraying and rolling process factors on styrene emission during the application of unsaturated polyester resins. *AIHA Journal*, **63**, 474-481.

95. Salim,S.M., Viswanathan,S., and Ray,M.B., 2006. Evaluation of source model coupled computational fluid dynamics (CFD) simulation of the dispersion of airborne contaminants in a work environment. *Journal of Occupational and Environmental Hygiene*, **3**, 684-693.

96. Scheff,P.A., Friedman,R.L., Franke,J.E., Conroy,L.M., and Wadden,R.A., 1992. Source activity modeling of freon« emissions from open-top vapor degreasers. *Applied Occupational and Environmental Hygiene*, **7**, 127-134.

97. Selway,M.D., Allen,R.J., and Wadden,R.A., 1980. Ozone production from photocopying machines. *American Industrial Hygiene Association Journal*, **41**, 455-459.

98. Shaw,C.Y., 1993. Application of Tracer Gas Techniques to Ventilation and Indoor Air Quality Investigations. *Indoor and Built Environment*, **2**, 374-382.

99. Sottas,P.E., Bruzzi,R., Vernez,D., Mann,S., Guillemin,M., and Droz,P.O., 2005. A bayesian framework for the combination of different occupational exposure assessment methods [Abstract]. In: *IOHA 6th International Scientific Conference, 19-23 September 2005, programme and abstracts. - Yeoville, South Africa : IOHA (International Occupational Hygiene Association) ; MVS (Mine ventilation Society of South Africa ; SAIOH (Southern African Institute for Occupational Hygiene) ,p 172.*

100. Spencer,J.W. and Plisko,M.J., 2007. A comparison study using a mathematical model and actual exposure monitoring for estimating solvent exposures during the disassembly of metal parts. *Journal of Occupational and Environmental Hygiene*, **4**, 253-259.

101. Tan,Y.M. and Flynn,M.R., 2002. Methods for estimating the transfer efficiency of a compressed air spray gun. *Applied Occupational and Environmental Hygiene*, **17**, 39-46.

102. Taylor,J.R., O'Shaughnessy,P.T., and Reynolds,S.J., 2004. Estimating personal exposures based on mass balance material usage rates: validation of a ventilation model in a spray paint booth. *Journal of Occupational and Environmental Hygiene*, **1**, 213-221.

103. von Grote,J., Hurlimann,C., Scheringer,M., and Hungerbuhler,K., 2003. Reduction of occupational exposure to perchloroethylene and trichloroethylene in metal degreasing over the last 30 years: influences of technology innovation and legislation. *Journal of Exposure Analysis and Environmental Epidemiology*, **13**, 325-340.

104. von Grote,J., Hurlimann,C., Scheringer,M., and Hungerbuhler,K., 2006. Assessing occupational exposure to perchloroethylene in dry cleaning. *Journal of Occupational and Environmental Hygiene*, **3**, 606-619.

105. Wadden,R.A., 2001. Characterization of publication rotogravure press emission rates and compositions. *Applied Occupational and Environmental Hygiene*, **16**, 471-481.

106. Wadden,R.A., Hawkins,J.L., Scheff,P.A., and Franke,J.E., 1991. Characterization of emission factors related to source activity for trichloroethylene degreasing and chrome plating processes. *American Industrial Hygiene Association Journal*, **52**, 349-356.

107. Wadden,R.A., Liao,S.L., Scheff,P.A., Franke,J.E., and Conroy,L.M., 1998. Application of receptor modeling to indoor air emissions from electroplating. *Applied Occupational and Environmental Hygiene*, **13**, 829-838.

108. Wadden,R.A., Scheff,P.A., and Franke,J.E., 1989. Emission factors for trichloroethylene vapor degreasers. *American Industrial Hygiene Association Journal*, **50**, 496-500.

109. Wadden,R.A., Scheff,P.A., Franke,J.E., Conroy,L.M., Javor,M., Keil,C.B., and Milz,S.A., 1995. VOC emission rates and emission factors for a sheetfed offset printing shop. *American Industrial Hygiene Association Journal*, **56**, 368-376.

110. Walker,K.D., Catalano,P., Hammitt,J.K., and Evans,J.S., 2003. Use of expert judgment in exposure assessment: part 2. Calibration of expert judgments about personal exposures to benzene. *Journal of Exposure Analysis and Environmental Epidemiology* , **13**, 1-16.

111. Walker,K.D., Evans,J.S., and MacIntosh,D., 2001. Use of expert judgment in exposure assessment. Part I. Characterization of personal exposure to benzene. *Journal of Exposure Analysis and Environmental Epidemiology*, **11**, 308-322.

112. Welling,I., Andersson,I.M., Rosen,G., nen,J., Mielo,T., Marttinen,K., and Niemela,R., 2000. Contaminant dispersion in the vicinity of a worker in a uniform velocity field. *Annals of Occupational Hygiene*, **44**, 219-225.

113. Whicker,J.J., Wasiolek,P.T., and Tavani,R.A., 2002. Influence of room geometry and ventilation rate on airflow and aerosol dispersion: Implications for worker protection. *Health Physics*, **82**, 52-63.

114. Wild,P., Sauleau,E.A., Bourgkard,E., and Moulin,J.J., 2002. Combining expert ratings and exposure measurements: a random effect paradigm. *Annals of Occupational Hygiene*, **46**, 479-487.

115. Wu,B. and Gebremedhin,K.G., 2001. CFD development and simulation of flow fields in ventilated spaces with multiple occupants. *Transactions of the Asae*, **44**, 1839-1850.

116. Zhang,J.S. and Christianson,L.L., 1990. Room air flow and turbulence characteristics in a 1/4th scale test room., pp. 249-254.

Annex I

QUESTIONNAIRE in OCCUPATIONAL HYGIENE
Occupational Hygienist

For representative results, it is important that you send back this questionnaire to IST in the attached envelope (even if not fulfilled).

The returned questionnaire will be treated in a confidential way. The individual data will not be, in any case, transmitted outside the Institute. The results will be presented in the form of global statistics. This document remains property of the Institute of Occupational Health Sciences.

Fill the gray boxes with your answers or flag the appropriate boxes (with a number inside) corresponding to your answer (It is essential to answer all the questions). Only one answer is usually expected, questions where several answers are possible are indicated as such.

1 Ref.: ☐

A. Background information

The following section aims to understand your background and basic activity in the field of occupational hygienist.

2. **Which of the following categories do you fall into?**

Industry/services (incl. branch solution) ☐ 1
Advisory or consulting body ☐ 2
Authority (SUVA, SECO…) ☐ 3
Other (specify): ☐ 4

3. **In which economic sector are you working (pharmaceutics, chemistry, metalworking.....) ?:** ☐

4. **What was your initial formation?**

Chemistry ☐ 1
Biology ☐ 2
Physics ☐ 3
Environment ☐ 4
Medicine ☐ 5
Other (specify): ☐ 6

5. **How did you get specialized in occupational hygiene? Through (several answers are possible):**

Practice, experience ☐ 1
Postgraduate course (in Switzerland) ☐ 2
Other specialized formation/course (international) ☐ 3

5.a **My specialization is recognized by SGHA/SSHT (MSST specialist in occ. hygiene)**

Yes ☐ 1
No ☐ 2

Please do not hesitate to contact us if you have any questions or problems with the questionnaire
(contact Raffaella Bruzzi , tel 021 3147434, Raffaella.Bruzzi@hospvd.ch)

6. **Years of experience in occupational hygiene:**

7. **What is your occupation rate dedicated to the occupational hygiene activity?**

Full time	(100 %)	☐ 1
Almost full time	(75 to 100 %)	☐ 2
Main activity	(50 to 75 %)	☐ 3
Secondary activity	(25 to 50 %)	☐ 4
Remote activity	(< 25 %)	☐ 5

8. **How often do you perform workplace exposure assessment (whatever the method used)?**

Daily	(≥1 per day)	☐ 1
Weekly	(≥1 per week, < 1 per day)	☐ 2
Monthly	(≥1 per month, < 1 per week)	☐ 3
A few times per year	(≥2 per year, < 1 per month)	☐ 4
1 time per year or less	(< 2 per year)	☐ 5

9. **In which kinds of environment (office work, laboratory, workshop, industrial processes,..) have you usually assessed workplace exposure?**

B. Methods for assessing workplace exposure (gas, vapour, dust)

The following questions are intended to understand the usual methods you use to assess indoor occupational exposures (chronic and sub-acute exposures).

How often are you employing the following techniques in order to characterize workplace environment?

		Between **100-80%** of cases	Between **80-50%** of cases	Between **50-10%** of cases	Between **10- 0%** of cases	**never**
10	**Assess exposure only on the basis of employee interview/ workplace visit (expert judgment)**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
11	**Measuring exposure (punctual, short term-measurement)**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
12	**Measuring exposure (direct reading or sampling during a significant part of the work activity)**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
13	**Biological monitoring**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
14	**Use of predictive mathematical and statistical model (exposure model)**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
15	**Making reference to literature/ existing exposure levels / state of the art (ex. good laboratory practices)**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
16	**Other (specify):**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5

Which of the following technique is, in your opinion, more efficient to assess chronic exposure ? Give a score from 1 (less efficient) to 6 (more efficient)

Efficiency= capacity to give an assessment of reasonable accuracy (sufficient to make a decision in regards of risk acceptance) at the lowest investment cost (time, money, other resources…)

		less efficient					more efficient	don't know
17	**Assess exposure only on the basis of employee interview/ workplace visit (expert judgment)**	1	2	3	4	5	6	☐
18	**Measuring exposure (punctual, short term-measurement)**	1	2	3	4	5	6	☐
19	**Measuring exposure (direct reading or sampling during a significant part of the work activity)**	1	2	3	4	5	6	☐
20	**Biological monitoring**	1	2	3	4	5	6	☐
21	**Use of predictive mathematical and statistical model**	1	2	3	4	5	6	☐
22	**Making reference to literature/ existing exposure levels / state of the art (ex. good laboratory practices)**	1	2	3	4	5	6	☐
23	**Other (specify):**	1	2	3	4	5	6	☐

Which degree of overall reliability (reliability of data used and precision of the assessment method) do you associate to each method? Give a score from 1 (less precise) to 6 (more precise).

		less reliable					more reliable	don't know
24	**Assess exposure only on the basis of employee interview/ workplace visit (Expert judgment)**	1	2	3	4	5	6	☐
25	**Measuring exposure (punctual, short term-measurement)**	1	2	3	4	5	6	☐
26	**Measuring exposure (direct reading or sampling during a significant part of the work activity)**	1	2	3	4	5	6	☐
27	**Biological monitoring**	1	2	3	4	5	6	☐
28	**Use of predictive mathematical and statistical model**	1	2	3	4	5	6	☐
29	**Making reference to literature/ existing exposure levels / state of the art (ex. good laboratory practices)**	1	2	3	4	5	6	☐
30	**Other (specify):**	1	2	3	4	5	6	☐

C) Use of the expert judgement.

The following questions are intended to compare the importance of the parameters observed by the specialists to assess the exposure situation (chronic and subacute exposures), without any objective measurements or empirical or theoretical exposure models.

How often do you employ the following factors in order to characterize workplace environment during exposure judgment?

		Between **100-80%** of cases	Between **80-50%** of cases	Between **50-10%** of cases	Between **10-0%** of cases	**never**
31	**Room size and shape**	1	2	3	4	5
32	**Opening of doors or windows (natural ventilation)**	1	2	3	4	5
33	**Location of air inlets and exhausts points (forced ventilation)**	1	2	3	4	5
34	**Wind speed and wind direction within the room**	1	2	3	4	5
35	**Movement of people/objects in the room**	1	2	3	4	5
36	**Air temperature gradient in the room**	1	2	3	4	5
37	**Overall quantity emitted (rough mass balance)**	1	2	3	4	5
38	**Evaporation area (solvent)**	1	2	3	4	5
39	**Vapour pressure or boiling temperature (solvent)**	1	2	3	4	5
40	**Composition, dilution (solvent)**	1	2	3	4	5
41	**Presence of air jet at the source (a vector gas)**	1	2	3	4	5
42	**Emission process: grinding, spraying…**	1	2	3	4	5
43	**The method and degree of manual handling (agitation, stirring..)**	1	2	3	4	5
44	**Activity intensity**	1	2	3	4	5
45	**Activity frequency**	1	2	3	4	5
46	**The presence of personal protective equipment**	1	2	3	4	5
47	**Dustiness, general cleanness**	1	2	3	4	5
48	**Sensations (smell, irritation effects, ...)**	1	2	3	4	5

49 **Comparing the following factors, which of them, in your opinion, influence mainly occupational exposure? Give a score from 1 (less influence) to 6 (more influence).**

		less influence →					**more influence**	**don't know**
50	**Room size and shape**	1	2	3	4	5	6	☐

51 **Opening of doors of windows (natural ventilation)**	1	2	3	4	5	6	☐
52 **Location of air inlets and exhausts points (forced ventilation)**	1	2	3	4	5	6	☐
53 **Wind speed and wind direction within the room**	1	2	3	4	5	6	☐
54 **Movement of people/objects in the room**	1	2	3	4	5	6	☐
55 **Air temperature gradient in the room**	1	2	3	4	5	6	☐
56 **Overall quantity emitted (rough mass balance)**	1	2	3	4	5	6	☐
57 **Evaporation area (solvent)**	1	2	3	4	5	6	☐
58 **Vapour pressure or boiling temperature (solvent)**	1	2	3	4	5	6	☐
59 **Composition, dilution (solvent)**	1	2	3	4	5	6	☐
60 **Presence of air jet at the source (a vector gas)**	1	2	3	4	5	6	☐
61 **Emission process: grinding, spraying…**	1	2	3	4	5	6	☐
62 **The method and degree manual handling (agitation, stirring..)**	1	2	3	4	5	6	☐
63 **Activity intensity**	1	2	3	4	5	6	☐
64 **Activity frequency**	1	2	3	4	5	6	☐
65 **The presence of personal protective equipment**	1	2	3	4	5	6	☐
66 **Dustiness, general cleanness**	1	2	3	4	5	6	☐
67 **Sensations (smell, irritation effects, ...)**	1	2	3	4	5	6	☐

D) Quantitative exposure assessment: exposure parameters

The following questions are intended to understand the physic-chemical parameters used by practitioners during quantitative exposure assessment.

EMISSION ASSESSMENT

68 **Comparing the following chemical and physical parameters, which of them play, in your opinion, the most significant role in the emission phenomena? (Score from 1 to 6)**

AEROSOLS	**less significant**					**more significant**	**don't know**
69 **Molecular weight**	1	2	3	4	5	6	☐
70 **Particle size and distribution**	1	2	3	4	5	6	☐
71 **Air temperature**	1	2	3	4	5	6	☐
72 **Particle shape**	1	2	3	4	5	6	☐
73 **Kinematics viscosity**	1	2	3	4	5	6	☐
74 **Air speed and direction at the**	1	2	3	4	5	6	☐

	source							
75	**Cohesion force**	1	2	3	4	5	6	☐
76	**Separation force in relation with the emission process (grinding, air jet pressure…)**	1	2	3	4	5	6	☐
77	**Other (specify):**	1	2	3	4	5	6	☐

SOLVENTS		less significant					more significant	don't know
78	**Molecular weight**	1	2	3	4	5	6	☐
79	**Vapour pressure**	1	2	3	4	5	6	☐
80	**Air temperature**	1	2	3	4	5	6	☐
81	**Molecular diffusivity**	1	2	3	4	5	6	☐
82	**Air speed and direction at the source**	1	2	3	4	5	6	☐
83	**Evaporating surface (area)**	1	2	3	4	5	6	☐
84	**Evaporating surface (agitation, stirring)**	1	2	3	4	5	6	☐
85	**Other (specify):**	1	2	3	4	5	6	☐

DISPERSION ASSESSMENT

In your opinion, what is the importance of the following parameters during the transfer from the source to breathing zone air? Give a score (from 1 to 6)

		less important					more important	don't know
86	**Air speed and direction in the room (general air movements)**	1	2	3	4	5	6	☐
87	**Air speed and direction near the source (local air movements)**	1	2	3	4	5	6	☐
88	**Presence of air jet (a vector gas)**	1	2	3	4	5	6	☐
89	**Room size and shape**	1	2	3	4	5	6	☐
90	**Air temperature changes with Height or across the room**	1	2	3	4	5	6	☐
91	**Molecular diffusivity**	1	2	3	4	5	6	☐
92	**Particles size and shape (for aerosols)**	1	2	3	4	5	6	☐
93	**Other (specify):**	1	2	3	4	5	6	☐

E) Quantitative exposure assessment: exposure models

The following questions are designed to understand the level of use of the emission and dispersion models, as well as reliability and effectiveness perceived.

94 **Have you employed any exposure model to assess occupational exposure?**

Yes ☐ 1

No ☐ 2

If you've rarely or never used exposure model, why? (Several answers are possible)

95 **You don't know any exposure model** ☐

96 **You feel that estimation outputs are not accurate and precise** ☐

97 **Too time consuming** ☐

98 **You find difficult to place real-life work situation in terms of model parameters** ☐

99 **Other reasons (specify):** ☐

Answer the following question if you use or have used exposure empirical or theoretical models.

<u>POLLUTANTS' GENERATION RATE MODELS</u>

a. If you use exposure models, how do you usually identify the generation rate?

	Between **100-80%** of cases	Between **80-50%** of cases	Between **50-10%** of cases	Between **10- 0%** of cases	**never**
100 **Making reference to literature/ existing standard**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
101 **Estimate through a mass balance (assess the mass of product release)**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
102 **Measuring exhaust air concentration**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
103 **Specific emission model**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
104 **Other (specify):**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5

b. Do you know the following generation rate models?
c. And how often do you apply them?

	b				**c**		
	YES	**NO**	Between **100-80%** of cases	Between **80-50%** of cases	Between **50-10%** of cases	Between **10- 0%** of cases	**never**
105 **Saturation vapeur pressure model (SVP)**	☐	☐	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
107 **Back pressure model**	☐	☐	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5
109 **Evaporation rate from flat surface**	☐	☐	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5

111	**Drum-filling models**	☐	☐	1	2	3	4	5
113	**Exponentially decreasing emission rate**	☐	☐	1	2	3	4	5
115	**Other (specify):**	☐	☐	1	2	3	4	5

POLLUTANTS' DISPERSION MODELS

a) Which of the following exposure models do you know?
b) And in how many cases do you apply them?

		a				**b**		
		YES	**NO**	Between **100-80%** of cases	Between **80-50%** of cases	Between **50-10%** of cases	Between **10- 0%** of cases	**never**
	Deterministic Mass Balance Model:							
117	Ideal Mixed Model	☐	☐	1	2	3	4	5
119	Two Zone Model	☐	☐	1	2	3	4	5
121	Eddy Diffusion Model	☐	☐	1	2	3	4	5
123	Gaussian plume dispersion model	☐	☐	1	2	3	4	5
125	Computational fluid dynamic	☐	☐	1	2	3	4	5
	Empirical Model:							
127	Job exposure matrix, etc....	☐	☐	1	2	3	4	5
129	EASE	☐	☐	1	2	3	4	5
	Others							
131	(i.e. EPA's tools: ChemSTEER, MCCEM, WPEM), specify:	☐	☐	1	2	3	4	5

Give your degree of satisfaction (from 1 to 6) for all the models you have used, considering respective level of efficiency and accuracy.

		less efficient →					**more efficient**
	Deterministic Mass Balance Model:						
132	Ideal Mixed Model	1	2	3	4	5	6
133	Two Zone Model	1	2	3	4	5	6
134	Eddy Diffusion Model	1	2	3	4	5	6
135	Gaussian plume dispersion model	1	2	3	4	5	6
136	Computational fluid dynamic	1	2	3	4	5	6
	Empirical Model:						
137	Job exposure matrix, etc....	1	2	3	4	5	6
138	EASE	1	2	3	4	5	6
	Others						
139	(i.e. EPA's tools: ChemSTEER, MCCEM, WPEM), specify:	1	2	3	4	5	6

		less accurate					more accurate
	Deterministic Mass Balance Model:						
140	Ideal Mixed Model	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6
141	Two Zone Model	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6
142	Eddy Diffusion Model	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6
143	Gaussian plume dispersion model	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6
144	Computational fluid dynamic	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6
	Empirical Model:						
145	Job exposure matrix, etc....	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6
146	EASE	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6
	Others						
147	(i.e. EPA's tools: ChemSTEER, MCCEM, WPEM), specify:	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6

In a mathematical model based on the mass balance of a substance, which of the following hypothesis may be assumed, in your opinion, without an important loss of accuracy? (score from 1 to 6)

		negligible loss of precision					severe loss of precision	don't know
148	**The incoming air is contaminant-free**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
149	**The generation and ventilation rates are constant over time**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
150	**Room air and ventilation air mix ideally**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
151	**The concentration approaches the equilibrium concentration**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
152	**Uniform concentration throughout the room**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
153	**Extinction of the chemical (by adsorption/ deposition on walls and equipment) chemical transformation, condensation of hot vapours, are negligible**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
154	**The effect of gravity on fine aerosol is negligible**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
155	**Exposure close to the source is unaffected by local stirring, worker position (ideal mix in the near field)**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐

For which of following substances or specific scenarios, have you found exposure models inappropriate? Give a score of satisfaction (from 1 to 6)

		less adequate →					more appropriate	don't know
	SUBSTANCE							
156	**Mixtures of solvents**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
157	**Liquid with very low vapour pressure**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
158	**Hot fumes (such as Welding fumes)**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
159	**Sprays Aerosols (with a vector gas)**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
160	**Other (specify):**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
	SPECIFIC SCENARIO							
161	**Emission during application phase**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
162	**Presence of multiple sources**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
163	**Passive emission (not directly associated with the process, e.g. re-suspension of settled dust)**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
164	**Irregular or not homogenous way of handling**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐
165	**Other (specify):**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6	☐

166 **In these particular cases, do you think is it necessary to develop other models easier to use?**

yes	☐ 1
not	☐ 2

167 **If not, why?**

In your opinion, which could be future improvements to the exposure models? (Give a score from 1to 6)

		less useful →					more useful
168	**Integrate more emission parameters in the dispersion models**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6
169	**Use more factors easily accessible field investigation**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6
170	**Take more into account local phenomena of dispersion (close to the source).**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6
171	**Other (specify)**	☐ 1	☐ 2	☐ 3	☐ 4	☐ 5	☐ 6

Annex II

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Modélisation de l'exposition au CO

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Stratégie d'évaluation de l'exposition

- **mesure:** fiable et objective, mais la validité de ces résultats est souvent limitée à la seule période de mesure.
- **jugement d'expert:** basé sur l'interprétation subjective des observations et des entrevues avec des employés, reste une méthode de faible précision.
- **modèle:** peu précis mais prédictif, résultats cohérents si choix de paramètres judicieux.

Objectifs

Des modèles prédictifs ont été utilisés dans le contexte de l'exposition au CO afin d'estimer les expositions prévisibles dans différentes situations professionnelles.

L'objectif était aussi de tester la flexibilité et la précision des modèles classiques d'exposition dans des situations d'exposition concrètes.

Méthode

Différentes situations d'exposition professionnelle au monoxyde de carbone ont été choisies: garage automobile, centre de karting, utilisation des tronçonneuses en extérieur.

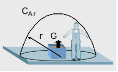
Pour chaque situation, différents scénarios d'émission et de ventilation ont été imaginés et intégrés dans des modèles d'exposition les plus adaptés.

Les profils de concentration calculés avec les modèles ont ensuite été comparés aux niveaux d'exposition rapportés par la littérature pour des situations similaires.

MODELES D'EXPOSITION:

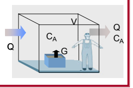
Modèle idéalement mélangé

$$V\cdot\frac{dC_A}{dt}=+G-Q\cdot C_A$$



Modèle de diffusion

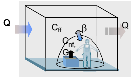
$$C_r=\frac{G}{4\cdot\pi\cdot D\cdot r}\cdot\left[1-erf\left(\frac{r}{\sqrt{4\cdot D\cdot t}}\right)\right]$$



Modèle à 2 compartiments

Near field: $V_{NF}\cdot\frac{dC_{A,NF}}{dt}=G\cdot+\beta\cdot C_{FF}\cdot-\beta\cdot C_{NF}$

Far field: $V_{FF}\cdot\frac{dC_{A,FF}}{dt}=\beta\cdot C_{NF}\cdot-(\beta+Q)\cdot C_{FF}$



SITUATIONS

Garage: - Volume de la pièce: 600 m^3 ,
- Renouvellement d'air: 10 h^{-1},
- Emission: démarrage à froid 50 g CO/dem.,
+ émission à chaud 80 g/h (10 Km/h)

Scénario
1. Déplacement d'une voiture sur un lift.
2. Déplacement simultané de 3 voitures
3. Déplacement en série de plusieurs voitures

Karting:

- Facteur d'émission:	15	g/km
- Vitesse moyenne	45	Km/h
- Volume de la halle:	2100	m^3
- Renouvellement d'air:	5	h^{-1}
- Max karts roulant	14	

Scénario
1. Normal
2. Basse saison
3. Haute saison

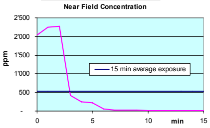
Tronçonneuses:

- type de moteur:	2 temps
- puissance:	2.3 kW
- facteur d'émission:	515 g CO/h
- exposition en extérieur	

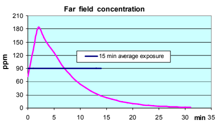
Scénario
1. Émission constante
2. Émission pulsée
3. Exposition à différentes distance de la source

Résultats: Garage, Modèle à 2 compartiments

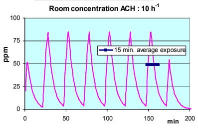
Scénario 1



Scénario 2



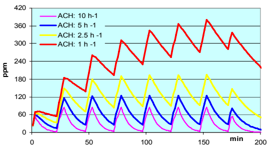
Scénario 3



	Moyenne 15 min ppm		Maximum ppm		Moyenne ppm
Scén.	NF	FF			
1	538	30	2300	60	
2		90		184	
3		50		85	28
Litterature I			**70 ; 900**		**14; 221**
II					**1;19;50**
III			**57; 120**		**31; 84**

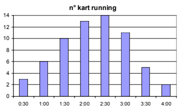
I) IST, Suisse II) Bundesanstalt fürArbeitsschutz und Arbeitsmedizin
III) Athens University

analyse de sensibilité pour différentes ventilations

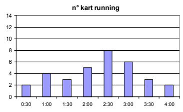


Résultats: karting, Modèle idéalement mélangé

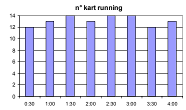
Scénario 1



Scénario 2

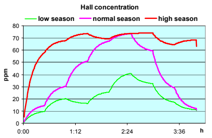


Scénario 3



Scén.	Moyenne ppm	Moyenne 15 min. ppm	Maximum ppm
1	42	74	74
2	22	40	41
3	66	73	74
Litterature I	**> 30**		**> 50**
II	**45**		**85**

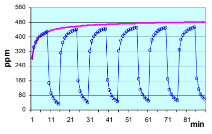
I) SECO, Suisse
II) Insitut national de santé publique du Québec



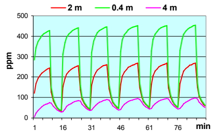
Résultats: tronçonneuses, Modèle de diffusion

Scénario 1

Scénario 2



Scénario 3



Pour les situations d'exposition à l'extérieur, le modèle de diffusion s'adapte mieux que les modèles à compartiments, en plus ce modèle permet d'estimer la concentration en fonction de la distance de la source.

Scén.	Moyenne ppm	Moyenne 15 min ppm	Maximum ppm
1	460	481	481
2	284	288	453
3	65; 171; 284	71; 169; 288	97; 268; 453
Litterature I	**16, 24**		**> 500**
II			**504**

I) Biinger et al, Am. Ind. Hyg. Ass. J. 58, [1997]
II) NIOSH

CONCLUSIONS

Les modèles représentent un outil intéressant de prévision de l'exposition lorsque l'estimation des facteurs d'émission et des paramètres de ventilation est réalisable. Bien qu'ils soient sensiblement moins précis que les mesures directes pour évaluer l'exposition sur une période donnée, ils présentent des avantages importants en terme d'analyse de sensibilité, de gamme de scénarios accessibles et de possibilités d'évaluation prospectives et rétrospectives. En ce sens, ils présentent une excellente complémentarité avec la métrologie.