A Simple Small Scale Method to Determine the Re-suspension Rate for Non-fixed Surface Contamination

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ABSTRACT

In the frame of the Co-ordinated Research Program of IAEA “Radiological aspects of package and conveyance non-fixed contamination” it has been identified that there is a lack of knowledge concerning the airborne release from surfaces with non-fixed contamination of radioactive materials during normal transport conditions. This has a direct effect on the models and parameters underlying the non-fixed contamination limits for packages and conveyances.

We have developed a simple and practicable methodology to investigate the suspension of non-fixed particle bound contamination from surfaces due to effects such as wind and vibration affecting packages during normal transport conditions. This method is based on the formation of a defined contamination layer on small test surfaces by bringing them into either liquid or airborne suspensions of surrogate particles. A time and particle size resolved detection technique covering the particle size range between 1 and 100 µm is employed to measure the particle re-suspension rate from the test surface exposed to controlled hydrodynamic and/or vibrational forces inside a narrow flow channel (4 mm channel height). All particles suspended from the test surface and incorporated into the airflow are size classified and counted by a clean room particle counter.

The re-suspension process is recorded in detail in order to distinguish between short term and long term effects as well as to perform statistical analysis of the re-suspension pattern enabled by the possibility of detecting single re-suspension events. For particles in the respirable size range the detection limit for the re-suspension rate is 10^{-6} [1/h]. The detection limit is determined by the maximum tolerable particle coverage of the investigated surface and decreases for larger particle diameters. The method is very versatile and allows for easy variation of particle and surface properties.

Measurements made with various test samples reveal consistent results which show an initially increased re-suspension rate as a short term effect followed by a much reduced long term rate. The long term re-suspension rate increases with flow velocity and decreases with particle size. For a high flow velocity of 30 m/s corresponding to a wall shear stress of 1 [N/m²] the long term re-suspension rate for 4 µm alumina particles deposited from the airborne state onto a smooth glass surface is 1 10^{-4} [1/h] with an estimated reproducibility of a factor of 2. The same particles deposited from an aqueous suspension show a re-suspension rate of at least a factor of 10 lower magnitude due to the remaining liquid forces between particles and surface even after overnight drying of the test samples. This causes also a considerably larger variation of results obtained in repeated experiments.

INTRODUCTION

There is a need for reliable data on the airborne release of radioactive materials in various transport conditions and for different modes of transport with their specific boundary conditions. Among this is the release of contamination from surfaces of transport casks and conveyances. This contamination usually occurs as a result of the transfer of radioactive material from areas in which these packages and conveyances are handled. Two types of surface contamination of radioactive material are distinguished, namely fixed contamination and non-fixed contamination. For practical purposes a distinction is made between contamination which, during routine conditions of transport, remains in situ (i.e. fixed contamination) and, therefore, cannot give rise to hazards from ingestion, inhalation or spreading, and non-fixed contamination which may contribute to these hazards. Non-fixed contamination appears mainly as particlulate matter which eventually can be released when exposed to hydrodynamic and vibrational forces. In the frame of the Co-ordinated Research Program of IAEA “Radiological aspects of package and conveyance non-fixed contamination” it has been identified that there is a lack of knowledge concerning the airborne release from surfaces with non-fixed contamination of radioactive materials during normal transport conditions. This has a direct effect on the models and parameters underlying the non-fixed contamination limits for packages and conveyances.

In describing the results of entrainment experiments or modelling the resuspension of non fixed contamination, there are two alternative terms used to characterise the transfer of particles from the contaminated surface to the surrounding air:

- Resuspension factor, F_{susp}, is defined as the ratio between the airborne concentration of a pollutant per cubic meter directly over a contaminated surface and the surface contamination. This definition needs detailed descriptions of the conditions (package type, room size, air exchange rate etc.). The unit is 1/m derived from Bq/m^3 (or number of particles per m^3) per Bq/m^2 (number of particles per m^2).
- Resuspension rate, T_{susp}, is given in h^{-1} derived from Bq/h (number of released particles per h per Bq (number of particles on the surface), and describes the percentage of activity present on a surface which is suspended from the surface into the surrounding air per hour. It mainly depends on the properties of the surface, the interaction between the contaminants on the surface and the forces acting on the particulates. It is independent of the size of the room, i.e. the volume of air into which resuspension occurs .

For an indoor situation the correlation between resuspension factor and resuspension rate is given by

$$F_{susp} = \frac{T_{susp} \times S}{V \times f_{ex}}$$

where S is the surface area of the package from which resuspension occurs, V is the room volume, f_{ex} is the air exchange rate of the room. When focussing on the resuspension risk of contaminated surfaces under various indoor and outdoor situations and creating data to be used in risk models the resuspension factor is less transparent than the resuspension rate. For this reason, in the following, we only consider the resuspension rate.

Numerous theoretical and experimental investigations exist in the literature on various aspects of the resuspension of sparse particles from the surface of substrates. Resuspension is influenced by many parameters determining the interaction between the particles and the surface i.e. adhesion and detachment, as well as the boundary layer flow over the contaminated surfaces and the action of relevant forces on the particles. Unless the boundary conditions are exactly specfied in all aspects, the results of theoretical studies as well as experimental results from the literature are of limited use for the practical problem of the resuspension of non-fixed contamination.

In this study, it was our aim to develop a simple and practicable small scale methodology to investigate the suspension of non-fixed particle bound contamination from surfaces due to effects such as wind and vibration affecting packages during normal transport conditions. This method should allow for

- simulating realistic routes of contamination of model surfaces with surrogate particles,
- applying well defined hydrodynamic and vibrational forces to the test surface and
- recording the complete time dependent resuspension processs with sufficient sensitivity.

DESIGN CONSIDERATIONS

It is well accepted in the literature, that for flow induced resuspension the controlling parameter for the hydrodynamic force is the wall shear stress, τ_0, of the flow also quantified by the so called friction velocity

$$u^* = \sqrt{\tau_0 / \rho}\ . \tag{1}$$

Here, ρ is the density of the air. For a turbulent flow between parallel plates the friction velocity is given by:

$$u^* = U\sqrt{\frac{f}{2}}$$

$$f = 0.32 \ \ \mathrm{Re}^{-1/4} \qquad (2)$$

where U is the flow velocity in the turbulent core and Re is the flow Reynolds number. This model can also be a first approximation of an airflow between the cooling fins of a transport cask. For other simple flow scenarios such as a free flow over a flat surface the friction velocity can be determined from boundary layer theory. For flows over complex structures the local surface shear stress has to be calculated using computanional fluid dynamics (CFD) models. Many experiments on resuspension have been carried out with spherical particles deposited on smooth surfaces. According to Reeks and Hall [1] a friction velocity of the order of 1 m/s is required to re-suspend 20 µm alumina spheres from a smooth steel surface. For rough surfaces the minimum friction velocity can be much larger than 1 m/s. Our set-up was designed to simulate wall friction velocities up to 4 m/s which is expected to cover the range associated with the surface flow conditions under normal transport conditions.

Surface vibrations cause particle detachment by the inertial force acting on the particle due to its acceleration. For harmonic oscillations, the maximum acceleration, a, is given by

$$a = A(2\pi \ \nu)^2$$

where A is the amplitude of the vibrations and ν is the vibrational frequency. Assuming an adhesion force of 10^{-6} N for a 10 µm particle attached to a smooth surface and a particle density of 4000 kg/m³ an acceleration of 5 10^5 m/s² is needed for particle detachment which would require an oscillation amplitude of 125 µm for a frequency of 10 kHz. Whether these conditions occur on the surface of casks during normal transports or for relevant accident scenarios is not known from the literature.

Resuspension processes show a characteristic time profile: an initially increased resuspension rate as a short term effect followed by a much reduced long term rate. The time scale is counted from the time point where the threshold conditions for resuspension occurs. Since, under realistic scenarios, these events cannot be predicted, short term as well as long term reuspension effects are important to be analysed. The experimental set-up should therefore be able to distinguish between these two time scales. This is only possible if an on-line method is employed for measuring particle resuspension from surfaces.

The detection limit of the set-up should allow measurements of resuspension rates at least as low as 10^{-5} [1/h]. These values were obtained in a recently conducted French study on resuspension of surfaces contaminated in the spent fuel storage pool of a power plant [2].

RESUSPENSION APPARATUS

The experimental approach of the methodology is based on counting and particle size classifying all, or at least a substantial fraction of the particles resuspended from a contaminated test surface. The resuspension flux given in particles/sec is followed in time and normalised to the number of particles initially deposited on the surface. This number has to be measured at the beginning of the resuspension experiment. From these two quantities the resuspension rate can be calculated. In order to control and exactly quantify the experimental boundary conditions a channel flow resuspension apparatus is used. A high throughput particle counter (Type LASAIR, PMT, Karlsruhe, Germany) with a flow rate of 28 l/min is used for particle detection. It classifies the particles in 8 size classes: 1-3, 3-5, 5-10, 10-20, 20-30, 30-50, 50-100, >100 µm, and, thus covers the particle size range affected by resuspension and which is relevant for the analysis of radiological consequences. The dimensions of the channel are chosen in order to generate sufficiently large surface shear stresses and to meet the sampling flow rate of the particle counter. In addition the number of particles deposited as a sub-monolayer on the test surface must be high enough to allow for total particle counts for an accurate measurement of resuspension rates of the order of 10^{-5} [1/h]. Based on this, the following design is chosen for the flow channel: height 4 mm, width 10 mm and length 76 mm.

Vibrations are induced into the channel by exposing it to ultrasound waves generated by a sonotrode (Type UIP 250, Dr. Hielscher). This device operates at 24 kHz and delivers ultrasound waves with a power of 200 W. No direct contact between sonotrode and channel is necessary. A g-sensor (Type ADXL 105 Analog Devices) is used to monitor frequency and amplitude of the vibrations. Variations of the accelerations were achieved by changing the output power of the sonotrode and varying the distance between sonotrode and channel surface. In the resuspension experiments in context with non-fixed surface contamination the channel is operated in a vertical orientation. The experimental set-up is shown schematically in Fig. 1.

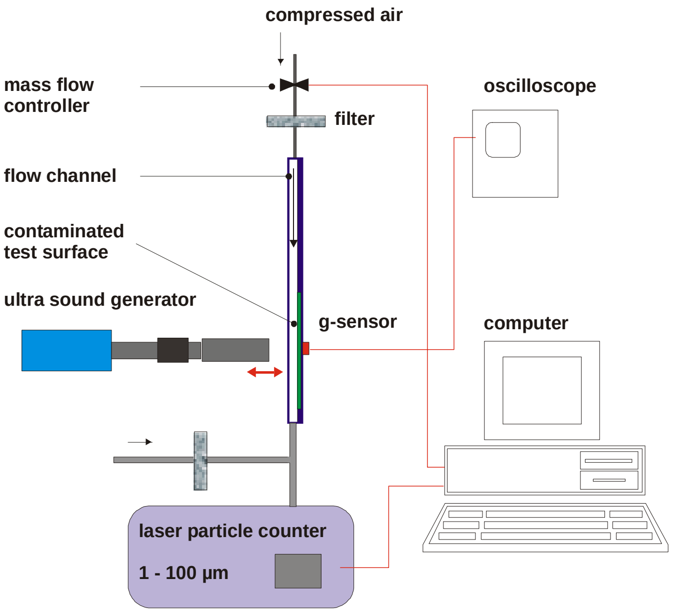

Fig. 1: Experimental set-up to measure particle resuspension from contaminated surfaces

The resuspension channel is made of two brass plates that can be firmly screwed together. One plate contains the flow channel (1 cm wide), the other one an indention (2 cm by 7.6 cm) to incorporate a thin plate with the contaminated test surface (Fig. 2). This set-up allows for easy variation of the test specimen in terms of the properties of the surface as well as the contamination.

SURROGATE CONTAMINATION

Two types of surface contamination methods were developed: particle settling in air, and particle settling in water. In order to achieve a homogeneous deposition layer on the test surfaces a settling chamber of 1.2 m height is used which was either empty or filled with water. Several test surfaces are placed at the bottom of the chamber. A pre-weighed amount of powder is introduced into the top section of the chamber. For the air settling chamber this is done by using an air driven dispersion nozzle in combination with a baffle plate for homogenisation of the aerosol cloud. After a fixed period of time (the settling time of the powder) the box is opened and the contaminated test surfaces are removed. For the liquid tank, a small amount of highly concentrated aqueous powder suspension is introduced into the water and is homogenised by a stirrer. At the end of the settling time the tank is drained and the surfaces are removed from the tank. For complete and homogeneous flushing of the surface a small quantity of surfactant has to be added to the water.

Several types of mineral dusts are used for contamination. In the demonstration tests we focused on two MIRA (Motor Industry Research Association) alumina dust grades, one covering the size range between 3 and 6 µm geometric diameter (MIRA Superfine) and a second one with particles in the range around 30 µm (MIRA G4) (see Fig. 3). The amount of powder used for contamination is selected in such a way that a submonolayer of contamination of the order of 50 % surface coverage which is about $2{\cdot}10^6$ particles/cm² for a particle size of 5 µm and $7{\cdot}10^4$ particle/cm² for the 30 µm particles results. The rationale behind this choice is to have a large particle number exposed to the resuspension forces in order to make the detection method as sensitive as possible for the low expected resuspension rate. At the same time we have to mimic the realistic situation of the spareness of the surface contamination and thus must avoid secondary resuspension effects such as saltation, the likelihood of which strongly increases at high contamination densities. Eight test surfaces of the size of a microscope sample glass can be contaminated simultaneously (wet and dry). The variation of the loadings between the single samples is less than 10% so that only one sample of the group has to be analysed for the initial particle number density which is needed for the evaluation of the resuspension measurement.

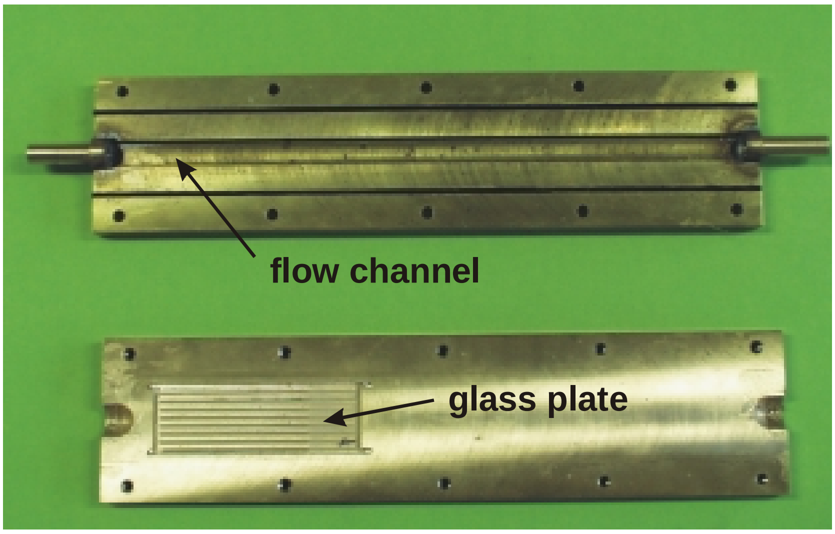

Fig. 2: Details of the resuspension channel. The flow is going from right to left. The flow is developing to a fully developed turbulent flow before it passes over the glass plate. Only particles in a strip of the width of the flow channel can be resuspended, altough the entire test surface (glass plate) is contaminated.

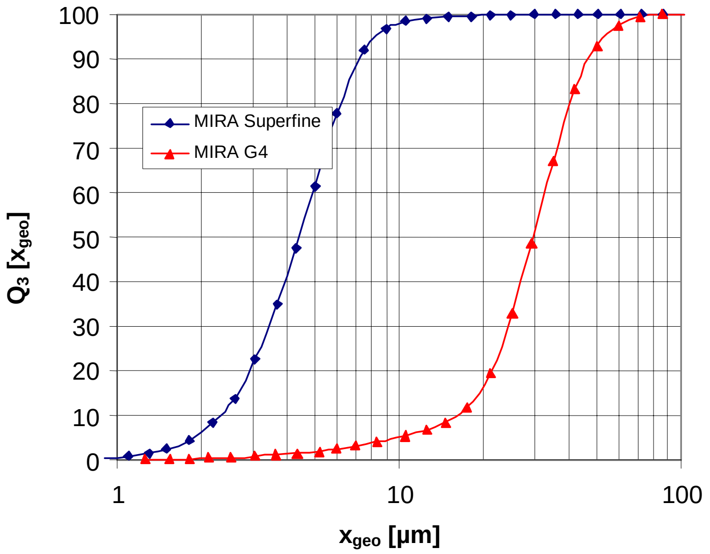

Fig. 3: Examples of the size distribution of two different particle systems for the simulation of non-fixed contamination.

DEMONSTRATION TESTS

Wind induced resuspension.

For measuring the wind induced resuspension the experimental protocol consists of two phases covering the short term, respectively the long term resuspension period. The short-term phase is treated separately because it is expected that the majority of the loosely bonded particles are resuspended during this phase. In order to detect the long term resuspension correctly it is necessary to remove these particles before measuring. While the flow velocity in the resuspension channel is increased linearly from zero to a final pre-set value, for example 30 m/s, and kept constant thereafter, the concentration of the resupended particles is measured continuously. At the beginning of each phase the total particle load on the test surface is measured by microscopic analysis.

A detailed look on the resuspension of particles in the 3-6 µm range is shown in Fig. 4. The blue curves represent the count rates in the relevant size range. The green curves are the velocity ramps. Obviously, resuspension starts at a velocity of 5 m/s corresponding to a friction velocity of 0.8 m/s. At the end of the first test period (at 4500 s) the sample was removed and a total of 10^7 particles was detected on the wind exposed surface. The long term resuspension rate measured in this experiment is $1.1 \cdot 10^{-4}$ [1/h]. It is obtained by deviding the count rate by the total number of surface particles and represents the average value over the time period indicated by the black bar. The value is based on appr. 9000 counts. We did not observe a temporal trend in long term resuspension rate. The peaks indicated in red are associated with the channels of the laser particle counter where no particles are expected to be found.

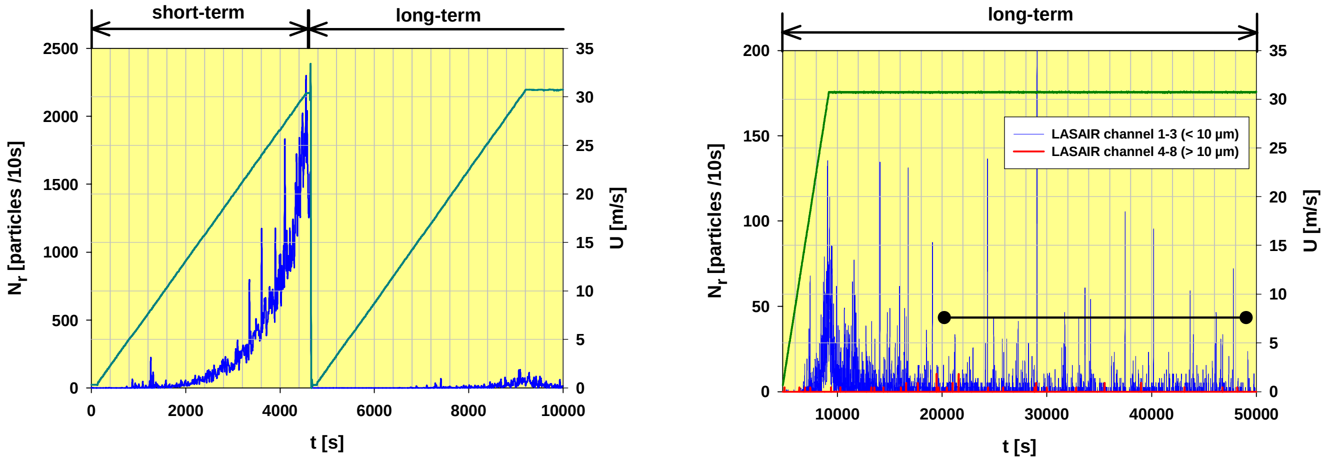

Fig. 4: Count Rates (left y-axes) of the LASAIR measured during a resuspension experiment with a glass surface contaminated with MIRA superfine particles (3-6 µm) by air settling. The velocity values are shown on the right y-axes.

Following this prolocol a small series of demonstration tests was conducted. The results are summarised in Table 1. The experiments show that particle resuspension from air contaminated surfaces is controlled by particle size and air velocity. Repeated experiments give results which are reproducible within a factor of 2. When the contamination is performed in water the resuspension rates are significantly smaller but associated with a much larger scatter. The results were not always as expected. Sometimes a smaller long term resuspension rate was measured for higher values of the air velocity. We attribute this effect to the fact that for liquid settling the adhesion forces between the particles and the surface are subject to substantial variation presumably due to the surfactant material remaining after the drying process. The error attributed to the counting statistics was in all cases smaller than 15%.

The demonstration tests show that the experimental system as part of the overall methodology allows for a quantitative analysis of the particle resuspension process induced by hydrodynamic forces. For the scenario of wind resuspension of particles from surfaces of transport casks during transport only the long term resuspension rate should be of interest since the surface has been cleaned beforehand using high velocity water jets inducing much higher resuspension forces on the contamination as expected from the airflow over the surface associated with the rail or road transport.

Table 1: Results of demonstration tests on wind resuspension of non-fixed surface contamination settled in air or water

U [m/s]	Resuspension rate [1/h]			
	3-6 µm		20-40 µm	
	air	water	air	water
10 [m/s]	2.9E-05	1.2E-05	8.0E-04	1.7E-04
30 [m/s]	1.1E-04	3.1E-05	1.2E-03	1.0E-05

Resuspension by vibrations

For vibration induced resuspension the situation is different. Short term resuspension bursts are important to be detected due to the possibility of shocks induced into the transport casks. These shocks may create surface vibrations associated with accelerations high enough for resuspension of contamination to occur. As demonstration tests we carried out a series of experiments using the MIRA G4 powder according to the following protocol: The contaminated surface was first exposed to an airflow of 21 m/s for a period of 5 minutes. The airflow was then reduced to 10 m/s. After an additional time period of 2 minutes the surface activation was turned on and the flux of the resuspended particles was measured over a time period of 10 minutes.

A relative measure of the acceleration amplitude was taken from the oscilloscope recording the signals of the g-sensor. The maximum achievable acceleration was set equal to 1. (An absolute value of the acceleration could not be recorded due to limited performance of the g-sensor available to us throughout the study.) Five tests were performed with relative surface accelerations in the range between 0 and 1. Fig. 5 shows the temporal pattern of the resuspension rate. Starting with virtually zero resuspension when only the air is flowing over the surface there is an initial burst of particles after turning on the vibrations and a levelling off of the curve after about 300 s. This constant value has a strong (exponential) dependence on the vibrational amplitude as demonstrated in Fig. 6.

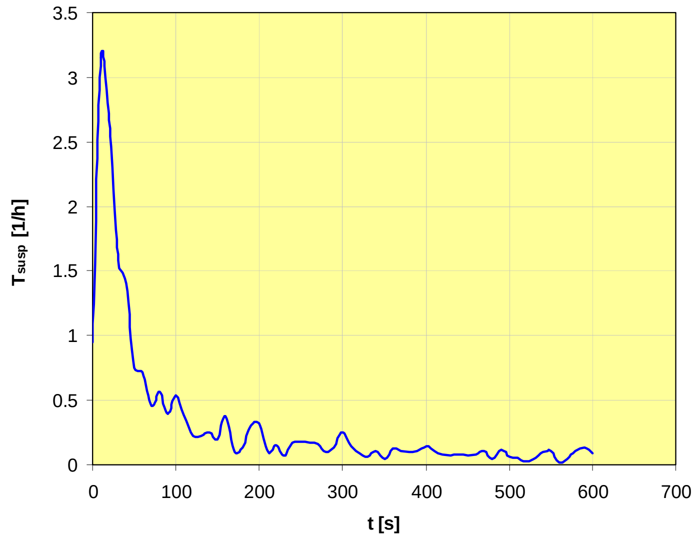

Fig. 5: Resuspension rate due to vibrations (largest amplitude)

The demonstration tests give consistent results. The experimental set-up is very promising since it allows at least in principle a good control on the acceleration. Improvements need to be made in view of aligning the ultrasound generator and the test channel, controlling the exact distance between generator and test channel, as well as measuring the acceleration by using a better g-sensor. This sensor could also be used to characterise the vibrational spectra of surfaces of transport casks, conveyors etc. under realistic transport and accident situations. These conditions could then be simulated easily in the experimental set-up.

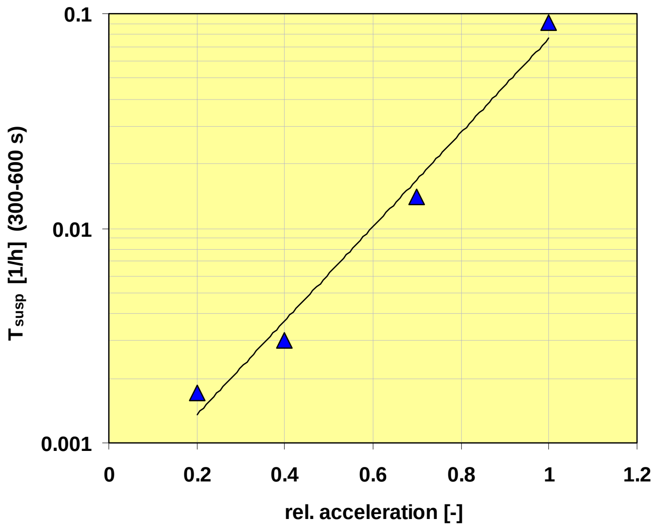

Fig. 6: Vibration induced long term resuspension rate as function of amplitude (acceleration). Frequency 25 kHz

CONCLUSIONS

A simple laboratory test method for measuring the flow and vibration induced resuspension rate of non-fixed surface contamination has been developed. The novelty of this method compared to existing methods is the time and size resolved counting of all suspended particles using a clean room particle counter. This enables a rather low detection limit for the resuspension rate of 10^{-6} [1/h] for particles < 10 µm, respectively, 10^{-5} [1/h] for particles in the size range between 10 and 30 µm. The method allows for quick variation of surface and particle properties. It can therefore be employed to simulate various types of packaging surface contamination and resuspension scenarios occuring under transport conditions. By measuring the resuspension rate instead of the resuspension factor scaling to surfaces larger than the test surface is easily possible. The small scale test results can therefore be directly used in the respective risk assessment models.

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