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Testing the performance of one and two box models as tools for risk assessment of particle exposure during packing of inorganic fertilizer

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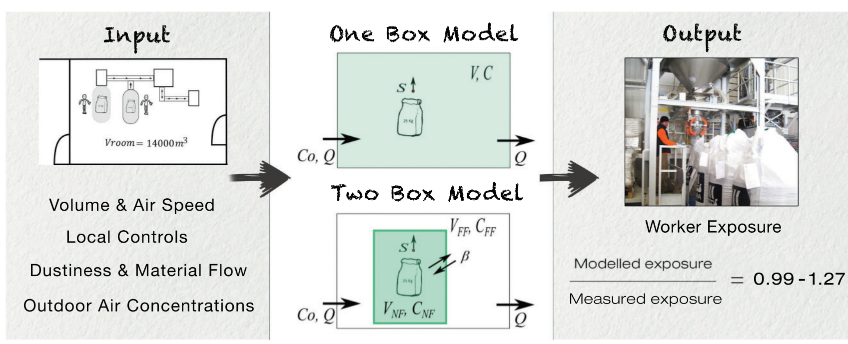
HIGHLIGHTS

- Occupational exposure to particles during industrial packing was assessed.
- No significant increases were found during packing of a granulate fertilizer.
- One and two box models predicted adequately actual worker exposure.
- Including outdoor concentrations in models was seen to improve their performance.
- Models parametrization was seen to be a key issue to adequately predict exposure.

GRAPHICAL ABSTRACT



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ABSTRACT

Modelling of particle exposure is a useful tool for preliminary exposure assessment in workplaces with low and high exposure concentrations. However, actual exposure measurements are needed to assess models reliability. Worker exposure was monitored during packing of an inorganic granulate fertilizer at industrial scale using small and big bags. Particle concentrations were modelled with one and two box models, where the emission source was estimated with the fertilizer's dustiness index. The exposure levels were used to calculate inhaled dose rates and test accuracy of the exposure modellings. The particle number concentrations were measured from worker area by using a mobility and optical particle sizer which were used to calculate surface area and mass concentrations. The concentrations in the worker area during pre-activity ranged 63,797–81,073 cm^{-3}, 4.6 × 10^6 to 7.5 × 10^6 μm^2 cm^{-3}, and 354 to 634 μg m^{-3} (respirable mass fraction) and during packing 50,300 to 85,949 cm^{-3}, 4.3 × 10^6 to 7.6 × 10^6 μm^2 cm^{-3}, and 279 to 668 μg m^{-3} (respirable mass fraction). Thus, the packing process did not significantly increase the exposure levels. Chemical exposure was also under control based on REACH standards. The particle surface area deposition rate in respiratory tract was up to 7.6 × 10^6 μm^2 min^{-1} during packing, with 52%–61% of deposition occurring in the alveolar region. Ratios of the modelled and measured concentrations were 0.98 ± 0.19 and 0.84 ± 0.12 for small and big bags, respectively, when using the one box model, and 0.88 ± 0.25 and 0.82 ± 0.12, when using the two box model. The modelling precision improved for both models when outdoor particle concentrations were included. This study shows that exposure concentrations in a low emission industrial scenario, e.g.

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during packing of a fertilizer, can be predicted with a reasonable accuracy by using the concept of dustiness and mass balance models.

1. Introduction

Industrial bag filling, packing and pouring processes have been pointed out as activities with high potential to emit airborne particles. Studies in different industrial sectors had reported from very low to high levels of worker exposure to particles, e.g.; during pouring and packing of paint pigments, packing of TiO_2, carbon black, fullerenes and carbon nanofibers (Ding et al., 2017; Fujitani et al., 2008; Koivisto et al., 2015, 2012a; Koponen et al., 2015; Kuhlbusch et al., 2004; Evans et al., 2010) as well as packing and pouring of cement materials (Notø et al., 2018; Peters et al., 2008). Additionally, differences in particle release have been observed when pouring different materials, different amounts, and using different types of mixing tanks (Koponen et al., 2015). Thus, every case is specific and further research is needed in order to understand emission patterns during packing and pouring.

Exposure to particulate matter (PM) is known to cause various adverse health effects, such as respiratory and cardiovascular diseases (Landrigan et al., 2017). Current epidemiological and toxicological studies consider $PM_{2.5}$ (with aerodynamic particle diameter $D_p \leq 2.5$ μm) as the most harmful component for human health (Gakidou et al., 2017; Landrigan et al., 2017; World Health Organization, 2016). Inhalation by humans of dust from inorganic complex fertilizers, which are the object of the present study, results in health effects which might be detected especially after long-term exposures. Inorganic complex fertilizers generally contain basic nutrients (nitrogen, phosphorus, potassium) as well as secondary and micronutrients (calcium, magnesium, boron, manganese) (Roy et al., 2006). Specifically, the fertilizer under study in this work is composed by ammonium nitrate, potassium nitrate and calcium fluoride. Ammonium nitrate, when inhaled, was seen to cause possibly meaningful pulmonary function changes (Kleinman et al., 1980) and to be irritating, cause coughing, bronchospasm, laryngospasm and laryngeal edema even at low concentrations (Gorguner and Akgun, 2003). Additionally, the clinical examination of workers of the ammonium nitrate production showed frequent cases of chronic bronchitis and radiculoneuropathy (Tsimakuridze et al., 2005). On the other hand, ammonium nitrate is known to be potentially explosive when confined. Potassium nitrate, has been seen to be irritating for the respiratory tract (INCHEM, 2001). Therefore, the study of packing of an inorganic fertilizer is of interest as workers can be exposed to relatively high concentrations of airborne fertilizer particles, which might cause respiratory health effects.

Exposure prediction models have been proposed as valuable risk assessment tools. Since the initial application of exposure prediction models, several research papers have been published regarding their theoretical aspects (Ganser and Hewett, 2017; Hewett and Ganser, 2017; Hussein and Kulmala, 2008; Nazaroff, 2004; Nazaroff and Cass, 1989). The two box model is a well-accepted exposure assessment tool in the risk assessment field as, even with its simplified assumptions, it is able to adequately simulate actual conditions for various processes including volatile compounds and PM emissions (Arnold et al., 2017; Jayjock et al., 2011). In the chemical industry, models have been tested in a variety of cases (Nicas, 2016; Sahmel et al., 2009 and references therein). However, when testing the models for PM in actual industrial environments, the number of studies decreases (Boelter et al., 2009; Johnson et al., 2011; Jones et al., 2011; Koivisto et al., 2015; Lopez et al., 2015). Recently, Arnold et al. (2017) conducted a study where the one and two box models, were evaluated under highly controlled conditions. Predicted exposure results for three industrial solvents when using near and far field models was categorized excellent and good to excellent under the ASTM Standard 5157 criteria (Arnold et al., 2017). However, in order to implement prediction models as trustable tools for worker risk assessment, additional real-world cases (including low and high exposure concentration scenarios) need to be evaluated, in order to validate model performance under real-world settings. Especially, performance of models on low concentration scenarios is relevant since real industrial exposure concentrations (especially for nanomaterials) are frequently low (Fonseca et al., 2018; Koivisto et al., 2012a; Koponen et al., 2015; Kuhlbusch et al., 2004). Thus, if models are to be used as tools for risk assessment, testing their performance in low emission and concentration scenarios is paramount. This, will favour understanding of the uncertainties related to critical parameters, such as the source characterization, local controls, and air mixing (Jayjock et al., 2011; Sahmel et al., 2009).

The objectives of the present study were 1) to perform a worker exposure and risk assessment study of packing of an inorganic complex fertilizer in an industrial plant, and 2) to test the one box and two box models performance in a real-world setting in order to contribute to the better understanding and validation of exposure prediction models. A real industrial case scenario, characterized by low particle emissions and subsequently low exposure concentrations, was selected for this purpose with the aim to test the applicability of models at the lower end of the particle concentration range. In this way, results are expected to be extrapolable to industrial settings dealing with nanomaterial exposures, which are typically low (e.g., Koivisto et al., 2012a; Kuhlbusch et al., 2004).

2. Methodology

2.1. Fertilizer chemical composition

The main chemical components of the fertilizer under study (commercial complex inorganic fertilizer) were ammonium nitrate; NH_4NO_3 (15–20%), potassium nitrate; KNO_3 (12.5–15%) and calcium fluoride; CaF_2 (2–3%), according to the material's safety data sheet. The fertilizer was granulated in 2.5 to 5 mm diameter spherical pellets. The product is not classified as hazardous according to regulation EC 1272/2008. However, it may intensify fire (oxidizer) it causes serious eye irritation, the inhalation of its degradation products may cause health hazards, and serious effects may be derived following exposure (material's safety data sheet). According to the European Chemicals Association (ECHA), Derived No Effect Level (DNEL) for long term inhalation are 37.6 mg m^{-3} for ammonium nitrite, 36.7 mg m^{-3} for potassium nitrite and 5 mg m^{-3} for calcium fluoride. For calcium fluoride, the EU occupational exposures limit (OEL) time-weighted average (TWA) is 2.5 mg m^{-3}. Recommended controls are good general ventilation, the use of safety glasses with side-shields, chemical resistant gloves and respiratory protection in case of inadequate ventilation (material's safety data sheet).

2.2. Work environment and packing process

The measurements were carried out during packing of a fertilizer in two different packing lines between the 23th and 26th of January 2017 at an industrial facility located in Castellón, Spain.

The packing hall was only naturally ventilated and the replacement air came from outdoors and from adjacent industrial hall via doors, which were always open (Fig. 1). The packing lines were for small bags (25 kg) and big bags (600 kg) where the studied fertilizer was