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CFD Modelling of the 3D spatial and temporal distribution of 1-methylcyclopropene in apple fruit storage

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CFD Modelling of the 3D spatial and temporal distribution of 1-methylcyclopropene in apple fruit storage

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ABSTRACT

In this paper, a direct model based on explicit geometry of stacked products in boxes was developed and used to study the diffusion, convection and adsorption of 1-MCP gas in cool stores for apple fruit. The discrete element method was employed to generate random stacking of spherical products in a box. A three-dimensional finite volume based CFD model was developed, verified and used to study the distribution and partitioning of the 1-MCP gas inside loaded container. The study addressed the gas distribution in a 500L container with or without air circulation. For each case, 80 kg Jonagold apples at 1°C and a 1-MCP dose of $1\mu\text{L L}^{-1}$ was used to collect validation data. In the presence of air circulation, diffusion-convection in air and diffusion adsorption in the product was applied. Simulations were performed with an unstructured tetrahedral mesh using the software ANSYS-CFX, a Reynolds Averaged Navier-Stokes solver. The case without air circulation was modelled as a diffusion problem in air and diffusion coupled with adsorption inside the product. Convection-diffusion-adsorption model parameters that were previously developed and validated were applied. The estimated equilibrium distribution of the 1-MCP gas equals 11%, 34% and 55% as unbounded in fruit, bonded in fruit and remaining in container respectively. Profiles of free (unbounded) and adsorbed (bounded) 1-MCP concentrations inside fruit were estimated for reduced dosages: 0.5, 0.3, 0.1, and $0.02\mu\text{L L}^{-1}$.

Keywords:

1-MCP, Fruit storage; Computational fluid dynamics, Gas diffusion, Gas adsorption, ethylene, postharvest

1. INTRODUCTION

Ripening of fruits and vegetables during storage or transportation must be controlled for effectively maintaining quality in the postharvest supply chain. Suppressing ripening of fruits and vegetables during storage demands controlling the action of ethylene, a molecule that promotes ripening and senescence. (Blankenship & Sisler, 1989). Refrigeration and controlled atmosphere (CA) storage are used by growers to slow ripening. Both methods work by, amongst others, reducing the ethylene production and activity. 1-methylcyclopropene (1-MCP) that interferes with the ability of plants to respond to ethylene provides a tool for postharvest management of climacteric fruits (Sisler & Blankenship, 1996; Sisler & Serek, 1997; Blankenship & Dole, 2003; Sisler & Serek, 2003; Watkins, 2006; Huber, 2008). 1-MCP works by attaching to a binding site (receptor) in fruit tissues that normally binds ethylene (Serek et al., 1994; Hall et al., 2000; Binder & Bleecker, 2003). The binding of 1-MCP causes fruits to ripen and soften more slowly; therefore maintaining their quality for longer periods of time (Sisler & Blankenship, 1996; Sisler and Serk, 1997; Golding et al., 1998; Abdi et al., 1998; Ku and Wills, 1999; Sisler et al., 1999).

The larger part of studies on 1-MCP describes plant responses to single or multiple doses of the gas at various temperatures and duration of treatment. A comprehensive review of these studies can be found in Blankenship & Dole (2003). For apple fruit, responsiveness to 1-MCP differs between cultivars and the exact reason of this variation is not fully known (Watkins et al., 2000). Responses to treatment with 1-MCP are influenced by a number of factors including method of application, commodity type, concentration during treatment, duration of exposure, fruit maturity, temperature at the time of

1 68 treatment, and the interval between harvest and treatment (Blankenship & Dole 2003,
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3 69 Jayanty et al. 2004). In some situations higher doses can cause excessive delay of
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5 70 ripening (Calvo & Sozzi, 2004; Ekman et al., 2004). For pear fruit, for instance, a dose
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7 71 higher than 0.5 $\mu\text{L L}^{-1}$ can cause excessive delay of ripening and alter ripening related
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9 72 developmental processes sufficient to significantly reduce product quality (Ekman et al.,
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16 74 The effective dose of 1-MCP for maximum efficacy of treatments is not yet clearly
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18 75 known. In fact, doses differ from country to country. In the United States and Canada, the
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20 76 labelled treatment dosage for apple is 1.0 and 0.6 $\mu\text{L L}^{-1}$, respectively (Pest Management
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22 77 Regulatory Agency, Health Canada, 2004). In Europe, the minimum use rate is 0.545 μL
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25 78 L^{-1} , and 1 $\mu\text{L L}^{-1}$ is prescribed as a critical use rate (European Food Safety Authority, 2005).

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29 79 Loss of 1-MCP gas due to sorption by non-target materials found in treatment rooms or to
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31 80 non-specific sorption sites inside fruit tissue can be significant. Vallejo & Beaudry (2006)
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33 81 suggested that 1-MCP levels can be compromised by wooden and cardboard bin and bin
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35 82 liner materials, but not by plastic bin materials or wall surface materials. Ambaw et al.
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38 83 (2011) modeled the diffusion-adsorption kinetics of 1-MCP in apple fruits and in several
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40 84 solid materials and estimated the amount of 1-MCP gas bounded (adsorbed), unbounded
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43 85 (free) in apple tissue (or other solid materials) and remaining free in air during treatments.

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46 86 The quantitative results suggested higher adsorption capacity of wooden and cardboard
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48 87 bin materials in line with the observations made by Vallejo & Beaudry (2006). In
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50 88 addition, the amount of adsorbed 1-MCP in fruit was several thousand folds higher than
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53 89 the amount of ethylene binding sites estimated by Blankenship & Sisler (1989), indicating
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56 90 the existence of multiple of non-specific 1-MCP sorption sites in fruit tissues. Choi &

1 91 Huber (2009) did a rigorous study on the nature and multiplicity of non-specific 1-MCP
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3 92 sorption sites in fruit and vegetable tissues and concluded that 1-MCP sorbs to several
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5 93 cellular targets, and hydrophobic components are preferred sorption sinks.
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8 94 In commercial applications the 1-MCP gas is released from a formulated cyclodextrin
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10 95 powder through aqueous dissolution. The gas, after escaping from the aqueous-solution
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12 96 complex, diffuses in air and is distributed by diffusion and convection to reach the
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14 97 produce or other solid materials in the storage room. The non-equilibrium distribution of
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16 98 the gas between the solid phase and the fluid phase then initiates the diffusion of the gas
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18 99 into the solid materials. The distribution of the gas is governed by the transport properties
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20 100 of the gas in air and in solid plus the nature of the adsorption. The airflow inside the
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22 101 storage room also has a profound effect over the overall distribution of the gas in
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24 102 treatment units. Hence, the design of the room, the stacking of fruit bins, the bin design
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26 103 and materials of construction are likely to affect the distribution of the gas in the storage
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28 104 room. Generally the transport phenomena (airflow, heat and mass transfer) in such
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30 105 systems are complex coupled processes and mathematical models are recommended to
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32 106 support understanding and designing the systems (Hoang et al., 2003; 2004,
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34 107 Nahor et al., 2005; Smale et al., 2006; Verboven et al., 2006; Alvarez & Flick, 2007;
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36 108 Ferrua & Singh, 2009).
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46 109 Verboven et al. (2006) gave a comprehensive review of different mathematical
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48 110 approaches to model the transport phenomena in food bulks, packages and stacks and
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50 111 suggested the use of computational fluid dynamics (CFD) as a valuable tool in the design
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52 112 and optimization of postharvest storage facilities. Review of mathematical methods in
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55 113 agro-food applications (Xia et al., 2002; Wang & Sun, 2003; Norton & Sun, 2006; Delele
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et al., 2010; Smale et al., 2006) indicated the added advantages and the increased use of CFD modelling techniques in the area. The reviews also indicated the presence of a substantial degree of approximations. In most cases, porous media approaches are still being used. Computational requirements for direct CFD simulations are still large in order to accurately resolve every detail in postharvest systems. Hence, assumptions and approximations are still the central part of CFD modeling. There is strong interest towards using CFD on detailed geometrical models of products and packages to explore further the transfer processes in postharvest storage (Delele et al., 2008; Ferrua & Singh, 2009).

In this paper the modeling of the three dimensional (3D) spatial and temporal distribution of 1-MCP gas in apple storage containers subject to diffusion, convection and adsorption is addressed. The objective was to understand the mechanisms of the 3D distribution of 1-MCP gas in storage spaces for apple fruit and develop a physically realistic model, based on computational fluid dynamics (CFD). Validation of the 3D model was performed by dedicated gas concentration measurements. The model was used to investigate uniformity and the effect of application dose.

2. MATERIALS AND METHODS

2.1. Apple fruits

‘Jonagold’ apples (*Malus domestica* Borkh., cv. Jonagold) were purchased directly after harvest from a local grower in Belgium in September, 2010. All the fruits used for the test were free of visual defects. Fruits were stored at 1°C in normal atmospheric air before and during the experiment that took place within a few days. The mean $\pm$ standard deviation

(n = 10) of the mass and volume of fruits were $240\text{g} \pm 22\text{ g}$ and $300\text{ mL} \pm 30\text{ mL}$, respectively.

2.2. The container

The container used for the study was a stainless steel rectangular box with gas tight glass door placed inside an automatically controlled cool store at the Flanders Centre of Postharvest Technology (Leuven, Belgium). The dimension of the container was $0.74\text{ m} \times 1.00\text{ m} \times 0.67\text{ m}$. One ETRI[®] AC axial flow fan with a diameter of 10.5 cm and a capacity of 44 L/s was placed at the container symmetry plane as shown in Fig. 1. Four slotted High-density polyethylene (HDPE) plastic bins (Euro Pool System International B.V., Rijswijk, The Netherlands) with dimension of $0.67\text{ m} \times 0.37\text{ m} \times 0.23\text{ m}$ were used to hold the fruit. The ventilating slots were spread over the five faces of each box with a vent-hole ratio of 20% at side faces and 3% at the bottom face of the box (Fig. 2). Each bin contains $20 \pm 0.2\text{ kg}$ of fruits and the boxes arrangement was in two rows and two columns (Fig.1). 1-MCP gas at target concentration of $1\mu\text{L L}^{-1}$ was generated by placing 0.86 g SmartFresh powder (SmartFresh[™], AgroFresh, Springhouse, PA), which had an active ingredient concentration of 0.14%, in a 300 mL glass beaker to which 4 mL distilled water was added. The placement and arrangement of bins, the circulating fan, gas generating beaker and gas sampling locations inside the container were in consideration of symmetry for the modelling and simulation process. The container was positioned in a cool room controlled at a temperature of 1°C. A leakage test was conducted by generating 1-MCP gas inside the empty container. The container was fitted with eight gas-sampling plastic tubing at different positions for measuring 1-MCP at regular intervals during a period of 24 hours.

2.3. Gas sampling and analysis

The concentration of 1-MCP was measured by capillary type gas chromatography (Compact GC, Interscience, Louvain-la-Neuve, Belgium) fitted with a 15 m long, 0.53 mm stainless steel column coated with a 0.5 μ m thick layer of MXT-WAX and equipped with a flame ionization detector (FID). A 10 mL of gas sample was injected into the gas chromatograph. Air and hydrogen gas flow rates in the FID were 300 mL min⁻¹ and 30 mL min⁻¹ respectively. Helium was used as a carrier gas. The column oven was kept at 42°C.

There were eight sampling points: at the void space between the fruits in the bottom bin, at the void space between fruits in the top bin, at the right and left free spaces of the container, at the top and bottom free spaces of the container and at the free space between two side-by-side bins (on symmetry plane) (Fig. 3). At sampling points, ends of tubes of equal length (1.5 m) were fixed. The other end of each sampling tube extended outside of the container and attached to a valve. Eight glass bulbs of size 25 ml fitted with a 20mm combination seal (Alu. Cap, Grace Alltech, Deerfield, Illinois) were vacuumed to nearly 200 mbar absolute pressure and used to draw gas samples from each sampling point. The dilution effect due to using this sampling technique was calibrated by drawing known concentrations of gas samples from separately prepared standards.

2.4. Experiments

The 1-MCP gas distribution in the loaded container was assessed with and without air circulation for a target concentration of 1 μ L L⁻¹. The container loaded with 80 kg of fruit in 4 bins was kept in a cool room at a constant temperature of 1°C. The initial temperature

of the fruit was 1°C before loading and experiments were started after equilibration inside the container overnight to ensure isothermal conditions. Then the beaker with the SmartFresh solution was added to the container. The times of 1-MCP concentration measurements at each of the 8 sampling points were 2, 4, 6, 8, 10 and 24 hours after closing the container.

Velocity measurements were performed in a separate test in a loaded container by means of air velocity transducers (TSI 8465 and TSI 8475, TSI Incorporated, Shoreview, MN, USA) which measure velocities using thermal anemometry. The windowless (Model 8465) probe was used to measure higher velocities near the fan exit and around the stack of fruits. The Omnidirectional (Model 8475) was used to take velocity measurements inside the stack (in the void space between fruits). Velocity measurements were taken from five points along horizontal lines: one point at the left side of the stack, three points between the fruit inside the stack and one additional point at the right side of stack. Such measurements were taken on five horizontal planes at the middle of bottom bin, above the fruits of the bottom bin, at the middle of the top bin above fruits of the top bin, and in the top of the container (Fig. 3).

2.5. Mathematical model

In the presence of air circulation, diffusion-convection in air and diffusion-adsorption in the product were considered. The case without air circulation was modelled as a diffusion problem in air and diffusion coupled with adsorption inside the product. Because HDPE does not significantly adsorb 1-MCP (Ambaw et al., 2011), only adsorption by the fruit was taken into account.

2.5.1. Model concept and assumptions

1 203 The mathematical model describing the diffusion of 1-MCP gas in air and in various solid
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3 204 materials was obtained from our previous work which modeled the kinetics of adsorption
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5 205 of 1-MCP in apple fruit and several non-target solid materials found in apple storage
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7 206 rooms (Ambaw et al., 2011). For a loaded container with air circulation, convection was
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9 207 added to the diffusion-adsorption process by explicitly modelling the airflow.

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13 208 The level of concentrations of 1-MCP gas used in the treatment is very dilute; its effect on
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15 209 the airflow was, therefore, neglected. Therefore, scalar transport equations were used to
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17 210 describe the transport and adsorption of 1-MCP. Two transport equations were defined,
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19 211 one to describe the unbounded 1-MCP gas in air and in solid, another to account for the 1-
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21 212 MCP bounded in solid.

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26 213 The length scale in which the 1-MCP gas transfer takes place ranges from around 0.001 m
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28 214 at the thickness of the HDPE plastic bin and between spheres, to about 1 m at the
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30 215 container level. Due to this range of scales, the number of grid points during meshing
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32 216 increases dramatically near the bins and between fruits, which directly affects the
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34 217 computational time and may exceed the present computing capability. To overcome this
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36 218 difficulty, volume meshing of the HDPE plastic bins was dropped and the accompanying
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38 219 adsorption of 1-MCP by it was neglected (justified by the low adsorption by this material,
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40 220 Ambaw et al. (2011)).

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47 221 Isothermal conditions were assumed in the model. This is reasonable for the fact that
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49 222 fruits were stored at 1°C in normal atmospheric air before and during the experiment. In
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51 223 practice, however, 1-MCP may be applied while performing cooling of the fruits. Produce
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53 224 generally takes several hours to reach to the equilibrium temperature depending on the
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55 225 cooling method. During this cooling period, variation of temperature in space and time is
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evident. The effect of this non-isothermal condition on the distribution of the 1-MCP gas is not yet understood and needs a dedicated investigation.

The concentration level of 1-MCP prescribed is very low (in the range of part per millions). Hence, it is reasonably safe to assume concentration-independent diffusion and adsorption coefficients.

2.5.2. Governing equations

The presences of complex wall geometries together with the high air velocities from the axial flow created by the axial flow fan suggest turbulent conditions around the bins. Under these flow conditions, the conservation of mass, momentum and scalar quantities within the system are given by Eqs. (1), (2) and (3) respectively.

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{U}) = 0 \quad (1)$$

$$\frac{\partial (\rho \mathbf{U})}{\partial t} + \nabla \cdot \{\rho \mathbf{U} \otimes \mathbf{U}\} = \nabla \cdot \{\sigma - \rho \overline{u \otimes u}\} + S_M \quad (2)$$

$$\frac{\partial (\rho \Phi_1)}{\partial t} + \nabla \cdot (\rho \mathbf{U} \Phi_1) = \nabla \cdot (D_a \nabla \Phi_1 - \rho \overline{u \Phi_1}) + S_{\Phi_1} \quad (3)$$

where ρ is the density of the fluid, $\mathbf{U}$ is the velocity vector, $\otimes$ is the dyadic operator (or tensor product), ∇ is del operator (vector differential operator), σ is stress tensor including pressure, S_M is the momentum source, S_{Φ_1} is scalar (1-MCP) source, D_a is the molecular diffusivity of 1-MCP in air calculated from kinetic theory of gases (Ambaw et al., 2011) and Φ_1 is the concentration of 1-MCP in air. The momentum and scalar transport equations contain turbulence flux terms additional to the molecular diffusive fluxes. These are the Reynolds stress, $\overline{u \otimes u}$, and the Reynolds flux, $\rho \overline{u \Phi_1}$. These terms describe

the fact that convective transport due to turbulent velocity fluctuations will act to enhance mixing over and above that caused by thermal fluctuations at the molecular level (Eq. 4 & Eq. 5).

$$-\rho \overline{u \otimes u} = -\frac{2}{3} \rho k \delta - \frac{2}{3} \mu_t \nabla \cdot \mathbf{U} \delta + \mu_t (\nabla \mathbf{U} + (\nabla \mathbf{U})^T) \quad (4)$$

$$-\rho \overline{u \Phi} = \Gamma_t \nabla \Phi_1 \quad (5)$$

Here, μ_t is the eddy (turbulent) viscosity, $\Gamma_t = \frac{\mu_t}{Pr_t}$ is the eddy diffusivity. Pr_t is the turbulent Prandtl number. k is the turbulence kinetic energy and is defined as the variance of the fluctuation in velocity. Turbulence models close the Reynolds-averaged equations by providing models for the computation of the Reynolds stresses and Reynolds fluxes. By performing rigorous assessment on heat transfer simulations carried out for different test cases with experimental heat transfer data, on a series of three grids, Menter et al. (2003) showed that the SST (Shear Stress Transport) model to be superior, as it gives more accurate predictions and is less sensitive to grid variation. Delele et al. (2009) also performed a comparative study on the use of the k- ϵ , k- ω and SST turbulence models in their CFD modelling of airflow through random stacking of horticultural products in vented boxes. After comparing the convergence and the accuracy of the solution from different turbulence models (standard k- ϵ , standard k- ω and SST), they used the SST model. For these reasons, in this study the SST turbulence model is employed.

1-MCP gas inside solids exists in two different forms: free in the internal void spaces and adsorbed on specific sorption sites. The free 1-MCP inside the product is subject to a diffusion-adsorption mechanism as given by Eq. 3. The adsorption is given as a sink term in the internal void space (Eq. 5). The adsorption of 1-MCP in the product is assumed

irreversible and its accompanying transport equation is given by a pure source term in the solid domain (Eq. 6 and Eq. 7).

$$\frac{\partial \Phi_1}{\partial t} = \nabla \cdot (D_s \nabla \Phi_1) - k_s (\Phi_{s,max} - \Phi_2) \Phi_1 \quad (6)$$

$$\frac{\partial \Phi_2}{\partial t} = k_s (\Phi_{s,max} - \Phi_2) \Phi_1 \quad (7)$$

where D_s is the effective diffusivity of the 1-MCP gas in the product ($\text{m}^2 \text{s}^{-1}$); k_s is the adsorption rate constant per binding site ($\text{m}^3 \text{mol}^{-1} \text{s}^{-1}$), Φ_1 is the concentration of unbounded 1-MCP per unit volume (mol m^{-3}), Φ_2 is the concentration of irreversibly adsorbed 1-MCP gas per unit volume of solid (mol m^{-3}), $\Phi_{s,max}$ is the total amount of available 1-MCP binding sites per unit volume of solid (mol m^{-3}).

2.5.3. Geometry, boundary and initial conditions

A random stacking of 90 normally distributed spheres of size 80 - 86 mm in diameter in the box was generated by using discrete element (DE) method. the same configuration of random stacking was used for every box. The DE method is a numerical technique for solving Newton's equation of motion of an assembly of interacting particles. The gravitational force and forces due to collision between spheres or walls are the force accounted in the DE method. A detailed description of the DE method applied here can be obtained from Tijskens et al. (2003). The size and position of the randomly generated spheres were imported to Ansys Design Modeler 12.1 and assembled together with the geometry of half of the container, the two HDPE plastic bins, the fan and the 1-MCP gas generator (Fig. 2). The geometric model was loaded into ANSYS ICEM CFD 12.1 and discretised using a tetrahedral mesh. Mesh generation was based on a guideline obtained from a grid assessment study made on a simplified model.

The axial flow fan was modelled as a momentum source at the position of the fan. The magnitude of the momentum source was gauged by comparing the calculated velocity profile of the steady state solution with velocity measurements taken at selected spots inside the loaded container.

Internal surfaces of the container and the walls of the EPS boxes were defined as a no slip boundary. Fruit surfaces were set to a no slip conservative interface flux type boundary. The generation of the 1-MCP gas was modelled explicitly by setting a scalar source at a wall boundary at the gas generator. The 1-MCP release profile from the beaker was approximated by a step function (Eq. 12).

$$g(t) = \begin{cases} \dot{m}, & t \leq te \\ 0, & t > te \end{cases} \quad (8)$$

where $\dot{m}$ is a constant representing the release of 1-MCP with time per unit cross section, $\text{kg m}^{-2} \text{s}^{-1}$; t is time, s; te is the time needed for total release of the 1-MCP gas, s.

The time for complete release of the 1-MCP gas was about 3h and 5h for the cases with and without air circulation, respectively. These values were obtained from separate tests on the generation profile with time inside an empty container for the two cases. Initially the concentration of 1-MCP inside container was zero. The parameters used in the CFD model are listed in Table 1.

2.5.4. Grid sensitivity study

A grid sensitivity study was accomplished by performing multiple simulation runs on a simplified geometry that allowed careful control of the grid size on the surface of the products. In the simplified model, the complex geometry of the HDPE plastic bins were excluded and only two spheres placed side by side were considered. On the simplified

model, a steady convective-diffusive heat transfer problem (forced air-cooling) was defined, which is analogous to the present scalar transfer problem. In place of the axial flow fan a velocity inlet and pressure outlet boundaries were used. At the inlet, the temperature and velocity of the fluid were specified as 274 K and 4.5 m s⁻¹. At the outlet, the relative pressure was set to 0 Pa. The surface temperature of the spheres was 298 K. The grid sensitivity was studied by assessing the resulting wall heat transfer coefficient to the spheres as a function of grid size. The wall heat transfer coefficient was manually calculated during post-processing using Eq. (9)

$$h_w = \frac{q_w}{(T_w - T_{ref})} \quad (9)$$

h_w is the wall heat transfer coefficient [W m⁻² K⁻¹]; q_w is the calculated area average heat flux at sphere surface [W m⁻²]; T_w is the defined surface temperature [K] and T_{ref} is the inlet temperature of the fluid. Mesh refinement was continued until the change in h_w between consecutive grid sizes was negligibly small, Table 2. The finest grid approaches a grid-independent solution of the surface heat transfer coefficient, $h_w = 8.5$ W m⁻² K⁻¹. Martins et al. (2010) studied forced convection cooling of two apple fruits placed in tandem arrangement and reported a correlation equation to calculate the Nusselt number as a function of Reynolds number. The Nusselt number from the finest grid (15) is in a good agreement to the value range (14) obtained by using the correlation from Martins et al. (2010).

The grid sensitivity analysis obtained from the simplified model was then used to specify the settings of the mesh generation of the actual model of the container. The resulting mesh statistics of the final space discretisation looks as shown in Table 3. The total

number of mesh elements in fruit domain in the final model comes to 672,342. For 180 fruits in the domain, the average number of mesh element per fruit becomes 3800, which is in the level between M3 and M4 in Table 2. Mesh quality of the final model was further assessed by using indicators like orthogonality angle, aspect ratio and edge length ratio were calculated. The mesh quality indicators are in the acceptable range (ANSYS CFX-Solver Theory Guide, 2009).

2.5.5. Solution strategy

The problem was solved with the code ANSYS-CFX (ANSYS, 2009). A steady state calculation was performed and a converged solution of the airflow inside the container was obtained without any release of the 1-MCP gas. The steady state solution was then used as initial value to the subsequent transient convection-diffusion-adsorption problem. The simulation uses second order backward Euler, high resolution spatial differencing (i.e., a blend between central differencing and upwind differencing locally) and first order schemes for the transient, advection and turbulent terms, respectively. Several time step sizes (3600s, 360s, 72s and 36s) were assessed. Based on the computational time required and improved accuracy obtained by decreasing the time step, 72 s with 15 iterations was selected as the optimum. Under the selected optimum solver format, a single full simulation took 120 h on a 64-bit, Intel (R) Core (TM)2 Quad CPU, 3 GHz, 8 Gb RAM, Windows 7 PC.

In the case without air circulation, the diffusion-adsorption problem was solved with a time step and iteration number of 360 s and 15, respectively. This simulation took 12 h on a 64-bit, Intel (R) Core (TM)2 Quad CPU, 3 GHz, 8 Gb RAM, Windows 7 PC.

3. RESULTS AND DISCUSSION

3.1. Airflow pattern and turbulence

The air velocity at the fan was 4.5 m s^{-1} . The air flows directly to the opposite wall (right side of container). After hitting the wall, the air flows down around the stack (Fig. 3). The air velocity at the left side of the container was still as high as 2 m s^{-1} , and was higher near the container wall and lower near the the stack (Fig. 4). The airflow passes mainly around the stack to reach the left side of the container and return to the fan. This side of the container is characterized by a more uniform velocity distribution than the right side of container (Fig. 4) with velocity values ranging from 0.2 to 0.4 m s^{-1} . The model predictions of air velocity at various locations inside container agreed with the pattern obtained from the experimental measurements. The model clearly captured the high and low flow regions (Fig. 6).

The region immediately above the fruits in the top bin is is a low velocity recirculation zone from the main airjet in the middle of the container. Compared to the same region in the bottom bin, it actually has a lower velocity. Air velocity in this region ranges from 0 to 0.3 m s^{-1} , with the higher end of the velocity occuring in the region near to the symmetry plane (Fig 4 (b)). The stack interiors have very low velocities that fall below the detection limit of the velocity transducer (0.05 m s^{-1}) confirmed by the simulations (Fig. 6 (c) and (e)). The model shows slightly higher velocity magnitudes in the bottom region of the bin than the interior (Fig. 5 (a) and (b) for the top and bottom bins respectively). This is due to the venting holes at the bottom face of the bin. This side of the bin is with the lowest vent holes ratio (3%), and accordingly the venting from this side of the bin is poorer than the venting from the slots in the vertical faces (which has a 20% vent holes ratio).

1 381 The bottom bin is along the returning air stream. Uniformly distributed and higher flow
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3 382 velocity is observed in the region immediately above fruits in the bottom bin than the
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5 383 corresponding region in the top bin. Both the model and the experimental measurements
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7 384 confirm this fact. Airflow in this region is mainly at 0.4 m s⁻¹. Left, right, back lateral
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10 385 region, front lateral region, and bottom region of the bin are at the same magnitudes as the
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12 386 corresponding regions in the top bin.

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16 387 The low air velocity inside the stacks indicates that the air circulation created by the axial
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18 388 flow fan may not sufficiently ventilate the interior of the bins. Hence, the flow inside the
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20 389 stack might be governed predominantly by molecular transport processes. This is further
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23 390 discussed in the next section.

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27 391 For further investigation of the flow conditions, turbulence intensities on a vertical cross
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29 392 section are shown in Fig. 7. The equation used to calculate the turbulence intensity is
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32 393 given by:

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$$I \equiv \frac{u'}{U'} \quad (10)$$

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39 395 where u' is the root-mean-square of the turbulent velocity fluctuations and U' is the mean
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42 396 velocity (Reynolds averaged). From the turbulent energy k , u' can be computed as:

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$$u' = \sqrt{\frac{1}{3}(u_x'^2 + u_y'^2 + u_z'^2)} = \sqrt{\frac{2}{3}k} \quad (11)$$

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50 398 The turbulence intensity is much higher in the region outside the stack (Fig. 7).
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52 399 Especially, very high turbulence exists in the gap between the container right wall and the
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55 400 stack. The next highest turbulence intensity is in the region immediately above fruits in
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58 401 the top bin. Here turbulence intensity reached an average value of 24%. The airflow at the
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left side of the stack below the axial flow fan has a turbulence intensity of less than 10%.
The turbulence intensity inside the stack is very low. The low velocity observed in this region together with the low turbulence intensity suggests a pure diffusion dominated transport condition in this region.

The top fruits of the top bin generally experiences higher turbulence intensities than the same fruits in the bottom bin.

3.2. 1-MCP distribution in the container

The experimental measurements of the concentration of 1-MCP inside the container for the cases with and without air circulation are compared to the simulations in Fig. 8 (a) and (b) respectively. The concentration rises in the first 4 to 8 h and subsequently decreases gradually to a final equilibrium value. In the first phase, the 1-MCP gas generation rate was relatively higher than the rate of diffusion into the fruit. When all 1-MCP is released from the source, only diffusion and adsorption take place, resulting in a decrease of concentration in the air.

The model captured the overall trend of the concentration profile (Fig. 8). With air circulation, the 1-MCP concentration increased faster in the container, reached a peak sooner and levelled to a lower value at 24 hours than for the case without air circulation. Once the release is finished, the concentration in the air slowly decreases to a constant level, indicating saturation of the fruit with an overdose remaining in the container.

The spatial variation inside the container clearly reduces by adding air circulation; the airflow enhances mixing such that a nearly uniform 1-MCP concentration throughout the container is established for the whole duration of the application. Without air circulation,

the distribution shows more variability, with higher concentrations closer to the 1-MCP source during the period of 1-MCP release. At the end of the experimental period, also for the case without air circulation the distribution is quite uniform.

The model shows non-uniformity in spatial concentration values at the early hours of treatment for the case without air circulation (Fig. 9 2nd column). There are regions inside the container with zero concentration of 1-MCP after even three hours of treatment (Fig. 9 (a) 2nd column). Such variation was not observed in the measurements, but the experimental variation is smaller for the circulation case than for the no-circulation case. The gas sampling method employed may not capture the spatial variation properly. The vacuum sampling employed might disturb the pure molecular diffusion condition and create convection currents which may act to mix the fluid and reduce the spatial variation. There are to the knowledge of the authors no alternative detection methods available today for such practical evaluations.

A time scale analysis further demonstrates the importance of the different processes taking place in the container. The smallest time constant for 1-MCP supply is 3 hours (10,800s) in case of air circulation. The time constant of free diffusion inside the container is in the order of 13,000s, while for free convection this is less than 1s. The diffusion time constant of 1-MCP inside the stack is of the order $L^2/\epsilon D$, with L (0.11 m) half of smallest dimension of the bins, ϵ the porosity of the stack (0.44) and D the diffusivity of 1-MCP in air ($8.67 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$). This leads to a diffusion time constant of 3,200s. The convection time constant L/U , with U the superficial velocity inside the stack ($< 0.05 \text{ m s}^{-1}$), is 2s. The time constant of 1-MCP diffusion into the fruit equals 80,000 s. Finally, the adsorption rate time constant is $1/c_{\max}k_s$, and equals 3,000s. Clearly, the gas

supply rate, diffusion across the container and diffusion into the fruit are the limiting factors driving the process inside the container. Diffusion and convection into the stack are sufficiently fast to provide nearly uniform levels of 1-MCP to every fruit in the bins. The model confirms this analysis.

Fig. 10 demonstrates well the importance of the different processes involved in the distribution of the 1-MCP gas inside the container. The main gradients persist inside the fruit, due to the large time constant of diffusion in the fruit. In the case without air circulation, the diffusion across the container is also limiting the distribution during the first hours of 1-MCP application. Sixteen hours after the start of the treatment, the 1-MCP concentration in both cases was rather uniform, confirming the experimental observation.

There are noticeable quantitative deviations between the modelled and measured average profiles of 1-MCP in the container. In particular, the calculated curves show lower concentration values in the first few hours of the test and higher values afterwards. The lower values in the first 2 to 4 hours of the experiment could be attributed to the assumption of constant 1-MCP release rate. The simplified step function used to model the generation averaged the actual case, under estimating the release for the first few hours and over estimating for the subsequent hours. This is expected from the nature of the release, which might be better modelled by a continuous function, which can capture the change in rate of release with time smoothly. Simplification was initiated for the sake of reducing computation time.

Experimentally, almost 59% and 54% of the added amount of 1-MCP diffused into the fruit after 24h of treatment for the case with and without air circulation, respectively. For both cases, the model predicts a 45% uptake; which is 10 to 15% smaller. The

discrepancy between experimental results and the model estimates is due to some simplifying assumption made in the model plus the gas sampling and measurement errors. For instance, the high-density polyethylene (HDPE) plastic bin, though small, has a capacity to adsorb 1-MCP gas (Ambaw et.al, 2011).

3.3. 1-MCP transfer to the fruits

1-MCP gas flux into fruits, calculated as area averaged values, reaches up to 2.3×10^{-12} kg m⁻² s⁻¹ and 2.2×10^{-12} kg m⁻² s⁻¹ for the cases with and without air circulation, respectively (Fig. 10 (a)). This value is 6, 3 and 1.4 fold higher in the presence of air circulation than in the absence in the first 1, 2 and 4 h of treatment respectively. The limited amount of 1-MCP gas in air is rapidly depleted within the first 4 h in the presence of air circulation. The spatial variations of flux values were assessed by taking the ratio of fluxes in fruits in the top bin to that in the bottom bin (Fig 10 (b)). This ratio is clearly different for the two cases. The ratio rapidly reaches unity for the air circulation case indicating the uniform gas supply over all fruit surfaces, and negligible spatial variation in surface mass transfer rates. The high flux value of fruits in the top bin for the first few minutes related to the observed higher turbulence intensity in the region is noticeable.

Without air circulation, there is a significant variation of flux values between the top and bottom bin fruits throughout the treatment period (Fig 10 (b)). In the beginning, 1-MCP does not reach the top bin, therefore the flux is zero. As the 1-MCP is distributed, the flux in the top bin catches up with the bottom bin flux. The flux in the top bin then relatively remains higher than in the bottom bin during the flux decrease period.

3.4. 1-MCP inside fruit

1-MCP is present in two different forms inside the fruit: bounded to ethylene and other receptors and free in the internal pore spaces of the fruit. Experimental determination of the magnitude of these two concentrations is almost impossible. The model provides a framework to calculate and visualize the amount adsorbed inside each fruit in the container so that process of uptake and adsorption for the two cases can be compared. Fig. 11 show the contours of 1-MCP inside the fruit at different times during the process. Faster adsorption can be noticed due to air circulation, the equilibrium distribution is equal for the two cases.

After 24 h of treatment, the distribution of 1-MCP reached equilibrium for the cases with and without air circulation (Fig. 12). The estimated overall equilibrium distribution of the 1-MCP gas equalled 34% bound in the fruit, 11% free in the fruit, and 55% free in container. Clearly, there has been an overdosing.

3.5. Simulation of the effects of reduced 1-MCP doses

For the dose considered in this manuscript ($1 \mu\text{L L}^{-1}$), we found that the equilibrium concentration in the fruits reached the saturation level in the fruit with remaining gas in the air. At lower doses, this may no longer be the case and maybe gradients inside the fruit may occur.

Performing a 1-MCP balance calculation for the case of $1 \mu\text{L L}^{-1}$ dose, the maximum amount of 1-MCP adsorbed is about $5 \times 10^{-6} \text{ kg m}^{-3}$ (equivalent to about 1.2×10^{-7} moles/kg). The number of ethylene binding sites has been estimated to be 3.2×10^{-11} moles/kg for “Starkrimson” Red Delicious apple (Blankenship and Sisler, 1989). Assuming the same amount of ethylene binding sites for Jonagold apple, a dose of $1 \mu\text{L L}^{-1}$ brings about

4000 folds higher concentration of 1-MCP molecules to the available ethylene binding sites.

The distribution of 1-MCP gas under reduced doses was assessed using the CFD model.

The container without air circulation was used. Doses of 0.5, 0.3, 0.1 and 0.02 $\mu\text{L L}^{-1}$ were considered in addition to the 1 $\mu\text{L L}^{-1}$ reference case. The volume averaged and minimum 1-MCP bounded in the fruit with time were computed and are shown in Fig. 13 and Fig. 14, respectively. The minimum adsorbed 1-MCP concentration in fruit is the value at the core of the fruit. The volume-averaged as well as the minimum adsorbed 1-MCP reached the saturation level ($\Phi_{s,max} = 5 \times 10^{-6} \text{ kg m}^{-3}$) within 24 h only for the 1 $\mu\text{L L}^{-1}$ dose. After 24 h, the average adsorbed 1-MCP level reached 88, 65, 25 and 5% of the maximum attainable for the 0.5, 0.3, 0.1 and 0.02 $\mu\text{L L}^{-1}$ doses, respectively. At a doubled treatment time of 48 hours, the adsorbed 1-MCP goes to $\Phi_{s,max}$ for the 0.5 $\mu\text{L L}^{-1}$ but not for the other doses.

To saturate the entire fruit within 24 h of treatment, a dose higher than 0.5 $\mu\text{L L}^{-1}$ is thus required. This result agrees with a previous dose response study that indicated that between 0.5 and 1 $\mu\text{L L}^{-1}$ 1-MCP is required for maximum response (Jayanty et al., 2004).

Jayanty et al. (2004) suggested that lower doses being nearly as effective as higher doses under repeated (weekly) application. In their study, the response to 0.1 $\mu\text{L L}^{-1}$ dose was nearly as effective as 1.0 $\mu\text{L L}^{-1}$, while a dose of 0.02 $\mu\text{L L}^{-1}$ did not bring any noticeable suppression of repining for 'Redchief Delicious' apples under multiple applications (more than four times) of the gas on a weekly basis (Jayanty et al., 2004). The complete lack of suppression of ripening by the 0.02 $\mu\text{L L}^{-1}$ dose is interesting. It suggests that the degree

of adsorption achieved under this dose is insufficient to initiate any suppression of ripening. The researchers proposed a reversible way of 1-MCP sorption and suggested a gradual release of the 1-MCP out of the fruit body as the cause of absence of efficacy. Why treatments under higher dosage (say, 1 $\mu\text{L L}^{-1}$) then become fully effective even in single application is still requiring an explanation. If the sorption of 1-MCP is assumed reversible then it is apparent that adsorbed 1-MCP under higher doses could also be released out of the fruit and suppression of ripening be compromised to a certain extent.

With the present model, there is a degree of adsorption of 1-MCP gas under the 0.02 $\mu\text{L L}^{-1}$ dose. The minimum adsorbed 1-MCP level inside fruit for the 0.02 $\mu\text{L L}^{-1}$ is 17 fold higher than the estimated concentration of ethylene binding sites (Blankenship & Sisler, 1989) but with level of adsorbed 1-MCP concentration significantly lower than the available 1-MCP binding sites. This may indicate a condition of insufficient treatment to a degree that no noticeable suppression of ripening is observed. It appears that the level of the 1-MCP concentration must be much greater than the concentration of available ethylene sites for sufficient suppression of ripening. The 1-MCP binding sites may even have higher likelihood to take the 1-MCP gas than the intended ethylene binding sites. If this is the case, it is only after saturating the other binding sites that the ethylene binding spots get chance for the 1-MCP gas. Hence, treating fruits at lower dose may mean leaving the ethylene binding sites unblocked considerably. If the binding to these other 1-MCP sites is presumed reversible then all adsorbed 1-MCP gas will slowly desorb out of the fruit making multiple application at lower doses to have no effect as that suggested by Jayanty et al. (2004). Choi and Huber (2009) performed an extensive investigation on determining the nature and multiplicity of non-specific 1-MCP sorption sites in fruit and

vegetable tissues and reported existence of multiple cellular components contributing to non-specific sorption of 1-MCP.

4. CONCLUSIONS

The 3D spatial and temporal distribution of 1-MCP in apple fruit storage for two cases was assessed using CFD models. The results from the model simulation agreed sufficiently with experimental values of concentration and velocity measurements. The models demonstrated absence of significant spatial variation of the 1-MCP gas in container and in fruit after 24 h treatment. There are, however, noticeable spatial variations of concentrations in fruit at early stage of treatment, say, up to 12 to 16 h. The variation is higher for the case of treatment without air circulation. This indicates the potential presence of spatial variation of the gas under insufficient ventilation in large spaces. Properly designed stack arrangement, sufficient ventilation, and appropriately placed air circulating unit looks essential for large treatment storage facilities.

Treatments under reduced doses of 1-MCP gas were also investigated. The model indicated the presence of untreated region of fruit tissue for the $0.02 \mu\text{L L}^{-1}$ dose. Accordingly, other researchers indicated insufficient treatment leading to total absence of response from a $0.02 \mu\text{L L}^{-1}$ dose even under repeated applications. These suggest the presence of additional 1-MCP binding sites in fruit tissue other than the intended ethylene binding sites. Effective treatment may require saturating all 1-MCP binding sites altogether. Doses higher than $0.5 \mu\text{L L}^{-1}$ are required if fruit saturation is required within 24 hours.

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7. FIGURE CAPTIONS

Fig. 1. Schematic diagram of the full setup of the experiment.

Fig. 2. Location of velocity and concentration measurements. (a): Position of horizontal planes for velocity measurements: P1, mid of bottom bin; P2, top of fruits in bottom bin; P3, mid of top bin; P4, top of fruits in top bin; P5, top of container. The placement of velocity sensors on the planes are shown as S1, S2, ...S5. (b): 1-MCP gas sampling points: 1, top of container; 2, between fruit in top bin (not visible); 3, between fruit in bottom bin (not visible); 4, bottom of container; 5, left side; 6, right side; 7, front side; 8, back side (on symmetry plane).

Fig. 3. Stream diagram from the CFD model showing the major direction of airflow inside loaded container with air circulation.

Fig. 4. Velocity contours along horizontal planes of the airflow inside container. (a) through the axial flow fan; (b) above fruits in the top bin; (c) mid of top bin; (d) above fruits in bottom bin and (e) mid of bottom bin.

Fig. 5. Velocity vector along vertical plane through the stack. (a) Top bin; (b) bottom bin. The direction of arrow heads indicates the direction of the velocity. The background contours indicate the local velocity magnitude.

Fig. 6. Measured and calculated velocity profile inside loaded container with air circulation. (a), Top of container; (b) above fruits in top bin; (c) mid of top bin; (d) above fruits in bottom bin and (e) mid of bottom bin. Curves are CFD predictions and data points are velocity measurements at S1, S2, S3, S4 and S5 as shown in Fig. 2.

Fig. 7. Turbulence intensity contours along a vertical plane through bisecting the stack.

Fig. 8. Experimental and calculated concentration of 1-MCP remaining in container with time. (a) with air circulation; (b) without air circulation. Full curves are calculations and broken curves are for measurements. The 1-MCP dose was $1\mu\text{L/L}$, the temperature 1°C . Vertical bars are standard error.

Fig. 9. Contours of unbounded (free) 1-MCP concentration in the container and the fruit: left and right columns are for the cases with and without air circulation, respectively, after 3h (a); 6h (b); 10h (c); 18h (d). The 1-MCP dose was $1\mu\text{L/L}$, the temperature 1°C .

Fig. 10. 1-MCP fluxes analysis for with and without air circulation cases. (a) area averaged flux of 1-MCP across all fruit surfaces; (b) Ratio of area averaged flux of 1-MCP across fruit surfaces in top bin to fruit surfaces in bottom bin.

Fig. 11. Contours of bounded (adsorbed) 1-MCP concentration in the fruit. Left and right columns are for the cases with and without air circulation, respectively, after 3h (a); 6h (b); 10h (c); 18h (d). The 1-MCP dose was 1 μ L/L, the temperature 1°C.

Fig. 12. Model prediction of the distribution of 1-MCP inside the container and the fruit. The 1-MCP dose was 1 μ L/L, the temperature was 1°C. Curves are for the cases with air circulation (broken curves) and without air circulation (full curves).

Fig. 13. Simulation of volume average 1-MCP bounded in fruit under different doses of 1-MCP.

Fig. 14. Simulation of minimum concentration of 1-MCP bounded in fruit under different initial doses.

Fig. 1
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Fig. 2

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Fig. 5

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(a)

(b)

Fig. 6

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(a)

(b)

(c)

(d)

(e)

Fig. 7

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Fig. 13
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Fig. 14
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Table 1
Model parameters and their values

Parameter	Value
Kinematic Diffusivity of 1-MCP gas in air D_a [$\text{m}^2 \text{s}^{-1}$]	8.67×10^{-6}
Kinematic Diffusivity of 1-MCP gas in apple fruit, D_s [$\text{m}^2 \text{s}^{-1}$]	2.41×10^{-8}
Total amount of available 1-MCP binding sites per unit volume of solid, $\Phi_{s,max}$ [kg m^{-3}]	5×10^{-6}
Adsorption rate constant per binding site, k_s [$\text{m}^3 \text{mol}^{-1} \text{s}^{-1}$]	3.62
Dry air density, ρ [kg m^{-3}]	1.29
Porosity of apple (bulk), ϵ []	0.44
Turbulence Prandtl number, Pr_t	0.9

Table 2
Mesh dimensions used for the grid independence test.

Mesh Type	Total No. of mesh elements	NEF ^a	ES ^b	AEV ^c	SEV ^d	MEV ^e	AAHTC ^f
M1	56644	971	8.00E-03	6.46E-07	1.05E-07	1.21E-06	7.94
M2	62295	1382	7.00E-03	4.78E-07	6.78E-08	9.56E-07	8.15
M3	72810	2321	6.00E-03	3.03E-07	2.82E-08	6.33E-07	8.32
M4	89454	3993	5.00E-03	1.81E-07	2.24E-08	4.26E-07	8.56
M5	1E+05	6820	4.00E-03	1.23E-07	1.01E-08	4.42E-07	8.54

^a Number of elements in single fruit

^b Element size [m]

^c Average element volume [m⁻³]

^d Smallest element volume [m⁻³]

^e Maximum element volume [m⁻³]

^f Area average wall heat transfer coefficient at fruit surface [W m⁻² K⁻¹]

79 Table 3
80 Mesh statistics of the final computational domain.

Domain	Nodes	Elements	OA ^a	MOA ^b	MVAR ^c	MVALR ^d	AEV ^e
Container	941008	4547830	68.5484	14.7209	11.2977	9.54587	2.13 E -7
Fruit body	143348	672342	63.9249	40.8091	9.17248	8.76852	2.74E -7
Fan region	20277	112570	71.4552	49.5849	3.21072	3.02269	3.21E-9
All Domains	1,104,633	5,332,742					

81
82 ^a Orthogonality angle, the acceptable range is > 20°
83 ^b Minimum orthogonality angle, the acceptable range is > 10°
84 ^c Maximum value of aspect ratio, the acceptable range is < 100
85 ^d Maximum value of edge length ratio, the acceptable range is < 100
86 ^e Average element volume [m⁻³]