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Computers and Electronics in Agriculture

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Original papers

Phosphine distribution and insect mortality in commercial metal shipping containers using wireless sensors and CFD modeling

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ARTICLE INFO

Keywords:
Fumigation
Resistance
Monitoring
Computational Fluid Dynamics
Mathematical modeling

ABSTRACT

In the present work, the distribution of phosphine gas in metal shipping containers was modeled and compared with available data from phosphine sensors. Two different sizes of containers, 20 and 40 ft, were used in the experiments with different doses for each treatment. In each container, sensors were placed to monitor the distribution of phosphine, along with vials with phosphine –susceptible and –resistant insect populations. The insects used in the experiments were the Rhyzopertha dominica (F.) and Oryzaephilus surinamensis (L.), which are two of the most common species found in stored products. A Computational Fluid Dynamic (CFD) model was developed using the OpenFoam software and combined with phosphine sensors for precision fumigation practices. Gas transport and sorption effects of phosphine into empty and filled containers were considered in the CFD model. In light of our findings, in an empty container, the phosphine concentration was approximately similar for all locations, while in the filled container there were noticeable variations inside the fumigated area. Moreover, there was a time delay for phosphine to reach the sensors that were submerged inside the fumigated commodity, at the rear side of the containers. Regarding the simulations, the predictions of the computational model were in accordance with the phosphine concentration as recorded by the sensors. Concerning insect mortality data, in most of the cases, for both species, complete control was noted, regardless of the resistance level of the population tested. These results indicated that the CFD correlated well with the phosphine concentration and insect mortality and thus, a methodology for precision fumigation in containers can be further established.

1. Introduction

Phosphine (PH_3) is a widely used fumigant for controlling insects in different types of facilities globally including warehouses, containers, silos, tarpaulins, and ships (Collins, 2010; Warrick, 2011; Agrafioti et al., 2020a). It is used for the disinfestation of a wide range of durable commodities such as grains, cereals, legumes, tobacco, dried fruits, nuts, and processing facilities (Benhalima et al., 2004; Daglish, 2004; Collins et al., 2005; Wang et al., 2006). It has several advantages that make it suitable for industrial use, such as ease of application, inexpensive, and globally accepted as a residue-free treatment (Kaur and Nayak, 2015; Nayak et al., 2020). Due to the withdrawal of methyl-bromide (UNEP, 1995), the use of phosphine has been increased gradually since 2005 (Bell, 2000; Chaudhry, 2000). Nevertheless, its extensive use, as well as the continuous and unsuccessful applications of this insecticide lead to the development of resistance (Ahmad et al., 2013; Nayak et al., 2013; Kaur et al., 2015; Saglam et al., 2015; Afful et al., 2018; Agrafioti et al., 2019; Aulicky et al., 2019). Apart from the above, insufficient monitoring of the phosphine concentration during the fumigation process and inadequate sealing of the storage facilities, have also contributed to resistance development (Agrafioti et al., 2020a; Nayak et al., 2020). This phenomenon is global as resistant populations of stored product insects have been recorded in many parts of the world (Opit et al., 2012; Nayak et al., 2013, 2020; Kaur et al., 2015; Cato et al., 2017; Sakka et al., 2018; Agrafioti et al., 2019; Athanassiou et al., 2019; Nayak et al., 2020).

One of the most important parameters for a good fumigation practice is monitoring the phosphine concentration. Common methods for phosphine monitoring include digital or analogue equipment (glass

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https://doi.org/10.1016/j.compag.2021.106087
Received 25 January 2021; Received in revised form 27 February 2021; Accepted 4 March 2021

Table 1

General information for the fumigated containers and fumigation conditions.

Year of treatment (Month)	Shipping container number	Commodity	Size (ft)	Duration of fumigation (days)	Number of locations with insect vials	Number of sensors in different locations	Dose (type of PH_3)	Temperature range (°C)	Range of concentrations at the termination of day 1 (ppm)	Range of concentrations at the termination of day 3 (ppm)	Range of concentrations at the day of the termination of the fumigation (ppm)
2017 (December)	1	empty	20	10	6	6	1 plate	20–22	300–500	300–400	0–50
2017 (December)	2	empty	20	6	6	6	1 plate	20–30	100–150	180–280	80–120
2018 (February)	3	empty	20	3	6	6	37 tablets	20–35	100–150	250–320	250–320
2018 (May)	4	currants	40	11	4	4	4 plates	20–35	200–400	840–1000	80–480
2018 (May)	5	currants	40	5.5	9	9	3 plates	20–38	200–400	800–900	600–700
2018 (June)	6	currants	40	6	6	6	2 plates	20–40	200–400	500–600	380–420

tubes) that require air to be removed from the fumigated facility (Brabec et al., 2019). These methods are time-consuming and difficult to use because they require highly trained personnel who need to take multiple measurements during the fumigation period. A common practice is to measure phosphine concentration once a day, especially when there are multiple fumigations in progress in the same area, as in the case of containers. In this regard, wireless phosphine sensors are a promising new technology which helps users to achieve complete pest control in their fumigation processes, through continuous monitoring, which can then be modeled to predict insect mortality patterns. The first wireless phosphine sensor report was published by Athanassiou et al. (2016), where the authors successfully evaluated this technology in a commercial dried fig facility, illustrating the uneven distribution of this gas within the fumigated area. Recently, Agrafioti et al. (2020a) utilized wireless phosphine sensors to evaluate the distribution and efficacy of phosphine in different commercial facilities such as horizontal warehouses, ships, silos, tarpaulin, and containers, and reported that fumigation processes in containers achieved better results, as compared to other facilities. Brabec et al. (2019) evaluated the accuracy and responsiveness of this wireless devices in grain silos, and confirmed that the sensors provided a more detailed picture of the fumigation process, as compared with conventional monitoring devices.

Different mathematical models have been evaluated and developed for phosphine fumigations by many research groups throughout the world (Darby et al., 2009; Boac et al., 2014; Isa et al., 2016; Plumier et al., 2018; Agrafioti et al., 2020b). Isa et al. (2016) worked with mathematical models that could predict fumigant distribution in leaky silos by using fan-forced fumigation. In that work, the authors studied the fan-forced fumigation via an inlet at the base of the silo, showing that leak locations can significantly impact the fumigant distribution (Isa et al., 2016). A modeling approach able to combine the distribution of phosphine and the prediction of insect mortality is Computational Fluid Dynamics (CFD), which has been successfully evaluated in the case of fumigations in silos by Agrafioti et al. (2020b). In that work, it was found that phosphine distribution on cylindrical grain silos can be notably improved through the operation of a recirculation system, an approach that was validated with field trials (Agrafioti et al., 2020b). Apart from silos, there are several reports for other types of storage structures, such as food processing facilities (Chayaprasert et al., 2006), bulked grains (Plumier et al., 2018), and bunkers (Boac et al., 2014). Nevertheless, there is still inadequate information regarding the distribution of phosphine in containers. Considering the importance of containers for the international trade of durable commodities, the lack of a comprehensive model for a typical container fumigations is a serious gap that has to be examined more thoroughly.

Based on the above, the present study focuses on the prediction of phosphine distribution in containers using a CFD approach, and its validation with data from wireless phosphine sensors. This work is a continuation of previous evaluations and field validations carried out in silos, based on the correlation of insect mortality with field fumigation trials (Brabec et al., 2019, Agrafioti et al., 2020b). In this context, the present study focuses on CFD simulations of two sizes of containers, 20 and 40 ft, which are the most common types of containers that are currently in use globally.

2. Materials and methods

2.1. *Fumigation trials*

Fumigation trials were carried out in 2017 and 2018 in commercial containers in Greece (Table 1). Plastic cylindrical vials (3 cm diameter and 8 cm in height, Rotilabo Sample tins Snap on lid, Carl Roth, Germany) were the experimental units for the tests. The upper inner part of each vial was covered with Fluon (polytetrafluoroethylene; Northern Products, Woonsocket, USA) to prevent insects from escaping. Each vial was filled with 10 g of either soft wheat or oat flakes, for *R. dominica* and

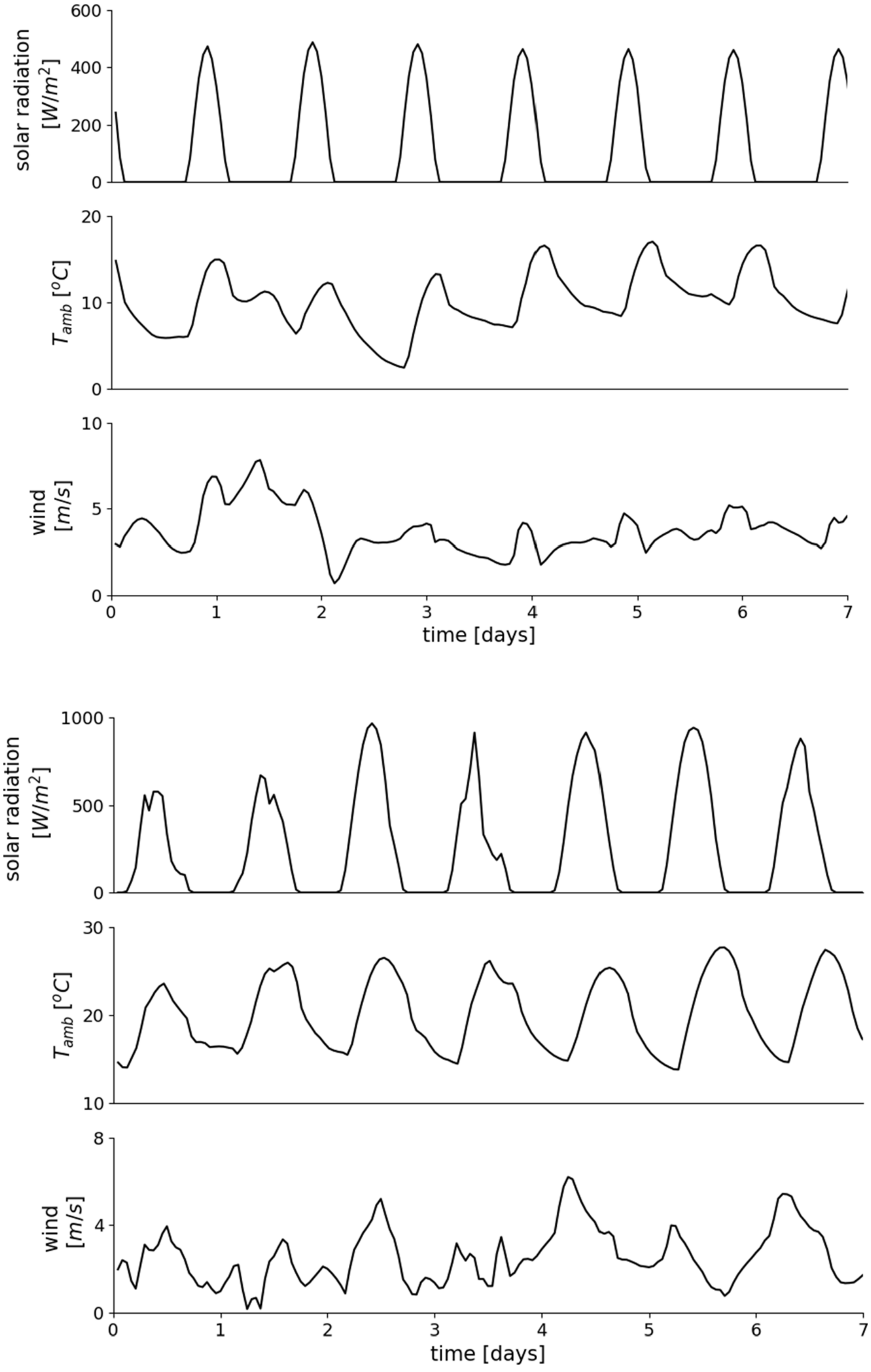

Fig. 1. Time variation of ambient conditions (solar radiation, temperature and wind velocity) for shipping container 2 (top) and shipping container 5 (bottom).

O. surinamensis, respectively. Afterward, ten adults of each species and population were introduced into each vial, using different vials for each species-population combination. For each fumigation trial, vials were placed in different locations within each container, including locations where sensors have been placed (Table 1). Additional vials with insects were used as controls and placed in untreated areas of each shipping container. After the termination of each fumigation trial, the vials were transferred to Laboratory of Entomology and Agricultural Zoology (LEAZ), Department of Agriculture, Crop Production and Rural Environment, University of Thessaly and adult mortality was noted. All vials were kept in incubators at the controlled conditions mentioned above and 65 days later progeny production was recorded.

2.2. *CFD-Simulations methodology*

CFD is a branch of fluid mechanics that uses numerical analysis and data structures to solve and analyze problems that include, but not limited to, fluid flows, heat, and mass transfer. A recent study published by Agrafioti et al. (2020b) presented a CFD model that was appropriate for phosphine fumigations and validated its performance in treated silos.

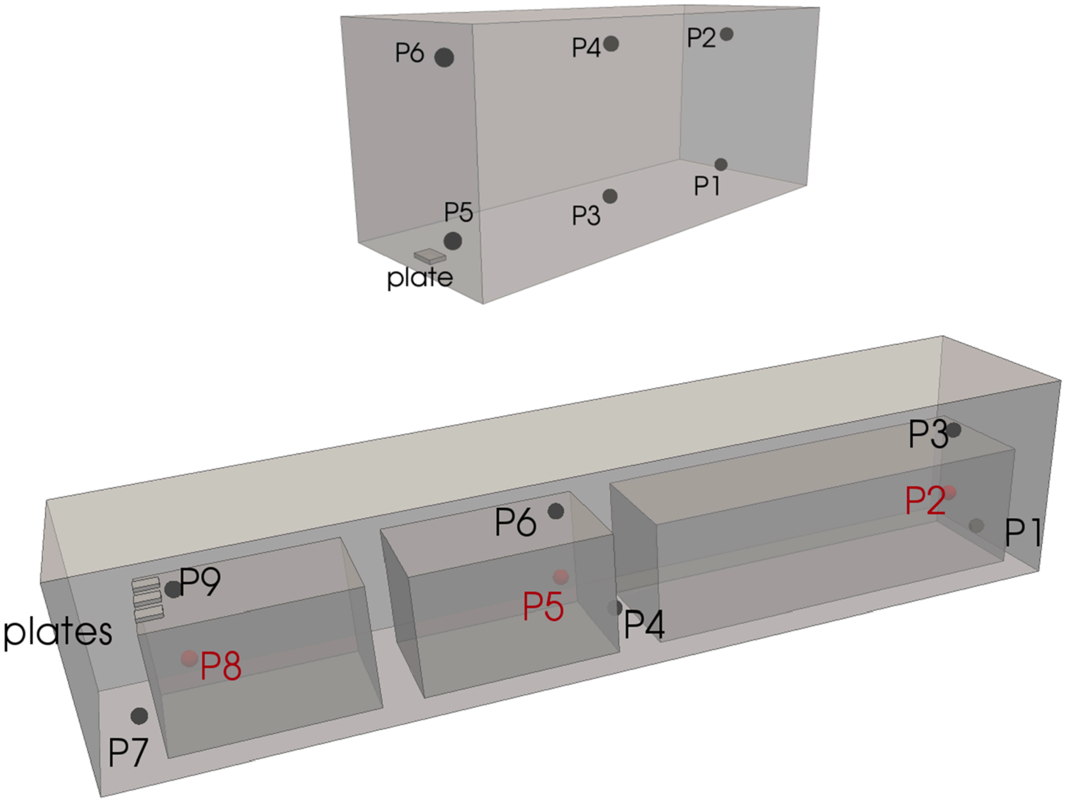

Fig. 2. The three-dimensional model of the two containers considered in this work (top: shipping container 2: empty container, bottom: shipping container 5: container with currants). Dots depict the location and the names of the wireless phosphine sensors. Furthermore, the position of the magnesium phosphide plates is shown.

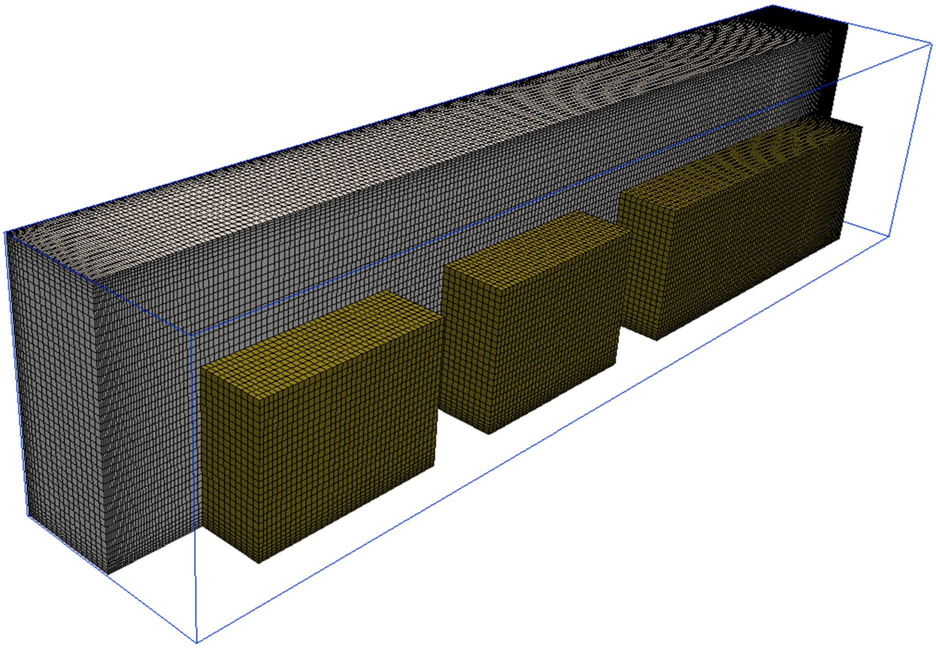

Fig. 3. The mesh used for the simulation of shipping container 5. The mesh resolution is increased near the walls of the container. The slice view reveals the pallets with currants (porous material) and their respective mesh.

In the aforementioned work, a detailed description of the model equations for air velocity, temperature, phosphine concentration, phosphine sorption, and their implementation in porous media was presented. The phosphine concentrations are associated with the mortality of stored-product insects based on an insect indicator function (Collins et al., 2005, Agrafioti et al., 2020b).

For the present work, two representative fumigation trials were further studied with the CFD methodology. The first one was an empty 20 ft container (shipping container 2) and the second one a 40 ft container with boxes filled with currants (shipping container 5)

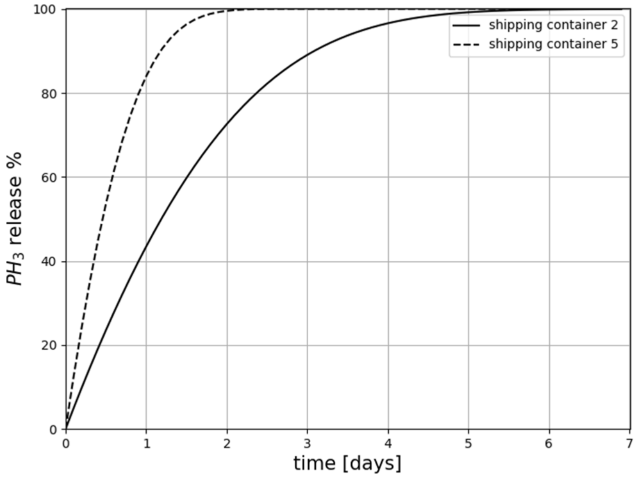

Fig. 4. Time evolution of the magnesium phosphide plates releasing phosphine gas (as a percentage of the total quantity available) for shipping containers 2 and 5. The gas release in the trial of shipping container 5 was considerably faster due to the higher ambient temperatures.

(Table 1). The cases were selected based on the variability of the key fumigation parameters (phosphine source, the influence of commodity, insect species, container size, weather conditions, etc.). The modeling methodology to address the other container fumigations is similar to the one presented below.

2.2.1 Boundary conditions

For accurate simulation predictions, the weather/ambient conditions of each location (Facilities 2 and 5) were considered as a boundary condition to the CFD model. An implementation of the boundary conditions to the model is described in Agrafioti et al. (2020b). In Fig. 1 the time series of ambient temperature, wind velocity, and solar radiation are presented which were used as inputs for the simulations.

2.2.2 Meshing

To solve the transport equations of the model, a discretizing procedure (meshing) of the geometrical domain (Fig. 2) is performed. In the present study, the mesh used for both facilities was structured, thus ensuring greater accuracy, while all cells were designed as hexahedra. For brevity, only the mesh of shipping container 5 is presented in Fig. 3 (mesh) since it is more complex than the empty container of shipping container 2. Furthermore, the mesh resolution was increased near the walls to properly capture large gradients. The number of cells for each case were approximately 200,000 and 410,000 for Facilities 2 and 5 respectively.

2.2.3 Phosphine gas release

To account for the phosphine gas release in the simulation model, the time evolution of the magnesium phosphide plates degassing was considered (Fig. 4). Modeling-wise, phosphine gas is inserted in the computational domain as a source term in the respective mass transport equation. In Fig. 4 is apparent that gas release in the trial of shipping container 5 was considerably faster due to higher ambient temperatures.

2.3. Wireless phosphine sensors

The phosphine concentration was measured with wireless sensors provided by Centaur Analytics (Centaur Analytics Inc., CA, USA). The devices are based on electrochemical sensors providing high accuracy and are equipped with wireless connectivity able to transmit data frequently (e.g. every 2 h) from inside the containers. The data were transmitted in real-time to a cloud platform, from which they were downloaded and further processed. The accuracy of the sensors for fumigation applications has been successfully evaluated by Brabec et al. (2019) and have been deployed to numerous fumigation applications (Kaloudis et al., 2018; Agrafioti et al., 2018, 2019, 2020a,b).

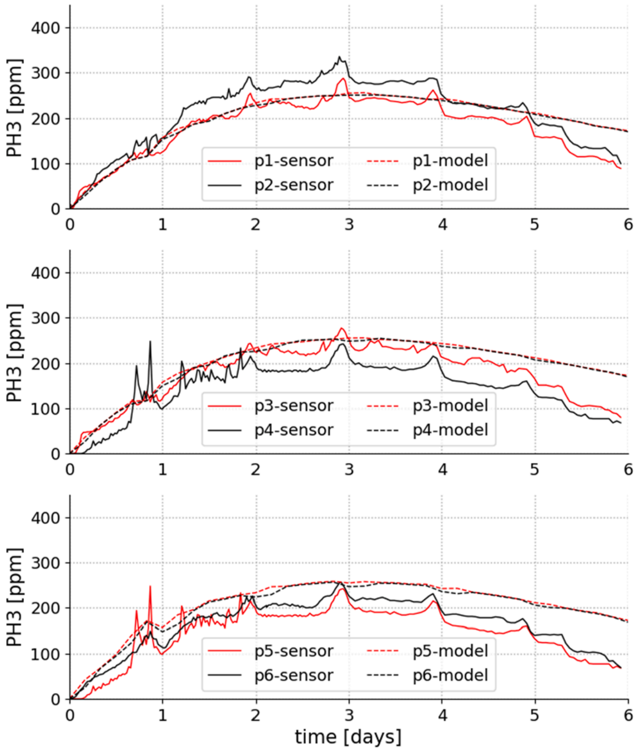

Fig. 5. Phosphine concentration (ppm) comparison of sensor data (solid lines) vs. simulation predictions (dashed lines) at 6 positions inside the container of shipping container 2.

In shipping container 2, six sensors were installed in the 20 ft container, three closer to the bottom and three near the top of the container (Fig. 2A). In shipping container 5, more sensors were installed to cover efficiently the larger size of the 40 ft container but also to evaluate the development of phosphine concentration inside the currant boxes (Fig. 2B).

2.4. Insects

The species used in the fumigations trials were adults of the lesser grain borer, *Rhyzopertha dominica* (F.) (Coleoptera: Bostrychidae) and the sawtoothed grain beetle, *Oryzaephilus surinamensis* (L.) (Coleoptera: Silvanidae), which are major pests of durable stored products. Both species were reared at the LEAZ, in incubators set at 25 °C, 65% relative humidity (r.h.) and continuous darkness, on soft wheat and oat flakes, for *R. dominica* and *O. surinamensis*, respectively. There were two populations from each species, one susceptible and one resistant to phosphine. The susceptible ones were the standard laboratory populations, which are kept in LEAZ for several decades, and the resistant populations labeled as *R. dominica* GA6 and *O. surinamensis* ASC11. The susceptibility and resistance of the above populations has been evaluated in a recent study by Agrafioti et al. (2019).

2.5. Data analysis on insect control

In all cases, control mortality was generally low, so the data for control mortality was not used in the analysis. All data, separately for each trial and insect species, were submitted to an Independent *t*-test,

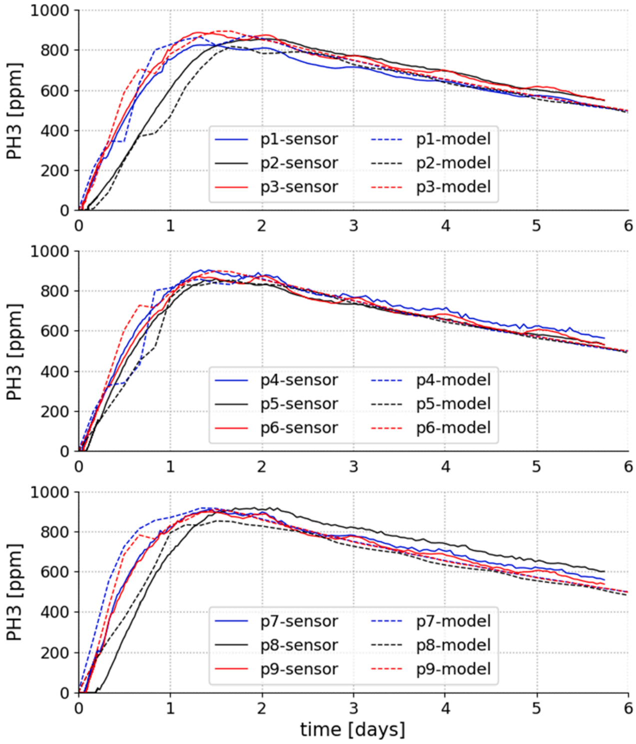

Fig. 6. Phosphine concentration (ppm) comparison of sensor data (solid lines) vs. simulation predictions (dashed lines) at 9 positions inside the container of shipping container 5.

with insect mortality as the response variable, to compare the two populations within each species. Means between resistant and susceptible populations of the same species were separated by using the *t*-test with a significance level set to 0.05. One-way ANOVA was used to determine the effect of location for each fumigation trial with insect mortality as the response variable and location as the main effect. The same procedure was followed in the case of progeny production.

3. Results

3.1. Phosphine gas distribution estimates and comparison with sensor data

The simulation process yielded the progress of phosphine concentration for the entire fumigation treatment as well as predictions of insect mortality. The predictions on phosphine concentration have been compared with sensor data to ensure the validity of the model (Figs. 5 and 6). The phosphine concentration (in ppm) comparison of sensor data with the simulation predictions at six positions inside the container (shipping container 2) is presented in Fig. 5. The dose for shipping container 2 was 1 Mg_3P_2 plate which released 33 gr of phosphine gas and for an empty, gas-tight 20 ft container should have resulted in approximately a concentration of 700 ppm. All sensors continuously reported concentrations below 350 ppm which was a clear indicator of gas losses. In general, phosphine concentration had similar values for all positions, with no apparent stratification. The local maximum observed on the sensor time series had a daily variation and coincided with the rise of temperature and solar radiation (Fig. 1).

Since the computational model took into account temperature variations from the ambient, air movements were also considered for their contribution to phosphine concentration. A minor discrepancy between the model and sensor data was observed on the final day of the fumigation process probably due to a higher gas loss rate. The gas loss rate

Table 2
Post-fumigation mortality (% ± SE) of parental adults of four insect populations in different types of facilities on which phosphine had been applied, and respective progeny production (number of adults per vial ± SE) 65 d later.

Shipping container number	Population	Mortality %	Mortality range among locations	Progeny production	Progeny production range among locations
1 (empty)	*O. surinamensis* ASC11	97.8 ± 1.3	93.3–100	0.2 ± 0.1	0.0–0.6
	O. surinamensis Lab*	100 ± 0.0	100–100	0.0 ± 0.0	0.0–0.0
	R. dominica GA6	17.2 ± 1.5 a	13.3–23.3	8.2 ± 0.1 a	7.6–8.6
	R. dominica Lab	99.4 ± 0.6b	96.6–100	0.1 ± 0.0b	0.0–0.3
2 (empty)	*O. surinamensis* ASC11	86.6 ± 4.0 a	76.6–100	2.2 ± 0.7 a	0.0–7.0
	O. surinamensis Lab	100 ± 0.0b	100–100	0.0 ± 0.0b	0.0–0.0
	R. dominica GA6	12.1 ± 5.4 a	3.3–100	8.8 ± 0.5 a	6.6–9.6
	R. dominica Lab	98.8 ± 0.7b	96.6–100	0.2 ± 0.1b	0.0–0.3
3 (empty)	*O. surinamensis* ASC11	73.8 ± 4.2 a	56.6–90.0	10.9 ± 3.7 a	3.3–25.6
	O. surinamensis Lab	100 ± 0.0b	100–100	0.0 ± 0.0b	0.0–0.3
	R. dominica GA6	11.6 ± 4.4 a	0.0–40.0	12.5 ± 2.0 a	4.3–22.3
	R. dominica Lab	89.5 ± 2.9b	86.6–100	1.9 ± 0.6b	0.0–5.3
4 (currants)	*O. surinamensis* ASC11	100 ± 0.0	100–100	0.0 ± 0.0	0.0–0.0
	O. surinamensis Lab	100 ± 0.0	100–100	0.0 ± 0.0	0.0–0.0
	R. dominica GA6	100 ± 0.0	100–100	0.0 ± 0.0	0.0–0.0
	R. dominica Lab	100 ± 0.0	100–100	0.0 ± 0.0	0.0–0.0
5 (currants)	*O. surinamensis* ASC11	100 ± 0.0	100–100	0.0 ± 0.0	0.0–0.0
	O. surinamensis Lab	100 ± 0.0	100–100	0.0 ± 0.0	0.0–0.0
	R. dominica GA6	100 ± 0.0	100–100	0.0 ± 0.0	0.0–0.0
	R. dominica Lab	100 ± 0.0	100–100	0.0 ± 0.0	0.0–0.0
6 (currants)	*O. surinamensis* ASC11	100 ± 0.0	100–100	0.0 ± 0.0	0.0–0.0
	O. surinamensis Lab	100 ± 0.0	100–100	0.0 ± 0.0	0.0–0.0
	R. dominica GA6	100 ± 0.0	100–100	0.0 ± 0.0	0.0–0.0
	R. dominica Lab	100 ± 0.0	100–100	0.0 ± 0.0	0.0–0.0

Within each shipping container and each species, means followed by different letters are significantly different, according to Student's *t*-test at $P < 0.05$. According to *t*-test, the parameters ***for parental mortality*** were: in shipping container 1 for *R. dominica* $t = -49.1$, $P < 0.001$, in shipping container 2 for *O. surinamensis* $t = -3.2$, $P = 0.002$, for *R. dominica* $t = -15.7$, $P < 0.001$, in shipping container 3 for *O. surinamensis* $t = -6.0$, $P < 0.001$, for *R. dominica* $t = -14.5$, $P < 0.001$.
According to *t*-test, the parameters ***for progeny production*** were: in shipping container 1 for *R. dominica* $t = 46.6$, $P < 0.001$, in shipping container 2 for *O. surinamensis* $t = 2.9$, $P = 0.006$, for *R. dominica* $t = 15.4$, $P < 0.001$, in shipping container 3 for *O. surinamensis* $t = 2.9$, $P = 0.006$, for *R. dominica* $t = 4.9$, $P < 0.001$. In all cases $df = 34$. Where no letters exist no significant differences are noted.
* "Lab" indicates phosphine susceptible insect populations.

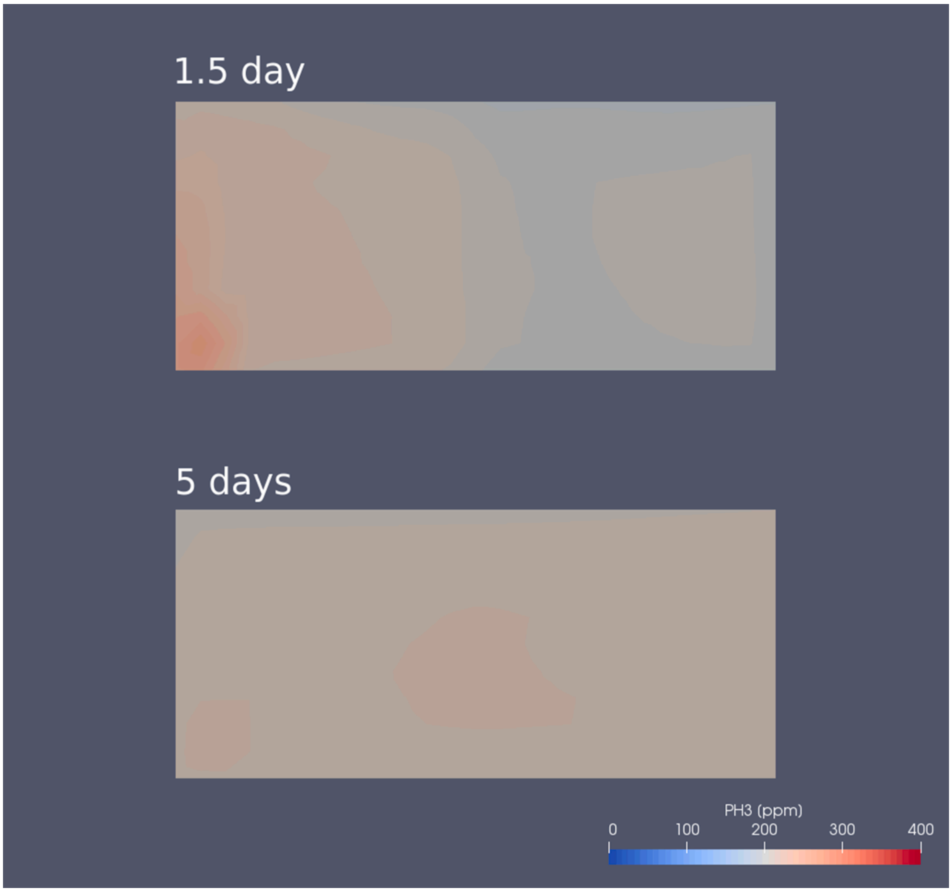

Fig. 7. Estimated phosphine concentration profiles (two-dimensional, mid-section view) at two time instances for shipping container 2.

coefficient is calculated based on an empirical equation that considers, among others, the wind velocity (Agrafioti et al. 2020b). The historical weather data were retrieved from a data service (www.soda-pro.com) and deviations from the actual wind velocity could be present due to microclimate conditions (e.g., a nearby building affecting the wind profile). A weather station installed on-site could potentially improve the simulation predictions. Nonetheless, the discrepancy of the 100 ppm and its duration was not sufficient to alter the predictions on the insect mortality. Specifically, the computational model predicted that there were no areas where 99.9% mortality was ensured which was in agreement with the trial results (see below for a comprehensive analysis).

The phosphine concentration comparison of sensor data with the simulation predictions at the nine locations (Fig. 2) inside the container of shipping container 5 is presented in Fig. 6. The dosage for this treatment was 3 Mg_3P_2 plates, which for an empty, gas-tight container, the phosphine concentration is expected to reach 1050 ppm. The sensor data in Fig. 6 showed a maximum concentration of 900 ppm and then a decline to 600 ppm at the end of the 6th day. This progress of phosphine concentration was expected due to the sorption effect by the fumigated commodity (currants) and the gas losses to the ambient. Furthermore, the maximum concentration was observed during the 2nd day, which was approximately 24 h faster than the treatment of shipping container 2.

In Fig. 6, the black lines indicated sensors that were placed inside the currant boxes (sensors P2, P5, P8) as shown in Fig. 2. As expected, there was a time delay for phosphine to reach these places due to the limited gas permeability of the carton boxes. This phenomenon was visible during the first 24 h of the treatment (Fig. 6) and as time progress, an equilibrium was reached between the concentrations on the inside and the outside of the boxes filled with currants. Similar to shipping container 2, almost all sensors reported the same concentration levels indicating intense mixing due to air flows originated by temperature gradients. The metal structure of a container was quite responsive to the daily temperature differences and boosted the phosphine gas mixing.

The predictions of the computational model were in agreement with the phosphine concentrations of the sensor data with few discrepancies, particularly in the first 24 h. Nevertheless, the computational model accurately predicted all the phenomena described above (time of maximum concentration, the time delay of P2, P5, P8 sensors, and the equilibrium of all sensors from the 2nd day and onward). Concerning insect mortality, the computational model predicted complete insect control for all locations of the container which was validated with the results of the field trials (Table 2).

3.2 Predictions on each location in the storage space

The overall performance of the CFD model was considered satisfactory, ensuring the validity of the phosphine concentration and insect mortality predictions for the entire container space as the ones presented in Figs. 7–9. Fig. 7 shows the phosphine concentration profiles (two-dimensional, mid-section view) at two-time instances for shipping container 2. The top view shows the prediction after 36 h since the treatment started. On the lower left (near the container door), an area of higher phosphine concentration was recorded. This is the position where the Mg_3P_2 plate was placed and at that time, the gas release process was still active. For the rest of the container area, the phosphine concentration was quite uniform for the reasons explained above (temperature gradients). The bottom view showed the phosphine profile after five days. Since the gas release has ended, there was no higher concentration near the door of the container and no stratification or other areas of higher concentration.

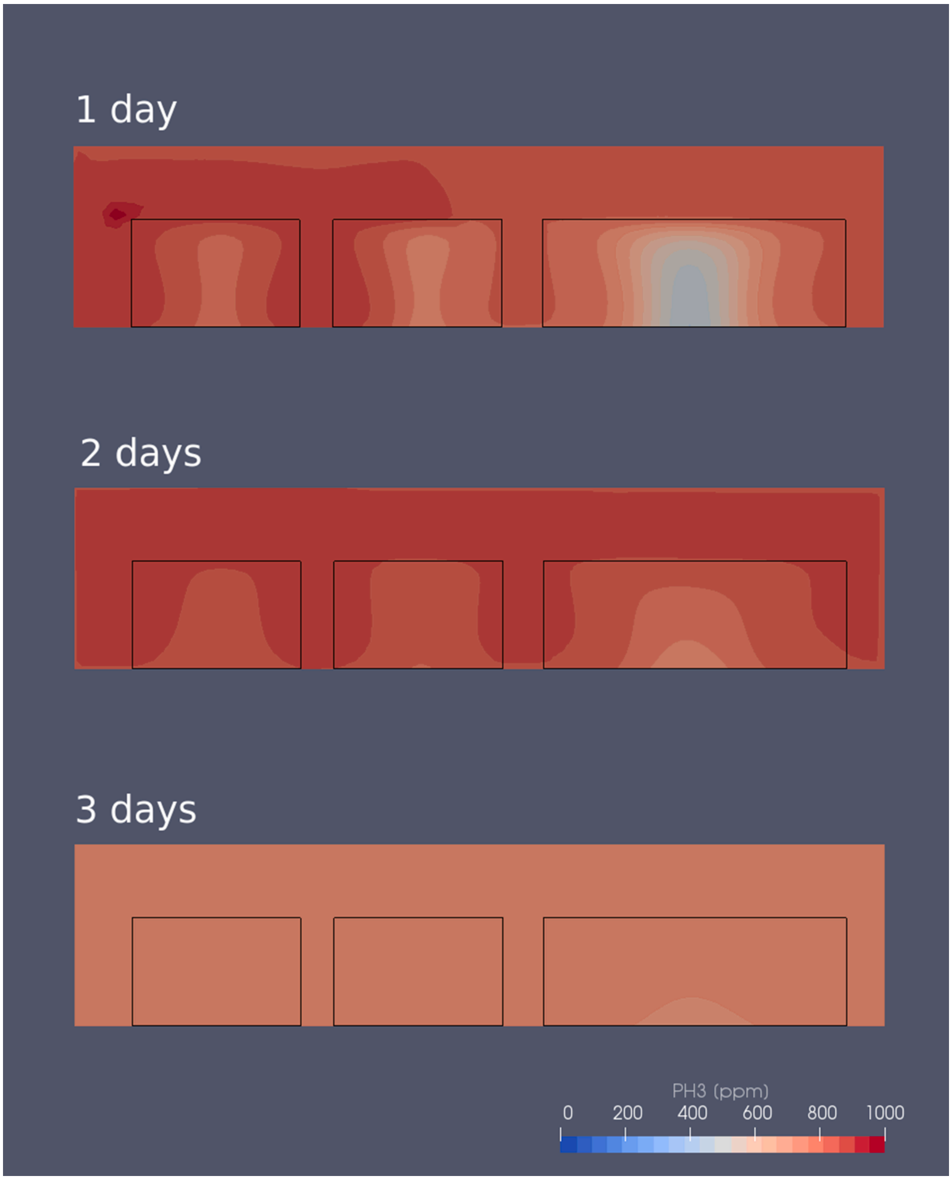

Fig. 8. Estimated phosphine concentration profiles (two-dimensional, mid-section view) at three time instances for shipping container 5.

In Fig. 8 the respective phosphine concentration profiles (two-dimensional, mid-section view) at three instances are shown for shipping container 5. The presence of pallets with boxes filled with currants disturbed the homogenous phosphine concentration profiles presented in Fig. 7. Specifically, at the end of the 1st day (top view), the air between the pallets had an almost uniform concentration near to 800 ppm except for the area near the front of the container where the three Mg_3P_2 plates were placed and were still releasing gas. Inside the boxes, the delay of the phosphine gas penetration is apparent. The largest stack of boxes, at the back of the container, had the lowest concentration, approximately 300 ppm. At the end of the 2nd day (middle image of Fig. 8), similar phosphine concentration profiles were obtained. The concentration outside the boxes was uniform and lower concentration was predicted inside the boxes. By the end of the 3rd day (bottom image of Fig. 8), the phosphine gas had evenly penetrated almost in every box of the stack, and equilibrium with the air concentration had been reached.

A useful augmentation of the phosphine concentration profiles was the prediction of insect control using the insect mortality model. Fig. 9 presents two-dimensional (mid-section view) insect mortality profiles, at three-time instances, for shipping container 5, depicting the areas in which *R. dominica* could not survive the fumigation. As expected, the areas near the Mg_3P_2 plates were the first ones that reached lethal levels, but there was a time delay in reaching higher concentration levels within the commodity. The profile in the middle (4.5 days), shows the progress of the insect mortality zones, where an area in the middle of the larger boxes stack at the back of the container has not yet reached complete insect control. Finally, at the end of the 5th day (bottom view) all areas in the container were treated successfully leaving no alive insects (Fig. 9). The field tests confirmed the simulation results as all insects were found completely controlled. Moreover, even though the model used was based on *R. dominica*, we found that *O. surinamensis* was completely controlled as well. Finally, for both species, our measurements showed complete mortality, regardless of the resistance levels of the populations tested (Fig. 9, Table 2). The same holds for progeny production capacity.

3.3. *Mortality and progeny production*

Regarding parental mortality, for all tested populations (resistant

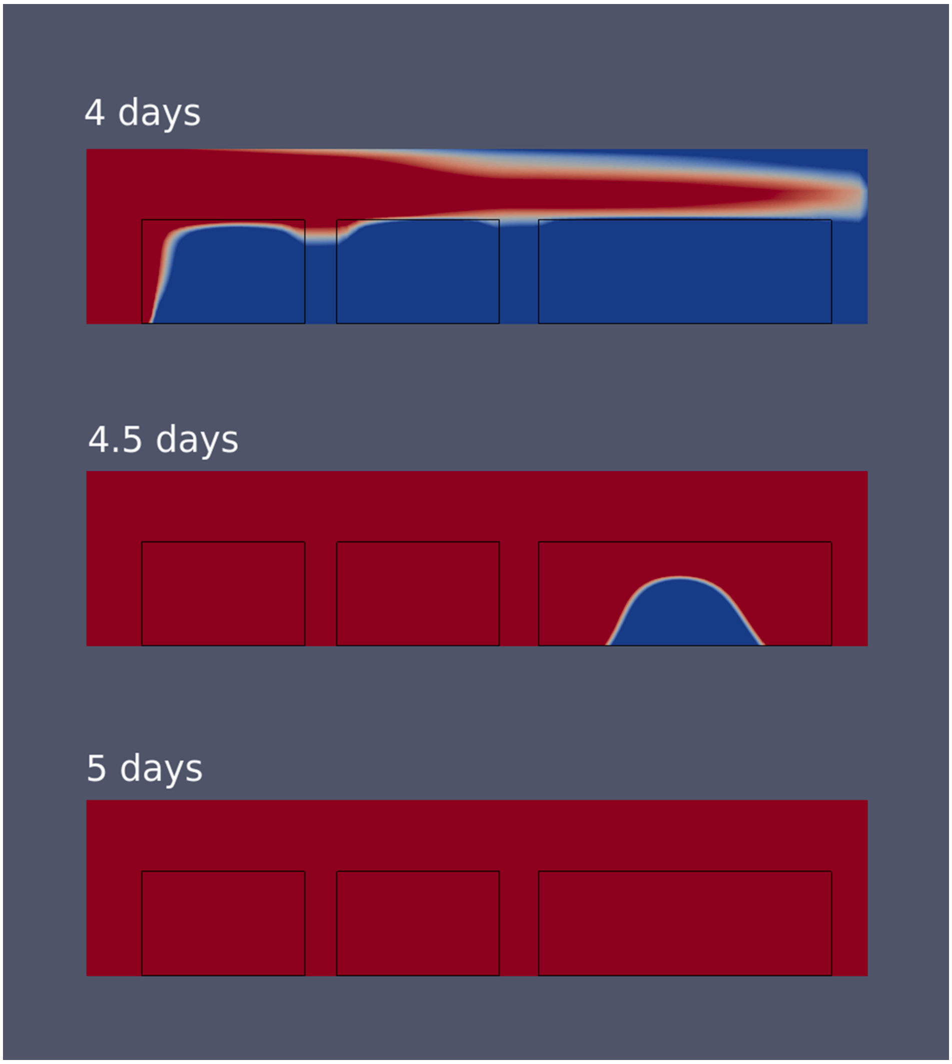

Fig. 9. Insect mortality profiles (two-dimensional, mid-section view) at three time instances for shipping container 5. Red color indicates zones with 99.9% insect mortality. Black boxes show the pallets filled with currants. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

and susceptible to phosphine) and locations, complete control was observed in most of the fumigations performed (Table 2). No significant differences in mortality levels were showed between resistant and susceptible insect populations within the tested locations in shipping containers 4, 5 and 6 (Table 2).

Regarding progeny production, no offspring emergence was noted with some exceptions which ranged between 1 and 13 adults per vial, depending on the population (Table 2). Significant differences in progeny production were observed between susceptible and resistant populations to phosphine in shipping containers 1, 2, and 3 (Table 2).

4. Discussion

Phosphine is highly effective for the control of stored product insects in an extremely wide range of storage facilities and commodities (Collins, 2010; Athanassiou et al., 2016; Agrafioti et al., 2020a,b). In the present study, we have estimated the spatio-temporal movement of phosphine in commercial containers by using wireless phosphine sensors. The distribution of phosphine gas in those containers was modeled and compared with available sensor data. Based on the results of the present work, the CFD model used can be used with success in fumigations in containers to predict the gas distribution and the concomitant insect mortality. So far, this model has been evaluated successfully only in the case of silos (Agrafioti et al., 2020b), and hence, this is the first time that the same model is validated under a “container-based” fumigation approach.

Two of the most important causes of failure in commercial fumigations is that phosphine is applied in leaky and poorly-sealed structures (Agrafioti et al., 2020a) and that phosphine absorbed by the commodity is rarely taken into consideration (Reddy et al., 2007). Apart from the predicted concentrations, and taking into account that phosphine is solely used for insect control, a fumigation can be considered as a failure if the target species are not effectively controlled for periods exceeding the duration of the fumigation process (e.g. offspring). Agrafioti et al. (2020a) found that fumigations in warehouses, ships, and silos are likely to fail due to increased leakages, which cannot be easily detected with the majority of the phosphine detection techniques. On the other hand, the commercial treatments on containers were the “best-case scenario”, since all insect life stages were dead, including the eggs, even for phosphine-resistant populations (Agrafioti et al., 2020a). In that study, after the initiation of the fumigation, a noticeable concentration increase was noted in all tested locations, which remained at high levels, i.e. 1500–2000 ppm during the entire treatment period, despite variations among locations. Moreover, in containers, phosphine distribution can be much more uniform than other larger structures, resulting in high insect mortality levels (Athanassiou et al., 2016; Agrafioti et al., 2018; 2020a).

Results of the present study, indicate that phosphine concentration levels are similar among the different locations in an empty container, as, for instance, in the case of shipping container 2 where phosphine concentrations were similar in all areas throughout the entire fumigation time, which correlates well with our model. The relatively small size of the container, led to, as expected, a uniform distribution of phosphine. However, in the cases of containers loaded with packaged products, the phosphine concentration profiles were altered. In those cases, the presence of the bulk commodity is likely to create an uneven gas distribution at the beginning of the fumigation, right after the application of phosphine. Increased concentrations were reported near the locations where the phosphine formulations (plates, etc.) are placed, behind the container door, and lower concentrations at the “far back” of the container. Still, after this interval, which we estimate in our study to last for 24 h or even less, a gas equilibrium is reached. In that case, the central part of the pallets or bulked commodities is expected to receive eventually less phosphine than the peripheral part.

Fumigant concentrations can be influenced by several factors including environmental conditions (temperature and moisture/humidity) (Reed and Pan, 2000; Darby, 2011), wind speed (Cryer, 2008), and sorption from the commodity (Daglish and Pavic, 2008). Based on our results, in Facilities 1, 2, and 3 (empty containers) the gas distribution was considerably affected by the natural convection currents, which can eventually reduce the gas concentration in leaky containers (Kaloudis et al., 2018). On the other hand, in Facilities 4, 5, and 6, the presence of boxes with currants might have partially alleviated this phenomenon, as air movement was reduced due to the presence of the product, at least as compared with the empty containers. Sorption is known to effect the concentration of phosphine, although phosphine sorption in the case of currants has not been studied in detail (Reddy et al., 2007; Daglish and Pavic, 2008; Darby, 2008). Apart from the apparent problems that sorption causes in a fumigation process, it seems that due to this phenomenon, the gas remains for longer periods close to the fumigated commodity, providing a gradual desorption activity (Plumier and Maier, 2018). This hypothesis needs to be investigated further.

Our bioassays with *R. dominica* and *O. surinamensis* show that there were no variations in mortality levels among populations of different susceptibility to phosphine, which could be attributed to increased gas concentrations. Thus, elevated phosphine concentrations maintained for sufficient periods, can control resistant individuals. Interestingly, in the case of the empty containers, parental survival, along with the concomitant progeny production, was much higher for the resistant populations, as compared with the susceptible populations. This is due to the fact that, despite the application of high dose of phosphine, concentration remained at relatively low levels, for reasons mentioned above. Low concentrations, significant gas fluctuations among locations, as well as and short exposures are responsible for increased insect survival, especially for resistant populations (Kaloudis et al., 2018; Agrafioti et al., 2020a; Nayak et al., 2020). Conversely, when the fumigations were carried out in loaded containers, complete (100%) parental mortality was achieved, which resulted in zero progeny production in the treated substrate. In these fumigations, the concentrations were higher than those of the empty containers, and remained at high levels in all locations, for the entire fumigation period. These mortality results were in agreement with the CFD model used in the present study.

The objective of this study was to correlate the phosphine wireless sensor data concerning the phosphine distribution and insect mortality in containers with a prediction model by using wireless sensors. Based on the results, the validation was successful, as the model can accurately predict distribution patterns and insect mortality, which may also occur in containers that are only partially loaded with product. At the same time, it was demonstrated that, if best management practices are followed in phosphine applications, insect control is high, regardless of the resistance status of the target populations. The proposed model can be further utilized in vessel/in transit fumigations, through a precision fumigation-oriented strategy, that provides real-time estimates for insect control.

CRediT authorship contribution statement

Paraskevi Agrafioti: Conceptualization, Methodology, Formal analysis, Investigation, Funding acquisition. **Efstathios Kaloudis:** Software, Validation. **Sotiris Bantas:** Validation, Software. **Vasilis Sotiroudas:** Conceptualization, Methodology, Investigation, Resources, Project administration. **Christos G. Athanassiou:** Writing - review & editing, Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This research is co-financed by Greece and the European Union (European Social Fund- ESF) through the Operational Programme «Human Resources Development, Education and Lifelong Learning» in the context of the project “Strengthening Human Resources Research Potential via Doctorate Research” (MIS-5000432), implemented by the State Scholarships Foundation (IKY).

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.compag.2021.106087.

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