

Numerical simulation of ozone concentration profile and flow characteristics in paddy bulks

R. Pandiselvam^{a*}, V. Chandrasekar^b and V. Thirupathi^c

^{a*}Physiology, Biochemistry and Post Harvest Technology Division,

^{a*}ICAR –Central Plantation Crops Research Institute, Kasaragod - 671 124, Kerala, India

^bICAR-Central Institute of Post Harvest Engineering and Technology, PAU post
Ludhiana, Punjab – 141004, India

^cDryland Agricultural Research Station, Chettinad – 630 102,

^cTamil Nadu Agricultural University, Coimbatore, India

*Corresponding author e-mail: anbupandi1989@yahoo.co.in

Abstract

BACKGROUND: Ozone has shown the potential to control the stored product insect pests. The high reactivity of ozone makes special problems when it passes through an organic medium such as stored grains. Thus, there is a need for simulation study to understand the concentration profile and flow characteristics of ozone in stored paddy bulks as a function of time.

RESULTS: Simulation of ozone concentration through the paddy grain bulks was explained by applying the principle of law of conservation along with continuity equation. A higher ozone concentration value was observed at regions near the ozone diffuser whereas a lower concentration value was observed at region away from the ozone diffuser. The relative error between the experimental and predicted ozone concentration values for the entire bin geometry was less than 42.8%.

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CONCLUSION: The simulation model described a non linear change of ozone concentration in stored paddy bulks. Results of this study provide the valuable source for estimating the parameters needed for effectively designing a storage bin for fumigation of paddy grains in a commercial scale continuous-flow ozone fumigation system.

Keywords: fumigation; ozone; diffusion; saturation time; half-life

1 INTRODUCTION

Ozone is a potential insecticide and powerful antimicrobial agent that has many beneficial applications in managing insect pests, fungi and micromycetes spore development in stored grains. The U.S. FDA approved its use as a direct additive for food.¹ It is attractive in food industry because ozone auto-decomposes into oxygen,² leaving no hazardous residue,³ can be generated on-site using oxygen cylinder/oxygen concentrator and ozone generator⁴ and eliminating the need to store chemicals.⁵ In addition to that energy input required for ozone fumigation treatment is much lower than thermal, radiation and microwave treatment.⁶ It requires only 4-5 kW.h of energy for processing of one ton of grain but electromagnetic field (microwave) required around 91 to 130 kW.h.⁷

Ozone is relatively stable molecule at low temperature and high relative humidity condition, only at higher concentration and higher temperature it rapidly decays to oxygen.⁸ Therefore low concentration and long exposure time is more practical,⁹ easy to generate, no significant changes in proximate composition of stored grain¹⁰ and environmentally friendly.¹¹ The problems in ozone application for insect control in stored

gains should be solved closely related with the grain bed thickness, moisture content,⁴ and selection of ozone concentration, exposure time,¹² mycobiotic contamination of grain surface and concentration of the ozone in outlet.⁹

The efficacy of ozone on stored product insect pests are presented in many scientific papers, however there is a paucity of information available in the published literature on the bulk flow behavior and concentration profile of ozone for the commercial application of ozone as a fumigant in stored grains.⁴ Therefore the bulk flow behavior and concentration profile of ozone in stored grains bulks needs to be understood before it can be used as a fumigant. High reactivity of ozone introduces several problems and ozone decay rate is easily accelerated by many types of organic and inorganic medium. Moreover studies of ozone application are mostly experimental, so they cannot be universally applied to different types of storage systems.

Hence, to verify the adaptability of ozone fumigation system in silos a study on the flow of ozone is highly essential. Therefore, this study was undertaken to investigate the flow characteristics and numerical simulation of ozone concentration in stored paddy bulks as a function of time.

2. MATERIALS AND METHODS

2.1 Sample preparation

Paddy (ADT 43) was procured from the local mill in Thanjavur of Tamil Nadu, India. The moisture content of the paddy was determined by oven method.¹³

2.2 Ozone fumigation system

Investigations on a bulk flow of ozone through paddy column were conducted at Indian Institute of Crop Processing Technology (IICPT), Thanjavur, India. Ozone

diffusion system consists of an oxygen concentrator, ozone generator, pilot scale storage bin (100 kg), ozone analyzer and ozone destructor.

2.2.1 Oxygen concentrator

The ozone generator fed with oxygen, generated by oxygen concentrator developed by Creative Oz-Air private limited, Noida, Uttar Pradesh, India (OZ-AIR HG5; Oxygen flow rate – 5 LPM maximum; Power consumption - 750 Watts; Oxygen pressure- 20-100 PSIG) with a purity level of greater than 95 percent. It separates nitrogen, oxygen and other gasses from the atmospheric air based on the principle of swing adsorption. During oxygen separation, it was ensured that there is no strong magnetic field around the equipment.

2.2.2 Ozone generator

Ozone gas was generated using ozone generator (ISM 5, material used-stainless steel; the size of ozone generator- 600×500×300 mm; Power consumption - 350 watts) developed by Creative Oz-Air PVT. LTD, Noida, Uttar Pradesh, India. It works under the principle of corona discharge method in which a spark is used to split the diatomic oxygen molecule into single valance oxygen atoms to produce ozone.

2.2.3 Pilot scale storage bin

Pilot scale storage bin was connected with an ozone generator to distribute the gaseous ozone evenly throughout a grain mass. The bin was made up of food grade stainless steel (SS 304) at 1.5 m height and 0.6 m diameter. It was designed in such a way that the grains can be loaded at the top and unloading can be done at the bottom. Ozone gas passed at the bottom of the storage bin. To measure the ozone concentration in the

storage bin, five concentration ports were placed (L_1 to L_5). L_1 is the lower level port where L_5 is the higher level port. L_1 is placed at a distance of 0.385 m from the bottom and the remaining ports were placed at a distance of 0.20 m. Each level of the concentration port has been fitted with a brass ball valve and connected with T and L-bend, with the help of 10 mm diameter polyurethane hose pipe. Each of the valves is interlinked and it is connected directly to the ozone analyzer. The ozone concentration can be measured at each port by turning the valves. Gas leakage was prevented with the help of gasket provided at the loading and unloading part of the storage bin. The bin consists of the perforated sheet at the bottom. The holes in the perforated sheet are made at 2 mm diameter, which helps to distribute the ozone gas uniformly throughout the bin.

2.2.4 Ozone analyzer

At every 5 min interval the ozone concentration present in paddy column was recorded by an ozone analyzer (BMT MESSTECHNIK GMBH-964, Gueterfelder Damm, Stahnsdorf, Germany). It is a dual beam photometer (UV 254 nm). The ozone concentration is displayed either in ppm, %wt/wt, or g/Nm^3 on a 16 character alphanumeric display.

2.2.5 Ozone destructor

The thermal ozone destructor developed by Creative Oz-Air private limited, Noida, Uttar Pradesh, India (Material: Size: 150D×500L (mm) Inlet and outlet size: $\frac{1}{4}$ "BSP, 50 Hz, Power consumption: 800 Watts, Power supply: 230V/AC, Insulating material: Glass wool and Temperature: 200°C) was connected with ozone analyzer to convert the ozone into oxygen. Where the left over ozone gasses passes through the zigzag path in

a destructor, due to high temperature the ozone was destructed to oxygen and released to the atmosphere.

2.3 Numerical modeling

Concentration profile and flow characteristics of ozone in the paddy bulks cannot be considered as constant during fumigation because it is the function of ozone concentration, time and bed thickness. Hence, a one-dimensional numerical model by applying the principle of law of conservation along with continuity equation was used to assess the concentration profile and flow characteristics of ozone of in the paddy bulks. Cylindrical coordinates were selected for the equation development. First law of thermodynamics is used for developing conservation equation for the fluid flow in the packed element at any time.¹⁴

Assumptions made for the numerical simulation are a) stored paddy bulks consist of smaller size grains compared to the bed dimension; b) grains are considered as uniform in size, shape, continuous mass and free flowing at same moisture content and variety; c) void fraction/porosity (void space in between the grains) of the grains are uniform throughout the bed; d) all the stored paddy bulks have the same physical, mechanical and aero dynamical properties in any direction of bed; e) frictional forces of grains are negligible during fluidization; f) grains are linearly distributed in ozone entry zone; g) effect of broken grains, foreign materials and bed compaction are not considered in mathematical model and h) ozone diffusion takes place in the vertical (Z axis) axis of the storage bin in unsteady steady state condition.

$$\frac{\partial c}{\partial t} = D_e \left(\frac{\partial^2 c}{\partial z^2} \right) - v_z \left(\frac{\partial c}{\partial z} \right)$$

The algebraic form of the equation is

$$c_{i,j+1} - c_{i,j} = A(c_{i-1,j+1} - 2c_{i,j+1} + c_{i+1,j+1} + c_{i-1,j} - 2c_{i,j} + c_{i+1,j}) - B(c_{i,j+1} - c_{i,j})$$

$$A = \frac{\partial t D_e}{2 \partial z^2}$$

$$B = \frac{\partial t}{\partial z}$$

where, c is Ozone concentration, %; t is time, s; D_e is diffusivity, m/s; and V_z is velocity of ozone in 'Z' direction.

2.4. Solution for numerical model

Experiments were conducted **thrice** to measure the ozone concentration profile and flow characteristics of ozone in the stored paddy bulks at $31 \pm 2^\circ\text{C}$ and 58% RH. The concentration of the ozone from ozone generator was 500 ppm. The ozone concentration was measured at every 5 min interval from each port. After summarizing experimental results, the numerical differentiation equation was linearized in to algebraic equation using finite difference method. A MATLAB (MATLAB 7.5, MathWorks, Inc, Massachusetts, USA) code was written to solve algebraic equation. Predicted values from the simulation and the experimental results were compared.

3 RESULTS

Simulated ozone concentration in paddy grains over the entire bin geometry presented in Fig. 1. Being the measured value, the time interval for ozone concentration prediction fixed at 5 min (Time step 1) for the simulation study. The governing partial differential equation (continuity equation) and the associated boundary conditions, on which the model was based, assume that the mechanism of transport through the porous

media is only convective mass transfer. The moisture content of the paddy grain taken for the study was 11.86 ± 0.11 (%) (d.b.). Two important parameters for ozone flow through the paddy media were the bulk density and the porosity of the sample; known to be 568 kg m^{-3} and 0.46, respectively.^{15,16}

Fig.1 shows that ozone concentration increases with increase in time. The decrease in ozone concentration values observed with increasing bed thickness (Fig 2). Fig. 3 depicted that at port L_1 , the concentration of ozone gas at the center of the bin was parallel to the bin floor. However, the denser contour lines observed in regions nears the perforated floor results in non-uniformity of ozone distribution within the stored paddy bulks.

The ozone concentration shows a significant decrease from 500 ppm (0.2 m) to 125 ppm (0.6 m) at 685 min fumigation time. The simulations run with the assigned boundary conditions and the initial concentration at every node in the grid set zero concentration. From the Fig. 4, it was observed that the regression line follows diagonal pattern. It shows that the predicted concentration has higher accuracy. The relative error of prediction and chi-square was calculated as

$$\text{Relative error (\%)} = \frac{(O_{\text{experimental}} - O_{\text{predicted}})}{O_{\text{predicted}}} \times 100$$

$$\text{Chi-square} = \frac{(O_{\text{experimental}} - O_{\text{predicted}})^2}{O_{\text{predicted}}} \times 100$$

The predicted ozone concentration data was compared against experimental data presented in Table 1. The predicted ozone concentrations were higher than the measured concentrations at every sampling point (up to 6 h). The relative error between the experimental and predicted values for the entire bin geometry ranged from 42.8 percent at

1 h to 1.04 percent at 11 h. The chi-square value (df=10; P<0.01) varied from 0 (2 h) to 10.64 (4 h). At sampling time 6 h and above, the predicted ozone concentrations were close to the measured values.

4 DISCUSSION

Previous experimental results confirmed that ozone absorption in the grain layer influenced by grain column height¹⁷ and moisture content.⁵ Pandiselvam *et al.*⁵ concluded that ozone penetration through the stored grains is inversely proportional to the bed thickness. Steponaviciene *et al.*⁹ found that ozone absorption higher in upper layers of the grains column. The effectiveness of ozone is decreases in insect pests control obtained in stored products with higher bed thickness.¹¹ The non uniformity of ozone distribution may be due to the variations in concentration gradient between such regions and regions away from the floor which creates high and low ozone concentration zones within the bulk grain. However, with increase in grain bed thickness, the lines take a parabolic shape, which indicates an increase in grain bed thickness, the parallel ozone flow paths begin to change to non-parallel lines.

The flow of ozone inside the grain bulks depends on property of ozone such as density, oxidation rate based on temperature and paddy grains property (porosity, moisture content, surface characteristic and orientation of grain). The high oxidation rate of ozone poses some distinctive challenges to meet desirable concentration of ozone through a paddy grain mass. Ozone degrades to oxygen within 1 h. It shows that ozone has an initial diffusion problem through the commodity, due to decomposition of ozone reducing its ability to kill the insect pests at deeper levels. The interaction of one ozone

molecules into other ozone molecules results in a breakdown of ozone to oxygen and, thus also a reduction in ozone concentration.¹⁸ Atmospheric temperature, relative humidity and ozone flow rate are also important factors for the half-life time of ozone. McClurkin and Maier¹⁸ found that ozonation of grain will be more effective in still air at low temperature and high humidity.

Ozone movement inside the pore space between grains not only depends on diffusion, because mass flow also occurred due to continuous generation by ozone generator. The time required to reach desired concentration is also based on flow rate, initial concentration and ozone delivery pattern. Fully perforated storage bin floors would create parallel ozone flow streamline and lower the risk of non ozonation zones in the grain at the bottom of the bin. However, the higher grain mound height configuration in the present case has resulted in the non-flow paths at grain surface. A single duct cannot offer uniform gas flow distribution.¹⁹ Such non-uniformity in ozone flow patterns within the grain bed can lower the efficiency of the fumigation process. But multiple ducts may facilitate in providing uniform ozone flow distribution across the storage bin. It may be improves the effectiveness of ozone fumigation method and thereby assures the pests free stored grain.

Ozone is a surface active material. Fumigation of grains using ozone gas, initially its reacts with grain surface because of the organic nature (active tissue) of the stored material, high surface contact area of the grains²⁰ and diffuse the internal layer.²¹ Therefore, it is difficult to maintain steady state ozone concentrations in the interstices of the material to ensure uniform treatment to all the material in a storage container. The grains considered to be in the phase one state at the start of the fumigation progress,

ozone gas consumed soon after it entered the stainless steel bin, similar to wheat,²² green gram,¹¹ and rice.⁵ As the ozone fumigation progressed, the rate of consumption gradually decreased resulting in ozone diffusion into the grain mass.²² It indicates the ozone gas flowed freely through the grain column with little degradation once the active tissues responsible for ozone degradation became saturated.¹⁰ Previously published data shows that ozone concentration decreases faster, if microflora present on grain surface.²³ Allen *et al.*²⁴ stated that the higher is the mycological contamination, slower the ozone diffusion into the grain mass.

Fig. 3 shows that ozone gas highly diffuses in a lateral direction as compared to a vertical direction. This may be due to the resistance of grains to gas flow is higher in a vertical direction because of the kernel orientation and flow paths varies with direction.²⁵ In practical situations, when the bin is filled with paddy, the slender shaped paddy kernels lie with the major axes horizontal, thus possibly providing more resistance to ozone flow in the vertical direction. Also, the density of ozone, (2.14 kg m^{-3}) at standard temperature and pressure is 1.5 times greater than the density of air (1.43 kg m^{-3}). The density difference may result in mass flow in addition to molecular diffusion; hence the rate of movement of ozone in the direction of the force of gravity (downward) would be greater than upward movement of ozone, against the force of gravity. This phenomenon observed during the experiments with ozone, movement through air in grains and the effective flow of ozone was caused by the pressure gradient.

5 CONCLUSIONS

From the present study it was understood that ozone diffuse upto a bed thickness of 0.6 m. The results suggest that ozone gas fumigation in paddy grains will be more

effective in still air at minimum bed thickness. Ozone concentration simulated using the present model showed good agreement with experimental results. The present numerical simulation model is giving acceptable accuracy in prediction, simple and requires less computational time. This study can offer solutions in predicting the total time required for ozone gas fumigation and design of bin geometry, ozonation duct designs and direction of ozone flow in flat bottom bins.

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Source file information

Figure 1. Simulated ozone concentration in paddy grains

Figure 2. Effect of grain column distance on ozone concentration

Figure 3. Ozone concentration changes as a function of fumigation time

Figure 4. Cross validation of ozone concentration

Table 1. Relative errors and chi-square value between experimental and predicted values of ozone concentration

Time (h)	Ozone concentration (ppm)		Relative error (%)	Chi-square
	Experimental	Predicted		
1	8	14	42.8	2.57
2	38	38	0	0.00
3	69	91	24	5.32
4	104	143	27	10.64
5	169	196	13	3.72
6	242	248	2.6	0.15
7	306	301	1.6	0.08
8	362	353	2.3	0.23
9	418	406	2.8	0.35
10	482	458	5.06	1.26
11	506	511	1.04	0.05

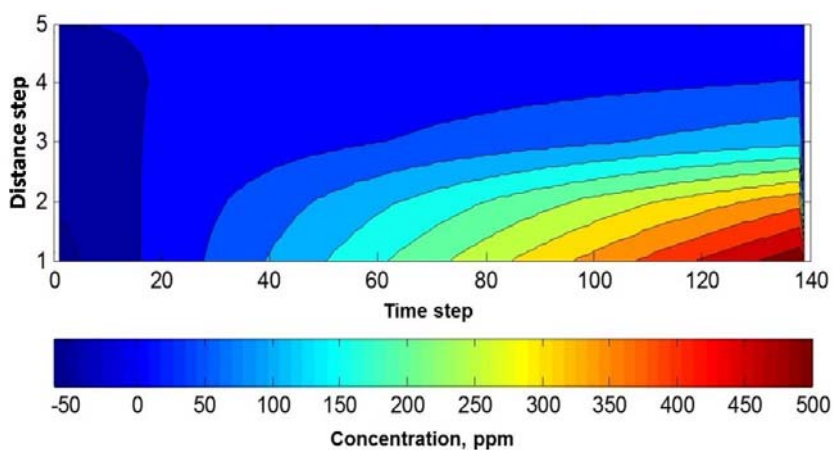


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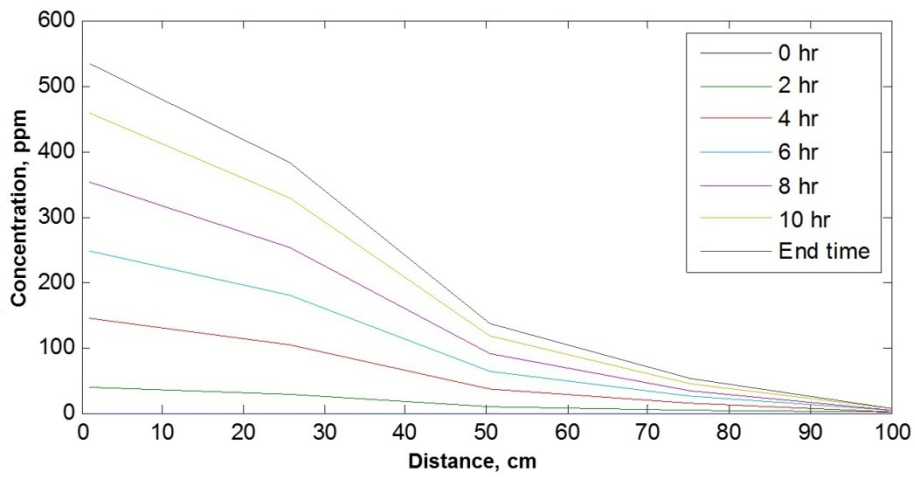


Figure 2. Effect of grain column distance on ozone concentration

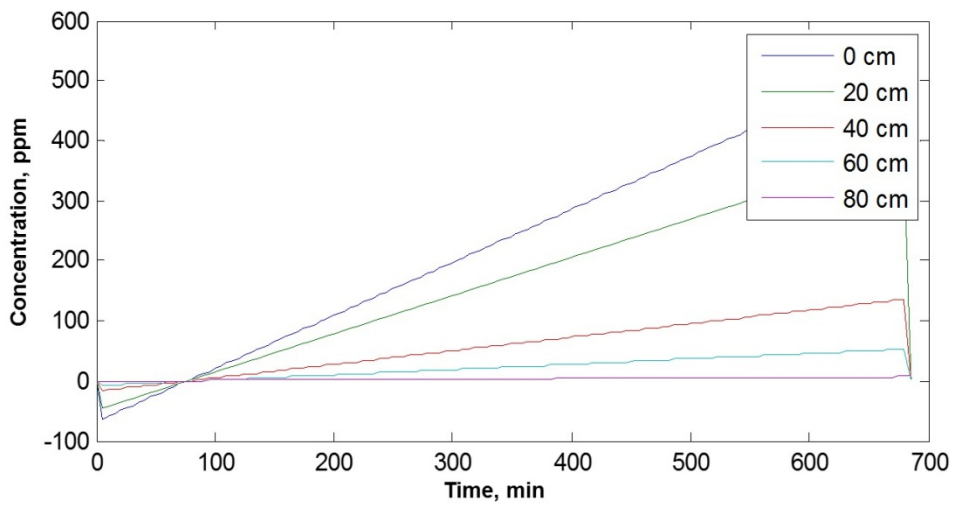


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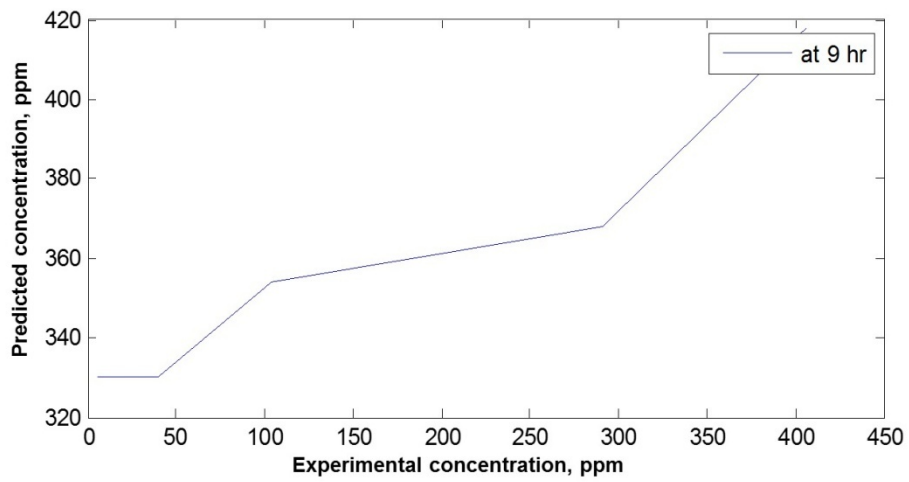


Figure 4. Cross validation of ozone concentration