Ethylene Removal From Fresh Produce Storage: Current Methods and Emerging Technologies

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Introduction

Ethylene is a colourless and odourless gas that acts as a plant ripening hormone. It is produced by plant tissues as a product of plant metabolism. Another source of ethylene is burning of hydrocarbons (Zagory, 1995). Among various effects of ethylene, it is responsible for rapid ripening and accelerated senescence in many climacteric and non-climacteric fresh produce (Ku et al., 1999; Saltveit, 1999). This effect of ethylene is often unwanted in postharvest storage and transport where the main focus is on extending shelf life of the produce. Thus, there are many existing and upcoming strategies of ethylene management and control. These strategies can be categorised into three groups; a) inhibition of endogenous ethylene production, b) inhibition of ethylene action and c) ethylene removal from the vicinity of the fresh produce. In the first category, products such as AVG (aminoethoxyvinylglycine) are used that affect the reaction pathway of ethylene biosynthesis and consequently the ethylene biosynthesis by the plant tissues is inhibited (Yuan and Carbaugh, 2007). Hypobaric storage (storage at low pressures <50 kPa), controlled and modified atmosphere storage (high CO_2 and low O_2) can also reduce ethylene production (Burg, 2004; Wills et al., 1982). In these techniques although ethylene production is suppressed, if the produce is subjected to exogenous ethylene from other sources, the detrimental effect of ethylene cannot be prevented. In the second category, products such as 1-MCP (1- methyl cyclopropane) are used for inhibiting the ethylene action i.e. the 1-MCP blocks the ethylene receptors on the produce surface and with no access to the receptors, ethylene is unable to affect the produce (Sisler and Serek, 1997). 1-MCP is being widely used and has resulted in favourable results in terms of delaying ripening and senescence and in some cases, reducing the occurrence of disorders (Blankenship and Dole, 2003). However, the effect of 1-MCP is variable on different products and can result in uneven ripening of the produce after prolonged storage (Mahajan et al., 2014). Moreover, it leaves a residue on the fruit surface and thus, cannot be used for organic products. The third strategy is to remove ethylene present in the vicinity of the fresh produce which may include different techniques such as venting, absorption and oxidation which have been shown in **Fig. 1**. Commercially available products based on different methods have been listed in **Table 1**. The detailed discussions of the various conventional and emerging methods under ethylene removal have been done in the following sections.

Current Methods

Ventilation

This is one of the easiest techniques to remove ethylene from surrounding areas of fruits and vegetables. The storage area can be replenished with atmospheric air at regular intervals. Air ventilation in long distance transport for maintaining low ethylene levels in banana transport was reported by Wills et al. (2014). This technique is generally used in cooler areas for reducing temperature

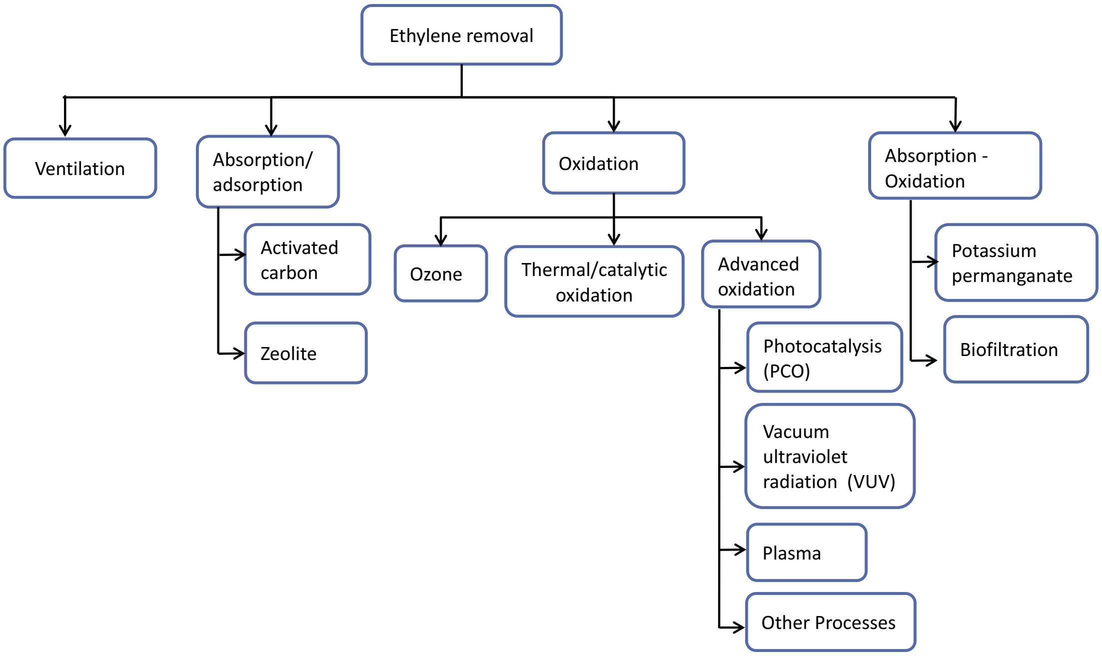

Figure 1 Different processes for ethylene removal.

Table 1 Some commercially available products for ethylene removal

Method	*Product name*	*Description*	*Company*
Adsorption, Adsorption-oxidation	It's Fresh!	Palladium impregnated zeolite sheets	It's Fresh! Ltd, UK
	PrimePro®	Sheets	DeltaTRAK, Inc., USA
	Extend-A-Life™ Produce Saver™	Filters, sachets	AgraCo Technologies International LLC, USA
	KEEPFRESH®	Sheets, bags	Teck Blue Systems, SL Keepfresh, Spain
	Peakfresh	LDPE film impregnated with mineral	Peakfresh, Australia
	Green Bags™	Clay	Evert-Fresh Corp., USA
	EC-3+	Zeolite impregnated with potassium permanganate in sachets, filters, filtration system	Ethylene Control Inc., USA
	Bi-On®, ETHYL STOPPER	Porous clay and potassium permanganate	Bioconservación S.A, Spain
	Ryan®	Natural clays and potassium permanganate ($KMnO_4$) in sachets and filters	Sensitec Co., USA
	ETI 25, 50	ABSOTIL/purafil- Alumina pellets	Fruit Control Equipments srl, Italy
	TMC Ethylene absorbers, TMC-5 Converter	Sodium Permanganate pellets	The Magic Cube Company Ltd., Hong Kong
Oxidation with ozone	Bio-Turbo	Ozone	Miatech, Inc., USA
High temperature catalytic oxidation	SWINGTHERM-BS	Air filtration equipment	Fruit Control Equipments srl, Italy
	Catalytic converters	Air filtration equipment	The Magic Cube Company Ltd, Hong Kong
	na	Air filtration equipment	Besseling Group BV, The Netherlands
	na	Platinum aluminium granules used in air filtration equipment	Van Amerongen, The Netherlands
Photocatalytic oxidation	ECOscrub	Titanium dioxide + UV light based air firtation equipment	Absorger, France
	AiroCide®	Air filtration	KES Science & Technology, Inc.
	FRESH+™	Catalyst + UV light	Fresh Plus International, USA

na: not available.
Adapted from Pathak et al. (2017a).

with the added benefit of ethylene removal. It is important to ensure that the replacement air must be ethylene free. As stated before ethylene is also produced from burning of hydrocarbons, thus, the exhaust from vehicles etc., may add to ethylene contamination in the air resulting in higher ethylene concentration in the air than normal (0.001-0.005 ppm) (Warton et al., 2000). Thus, conditioning of the outside replacement air may be required in non-optimal external atmosphere conditions. Excessive ventilation can also result in higher weight loss (Wills, 2015; Wills et al., 2012). Moreover, ventilation is not a suitable option in closed storages with controlled or modified atmospheres or refrigeration.

Adsorption/Absorption

Highly porous substances such as activated carbon and zeolite (aluminosilicate minerals) are effective in adsorbing ethylene. These materials may be used in form of sheets, filters or pads for ethylene absorption. Palladium incorporated into activated carbon was reportedly found to be more effective in fruit storage with tomato as the test product compared to activated carbon alone (Bailén et al., 2006). A heat cartridge connected to activated carbon with 1% palladium was developed by Martínez-Romero et al. (2009) to prevent saturation of the absorber. Palladium impregnated, zeolite based sheets yielded good results in terms of effective ethylene removal in green bananas and avocado(Smith et al., 2009; Terry et al., 2007). These sheets have been commercialised and are available as packaging films, sheets, pads etc. (It's Fresh! Ltd., UK). Other products such as Peakfresh (Peakfresh, Australia) made of LDPE film impregnated with naturally occurring minerals and Green Bags™ (Evert-Fresh Corporation, USA) based on clay are available. However, the absorbers may get saturated after a period of use or when high ethylene concentrations are present and thus, would require replacement or regeneration.

Oxidation

Using Ozone

Ozone is a strong oxidiser and its application in ethylene removal is well known (Smilanick, 2003; Suslow, 2004). It can oxidise impurities as well as disinfect microbes which makes it suitable for use in fruit and vegetables. An ozone concentration of 0.4 ppm effectively removed ethylene from apple and pear storage without affecting fruit quality. However, high ozone concentrations can be detrimental for stored fresh produce (Smilanick, 2003). Controlling ozone concentration especially with respect to the exposure to humans is a major challenge. The recommended safe limit of ethylene exposure in the USA is 0.1 ppm for an 8-h duration (US-OSHA (United States Occupational Safety and Health Administration)), and is even lower in other countries (e.g. New Zealand). Higher ozone concentrations are hazardous and can result in headaches, lung damage, chronic respiratory disorders and can even be fatal.

Filter systems based on ethylene removal using ozone are marketed by Bio-Turbo (Miatec, USA). Ethylene-laden air is drawn in from the fruit storage which is made to react with ozone inside the specialised chamber. After oxidation, clean CO_2 and H_2O is returned back to the storage. Use of a catalyser to oxide residual ozone ensures no ozone leakage into the environment as provided in the data by the company.

Catalytic Oxidation

At high temperatures, ethylene can be oxidised in the presence of catalysts into carbon dioxide and water. El Blidi et al. (1993) developed an industrial type prototype for oxidation of ethylene at temperatures 100–120 °C in presence of catalysts of manganese and copper. This prototype was tested in storage of golden delicious apples and was able to retain the quality close to the initial properties even at the end of storage. Commercial equipment based on these methods are available such as Swingtherm (Fruit Control Equipments srl, Italy). Companies such as Van Amerongen (The Netherlands), Absorger (France), Bessseling Group (The Netherlands) also manufacture similar systems for ethylene removal in fresh produce storage.

Photocatalysis or Photocatalytic Oxidation (PCO)

Photocatalysis or photocatalytic oxidation (PCO) is a relatively new technique for application in the postharvest sector for ethylene removal. It involves irradiation of a semiconductor with a suitable wavelength radiation. Semiconductors have a small energy gap between their valence band and conduction band. Radiation with a suitable wavelength can excite electrons from the valence band to the conduction band, simultaneously generating holes in the valence band. These electron–hole pairs are highly reactive and combine with surface adsorbed water and oxygen molecules to produce reactive oxygen species such as hydroxyl radicals, superoxide ions etc. These species eventually oxidise ethylene into carbon dioxide and water. Titanium dioxide is the most commonly used catalyst owing to its easy availability, stability and low cost. Other photocatalysts are also available (Si, Au oxides etc). Extensive research into modifying the catalyst for better efficiency and light radiation utilisation is being carried out (Chen and Mao, 2007; Hussain et al., 2010). Several modifications for the activation of the catalysts in the visible light region are also being carried out for better application (Licciulli et al., 2017; Lin et al., 2014). Commercial equipment based on photocatalysis such as ECOscrub (Absorger, France) and AiroCide® (KES Science & Technology, Inc) are available. However, scientific reports on the efficiency and effectiveness of such equipment in maintaining fresh produce quality are few. Application of a photocatalytic reactor inside a refrigerator and in cold storage was reported by Kartheuser and Boonaert (2007) on the other hand, quality of papayas in storage with a photocatalytic reactor was reported by Lourenço et al. (2017).

Oxidation Using Shortwave UV Light (Vacuum Ultraviolet Light VUV)

Shortwave ultraviolet light (often referred to as vacuum ultraviolet radiation, VUV ($\lambda < 200$ nm)) has high energy protons (~6 eV) that can dissociate water and oxygen molecules present in the atmosphere into reactive oxygen species. Commercially such lamps are available that produce major radiation at 254 nm (UV−-C) and minor radiation at 185 nm (VUV). Use of such lamps in banana storage for ethylene removal was demonstrated in the early 1970s (Scott et al., 1971). The use of VUV radiation is extensively studied for purification of air and has demonstrated good results in ethylene removal as well as in removal of microorganisms which could be a major advantage for storage of fruits and vegetables storage. However, production of ozone as a by-product limits this process. Pathak et al. (2017b) studied various factors affecting this process and also found it effective in reducing ethylene produced by apples. However, research into the effect of this process on all fruit quality aspects is needed.

Adsorption-Oxidation

Using Potassium Permanganate

Potassium permanganate ($KMnO_4$) is one of the widely used products for ethylene management. In a complete reaction with $KMnO_4$, ethylene is oxidised to carbon dioxide and water in a complete reaction leaving behind residue MnO_2 and KOH. Usually it is used with an additional adsorber/absorber material that absorbs the residue. These products are generally available as sachets (Purafil, USA) which can be placed close to the produce. In the first step, ethylene is absorbed by the absorbers and then oxidation takes place. In an incomplete oxidation, by-products such as potassium acetate may also be formed. Thus, by-products and end residue requires disposal. Moreover, in a study it was reported that $KMnO_4$ had reduced efficiency at high humidity conditions (Wills and Warton, 2004). Thus, frequent replacement may be required in long storage durations or with high ethylene concentrations. Bi-On®, ETHYL STOPPER by Bioconservación S.A, Spain are filtration equipment and sachets, respectively based on porous clay and potassium permanganate. Recently, some products based on $NaMnO_4$ have become available. According to the company (The Magic Cube Company Ltd.), it exhibits better efficiency than $KMnO_4$ in ethylene removal.

Bio-filtration

Another technique for ethylene removal is bio-filtration which employs a microbial load for ethylene removal. Biofiltration can be carried out using bioscrubbers, biofilters and biotrickling filters (Delhoménie and Heitz, 2005). In bioscrubbers microbes are suspended in an aqueous medium whereas in biofilters and biotrickling filters, the microbial load is immobilised on a filter material. Biotrickling filters differ from biofilters in terms of the movement of nutrient solution. As the name suggests, biotrickling filters have a continuously trickling or circulating nutrient solution through the filter bed whereas in bio filters there may be occasional replenishment with a nutrient solution. The filtering material may consist of one or multiple layers of porous substances such as peat, compost or soil. Some examples of different filter materials reported in literature are peat soil (Elsgaard, 2000), granular activated carbon (inoculated with *Bacillus* or *Pseudomonas* strains) (Kim, 2003), active sludge, peat wood chips, humus with organic soil (Moghadam et al., 2015), perlite and glass beads (Lee et al., 2010). Ethylene is first absorbed by the porous media and then oxidation takes place at the biological film formed by the microbes. The end product of ethylene oxidation is water, CO_2 and microbial cellular material. Kim (2003), by application of an activated granulated carbon biofilter, was able to achieve 100% ethylene removal in 14 min residence time for an inlet ethylene concentration of 331 mg m^{-3}. In another study (Moghadam et al., 2015), biofilter substrate was tested in a banana storage experiment by placing the substrate in cotton bags inside a fruit box containing bananas. At the end of 7 days storage at 30 °C, bananas stored with substrate of biofilter had ripened to a lesser degree than the untreated control.

Biofiltration is advantageous in terms of being of low energy intensive, moderate cost and has a low maintenance cost. However, it is a slow process. Biofilters may be associated with problems of pressure drop and require a large surface area. Moreover, for application in horticulture storage often low temperatures <15 °C are used, thus, it is important to ensure that the microbial strain being used has sufficient activity even at these temperatures.

Emerging Technologies

Advanced Oxidising Species Based Techniques

Several upcoming processes based on active oxygen species (similar to photocatalysis) have been shown to be effective in oxidation of ethylene and are being studied for potential application in ethylene removal in horticultural storage. In such processes advanced oxidation species such as hydroxyl radicals, superoxide ions are produced that convert ethylene into carbon dioxide and water in a complete reaction. Such processes may include various hybrid techniques, such as combination of PCO with ozone, PCO with VUV, combining VUV with ozone-assisted catalytic oxidation, and non –thermal plasma techniques. A hybrid process of VUV-PCO was reported to have higher ethylene removal efficiency compared to VUV, and PCO processes alone (Chang et al., 2013). Since ozone is produced in a VUV processes, coupling it with a secondary catalyst (Mn, Co, Fe, Ni supported on an aluminosilicate zeolite ZSM-5 (Zeolite Socony Mobil–5) or activated carbon with TiO_2) has been demonstrated for better ozone removal and at the same time, better volatile organic compound oxidation can also be achieved (Huang et al., 2016). Ethylene degradation using non-thermal plasma discharge techniques such as dielectric barrier, and corona discharge for plasma generation have been reported (Ma and Lan, 2015). In the plasma reaction, highly reactive radicals such as charged hydrogen and charged

Table 2 Advantages and limitations associated with various ethylene removal processes

Process	*Principles/characteristics*	*Advantages*	*Limitations*
Ventilation	Ventilation of storage units with fresh air from outside	● Easy ● Low cost	● Requires conditioning of fresh air ● Excessive ventilation results in weight loss of product
Absorption/adsorption	Use of porous materials such as activated carbon, zeolite	● Can be used with small packages	● Need replacement when saturated ● Regeneration is difficult
Ozone based oxidation	Use of ozone gas to oxidise ethylene	● Suitable for big storage spaces ● Disinfectant properties	● High concentration of ozone-injurious to plant tissues ● Hazardous to human health (>1 ppm) ● Handling difficulties
Thermal catalytic oxidation	Use of high temperature in presence of catalyst	● Continuous in operation	● Energy intensive ● Need elevated temperatures
Photocatalytic oxidation (PCO)	Use of ultraviolet light in presence of a catalyst for generation of electron hole pair and reactive oxidation species	● Continuous in operation ● Does not require elevated temperature or pressure	● Not efficient at low ethylene concentrations ● Catalyst deactivation may occur with time
Vacuum ultraviolet radiation photolysis (VUV)	Use of high energetic protons to generate reactive oxidation species	● Continuous in operation ● Does not require elevated temperature or pressure	● Production of ozone ● Needs ozone filtration
Plasma	Electric field and accelerated electrons required for production of reactive species such as charged hydrogen, charged oxygen	● Continuous in operation	● Production of ozone ● Needs ozone filtration
Potassium permanganate	$KMnO_4$ oxidizes ethylene into carbon dioxide and water	● Simple process ● Can be used within small packages	● Cannot be regenerated ● Needs frequent replacement ● Toxic residue disposal problems
Biofiltration	Oxidation by biofilm formed by microorganisms	● Easy ● Low cost ● Low maintenance	● Slow process ● Requires large surface area ● Requires optimal environmental conditions for microbial growth and survival

oxygen species are generated. Graham et al. (1998) developed a plasma reactor for potential use in the fruit and vegetable industry which was able to remove 78.2% of ethylene (initial 1 ppm concentration) in a single pass. Similarly, a reactor for application in cold humid fresh produce storage with a corona discharge plasma coupled with TiO_2 and activated carbon film was developed by Ye et al. (2013). However, these processes are in the fundamental stages of study. Although these techniques show good results in terms of ethylene degradation, complexities of these processes and ozone production are a challenge when it comes to actual application. Research on the impact of these processes on the quality of products, ease of application, and cost effectiveness is currently lacking.

Conclusion

Various ethylene removal techniques have their own advantages and limitations as summarised in Table 2. Currently, a variety of commercial products based on these techniques in the form of sheets, pads, and filtering equipment are available. Certain products give better results in terms of ethylene removal when placed inside fresh produce packages while others can be more suitable for large storage spaces. Thus, it is important to select the appropriate ethylene removal method depending on various factors such as ethylene sensitivity of the fruits, storage space, quantity of fruit etc. Comparable scientific data of the impact of different ethylene removal processes on fruit quality is missing which makes the comparison among these techniques difficult. Thus, for a comparative assessment of these products, scientific information regarding the efficiency of different commercially available products in actual storage conditions along with the cost analysis will be helpful. Overall, since ethylene plays an important role in ripening of many horticultural products, the advancement of ethylene removal technologies is much needed. Emerging technologies such as advanced oxidation-based processes still need further research in terms of practical applicability.

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