Honours Thesis

Installation of a Wood Combustion Catalyst to Reduce Winter

Air Pollution in Tasmania

By

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This thesis is submitted as a requirement for the degree of the

Bachelor of Health Science with Honours

January 2015

School of Health Sciences

University of Tasmania, Launceston

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Declaration

I declare that this thesis is my own original work submitted for the Bachelor of Health Science with Honours and not for another degree or diploma at any other institution. I certify to the best of my knowledge that all material of this thesis is my own original work except where there is clear acknowledgement or reference made to the work of others.

Olivia Johnston

Signature: *Johnstan* 30th January, 2015

Acknowledgements

Special thanks to my supervisors, Dr Fay Johnston and Dr John Todd. I very much appreciate your guidance, encouragement and support throughout the past year. It has been a pleasure working with you.

Thank you to Dr Grant Williamson for solving all of my computer problems and guiding me through the statistical analyses.

Thank you John Innis for your commitment to this study, especially for providing all the experimental data, taking street photos and answering all of my questions that I put to you.

Thank you to the Northern Midlands Council, The Examiner and members of the Perth Lions Club for your interest and support during this past year, helping to achieve successful promotion and implementation of the intervention.

Thank you to Smartburn Australia for donating catalysts and particularly Glenn and Lisa Fretwell, for your generous commitment to planning and implementing the intervention, supporting the volunteers and assisting with food and travel costs throughout our weekend in Perth.

Finally, I thank my family, especially my parents and my sister for helping distribute canisters in June and always encouraging me to do my best.

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List of Abbreviations

ABS Australian Bureau of Statistics
BAL bronchoalveolar lavage fluid
BLANkET Baseline Air Network of the EPA Tasmania
CI confidence interval
CO carbon monoxide
CO ₂ carbon dioxide
COPD chronic obstructive pulmonary disease
DNA deoxyribonucleic acid
EBC exhaled breath condensate
EC elemental carbon
EPA Environment Protection Agency (United States)
Environment Protection Authority (Australia)
GAM general additive model
GLM general linear model
GSH glutathione
HEPA high-efficiency particulate air (filter)
hPa hectopascal
HRV heart rate variability
HRV heart rate variability IHD ischaemic heart disease

IL-8 interleukin-8

µg microgram

µm micrometre

NEPM National Environment Protection Measure

NO nitric oxide

NO_x nitric oxides

OC organic carbon

OH hydroxyl

PAH polycyclic aromatic hydrocarbons

PM particulate matter

 PM_{10} coarse particulate matter up to $10\mu m$ in aerodynamic diameter

PM_{2.5} fine particulate matter up to 2.5µm in aerodynamic diameter

PM_{0.1} ultrafine particulate matter up to 0.1µm in aerodynamic diameter

POA primary organic aerosol

QGIS Quantum Geographic Information System

ROS reactive oxygen species

SEM standard error of mean

SOA secondary organic aerosol

SO₂ sulphur dioxide

TNF-alpha tumour necrosis factor-alpha

TRPA-1 transient receptor potential-ankyrin 1

VOC volatile organic compounds

Thesis Summary

Domestic wood combustion is the predominant source of winter air pollution in Tasmania. Inhalation of fine particulate matter of wood smoke ($PM_{2.5}$) is detrimental to respiratory and cardiovascular health. Fine particles have the capacity to lodge deep into lung tissue and translocate into the circulation, subsequently inducing systemic inflammation and oxidative stress. Catalysed combustion is designed to reduce emission of $PM_{2.5}$ generated from domestic wood heaters by enhancing the efficiency of combustion.

The present study was the first community trial to quantify the influence of the catalytic device, Smartburn[®], upon ambient air quality. The trial targeted Perth, a rural, northern Tasmanian town where approximately half of the residences use firewood for domestic heating. In June, 2014, almost 80% of wood heater residences accepted a catalytic device for installation in their firebox. Ambient $PM_{2.5}$ concentration was measured from fixed air monitoring stations at Perth and the control towns, Longford, Hadspen and Westbury. In order to identify a significant reduction in Perth pollution, the mean pre-intervention and post-intervention concentrations of Perth were compared. The mean post-intervention concentration of Perth was then compared with those of the control towns.

The main finding of this study was the absence of a significant association between community-wide distribution of the catalytic device and ambient air quality. Perth ambient pollution was not significantly reduced after the trial. Catalysed combustion remains difficult to achieve in a residential setting without sufficient control over factors associated with domestic operation of the wood heater and maintenance of the catalyst.

The first section of this thesis is a literature review of the health effects associated with fine PM exposure, the chemical characterisation of wood smoke, the technological aspects of wood combustion and previous interventions designed to reduce residential wood smoke. The

second section of the thesis is a research paper which investigates the potential for community-wide firebox installation of a catalytic device to reduce ambient $PM_{2.5}$ concentration in Perth, Tasmania. The manuscript is formatted according to the *Instructions for Authors* in the journal of *Atmospheric Environment* (Appendix A).

Chapter One LITERATURE REVIEW

1.1 Introduction

Ambient air pollution generated from domestic wood combustion is of global health significance. Wood combustion is a popular source of residential heating due to the affordability, accessibility and the renewable nature of wood, compared to fossil fuels (Sandradewi et al., 2008). However, it is also a prominent source of air pollution in residential areas. According to Bølling et al. (2012), atmospheric aerosols of wood combustion are heterogeneous mixtures of inorganic and organic material. Common constituents are chlorinated dioxins, carbon monoxide (CO), methane, volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs), nitric oxides (NO_x), sulphur dioxide (SO₂) and particulate matter (PM) (Riddervold et al., 2012).

The public health consequences of wood smoke exposure are adverse. Some PAHs, ROS and VOCs are potentially carcinogenic and PM is predominantly associated with respiratory and cardiovascular morbidity and mortality (Riddervold et al., 2012; Gold and Mittleman, 2013). Particle-induced bronchoconstriction, mucosal secretion and airway obstruction collectively exacerbate asthma and increase the risk of developing chronic obstructive pulmonary disease (COPD) (Fullerton et al., 2011). Endothelial activation induces atherosclerosis, thrombosis, hypoxemia and infarction (Bønløkke et al., 2014).

Variations in particle size, structure and surface area influence PM potency and capacity to transport metals and organic compounds via adsorption (Wang et al., 2010; Bølling et al., 2012). Coarse particles are classified as those with aerodynamic diameters between 2.5 micrometre (μ m) (PM_{2.5}) and 10 μ m (PM₁₀). Particles with aerodynamic diameters less than 2.5 μ m (PM_{2.5}) are fine and those less than 0.1 μ m in diameter (PM_{0.1}) are ultrafine (Donaldson et al., 2001). The increased surface area of fine and ultrafine particulates enhances their capacity to absorb chemical components, penetrate deep into lung tissue and stimulate inflammation (Strak et al., 2013). Activation of alveolar macrophages and pulmonary

epithelial cells releases pro-inflammatory mediators and ROS which rupture the respiratory membrane and translocate into the circulation (Danielsen et al., 2008). Systemic inflammation and oxidative stress overwhelm antioxidant defences, destroying host cells via lipid peroxidation and ultimately inducing apoptosis and carcinogenesis (Danielsen et al., 2011). The chemical composition of ambient pollution is highly variable according to the source of emission (Bølling et al., 2012; Su et al., 2013). Wood smoke is particularly variable in toxicity, according to fuel characteristics, conditions of combustion and the type of combustion appliance used (Heringa et al., 2011; Miljevic et al., 2010). The atmospheric fate of point source emissions is determined by land topography and meteorological conditions (Su et al., 2013).

This review will discuss the human health consequences associated with exposure to fine PM. Meteorological and topographical influences related to community-wide air pollution will facilitate understanding of the complexity associated with human dose-responses. Residential wood combustion will be the primary focus, with emphasis on the chemical characterisation of wood smoke. However, toxicological comparisons will be made with additional PM sources, particularly diesel exhaust and tobacco smoke. Potential strategies to reduce emissions and address the variable nature of residential wood combustion will be reported.

1.2 Respiratory implications of wood smoke exposure

Chronic wood smoke exposure has been associated with respiratory diseases, especially asthma, pneumonia, COPD and lung cancer (McGowan et al., 2002; Sanhueza et al., 2009; Fullerton et al., 2011). Particulates may also exacerbate symptoms of viral respiratory infection (McGowan et al., 2002). Victims of acute exposure experience symptoms of cough, nose and throat irritations and elevated frequency of respiratory tract infection (Sehlstedt et al., 2010; Fullerton et al., 2011). Comparisons between wood and charcoal cooking stoves in Malawi homes detected an elevated prevalence of respiratory tract infection among residents

exposed to wood smoke (Fullerton et al., 2011). Similarly, a population-based study of Nepal detected significant airway obstruction and impaired lung capacity among individuals exposed to biomass fuels, as opposed to non-biomass fuels (Kurmi et al., 2013). Shapiro et al. (2013) attribute the potency of wood smoke particulates to stimulation of transient receptor potential-ankyrin 1 (TRPA-1), a molecular sensor for oxidants in the respiratory tract expressed by bronchial, alveolar and endothelial cells. Particulate stimulation of the receptor induces mucosal irritation, bronchoconstriction and inflammation (Shapiro et al., 2013). According to Camp et al. (2014), COPD encompasses the phenotypes, bronchiolitis and emphysema. Biomass smoke induces bronchiolitis which is the airway-predominant phenotype associated with increased bronchial wall thickness and air trapping, or abnormal lung retention of air during expiration (Camp et al., 2014). Wood combustion emits significant concentrations of CO, which is a potent indicator of cardiovascular mortality (Fullerton et al., 2011; Steinvil et al., 2013). The product of incomplete combustion influences blood oxygenation, thereby reducing oxygen uptake and functional capacity. Exercise stress tests among healthy members of a population exposed to ambient CO revealed significant reductions in functional capacity as a result of chronic exposure (Steinvil et al., 2013).

Respiratory ramifications of acute exposure remain less clear. Riddervold et al. (2012) and Sehlstedt et al. (2010) observed no acute wood smoke exposure-related alterations in lung function and inflammatory cytokine concentration of plasma and bronchoalveolar lavage fluid (BAL) among exposed subjects. However, Riddervold et al. (2012) detected inflammatory biomarkers in exhaled breath condensate (EBC) and Sehlstedt et al. (2010) detected an increase in BAL antioxidant glutathione (GSH) concentration, representing an acute response preventing oxidative damage to the bronchial epithelium. Differential research conclusions may be attributed to the duration of exposure and nature of participant selection. Camp et al. (2014), Fullerton et al. (2011) and Kurmi et al. (2013) selected subjects based upon chronic histories of exposure to specific PM sources, while Riddervold et al. (2012) and Sehlstedt et al. (2010) measured respiratory responses to acute exposure among healthy volunteers with normal lung function and without bronchial hypersensitivity.

1.3 Cardiovascular and carcinogenic implications of wood smoke exposure

Inhalation of wood smoke PM is associated with systemic inflammation, oxidative stress and carcinogenesis. According to Danielsen et al. (2011), Bølling et al. (2012) and Strak et al. (2013), fine particulates induce systemic inflammation and oxidative stress via two distinct pathways: activation of pulmonary epithelial cells and alveolar macrophages to generate proinflammatory mediators and ROS, subsequently rupturing the respiratory, alveolar-capillary membrane and entering the circulation; or via direct translocation of particulates and adsorbed constituents through the alveolar-capillary membrane (Strak et al., 2013; Danielsen et al., 2011). Oxidative damage to DNA induces strand breaks and mutation, thereby initiating carcinogenesis (Danielsen et al., 2011). For instance, oxidation of guanine forms 8-oxoGua which instigates irreversible transversion mutations (Danielsen et al., 2008).

Tumour necrosis factor (TNF)-alpha, interleukin-6 (IL-6) and interleukin-8 (IL-8) are early biomarkers of inflammation detected in the studies of Danielsen et al. (2011), Stockfelt et al. (2013) and Forchhammer et al. (2012) as chemo-attractants of additional inflammatory cells and stimulants of the liver and phagocytes to produce acute phase proteins and ROS, respectively. Systemic inflammation and oxidative stress induce endothelial dysfunction and subsequent development of atherosclerosis, hypertension, hypoxemia and myocardial infarction (Stockfelt et al., 2013). Acute wood smoke exposure has been shown to cause retinal arterial narrowing and oxidative damage to DNA (Louwies et al., 2013; Danielsen et al., 2008). According to Unosson et al. (2013), wood smoke-induced imbalances in autonomic innervation and endothelial function influence alterations in central arterial stiffness and heart rate variability (HRV) during exercise. Endothelial dysfunction is induced by depletion of the

vasodilator, nitric oxide (NO) (Gill et al., 2011; Allen et al., 2011a). The enzyme responsible for NO synthesis is destroyed by inflammatory mediators in human aortic endothelial cells (Allen et al., 2011a). Unosson et al. (2013) observed three-hour exposure to filtered air during exercise, followed by three-hour exposure to wood smoke during exercise, to increase heart rate and central arterial stiffness, while reducing HRV and exerting no influence upon blood pressure (Unosson et al., 2013). Arterial stiffness and reduced HRV are prominent risk factors for cardiovascular disease. Reduced variation to the interval between consecutive heartbeats (HRV) stems from particulate inhibition of parasympathetic innervation and particulate stimulation of sympathetic innervation (Unosson et al., 2013; Gill et al., 2011). Such autonomic imbalance contributes to arterial stiffness and elevated heart rate (Unosson et al., 2013). These findings are consistent with those of a prospective, population-based study in Peru which found that residents cooking with biomass fuels recorded higher prevalence of carotid thickness, hypertension and carotid artery atherosclerotic plaques, compared to those cooking with cleaner fuels of kerosene and electricity (Painschab et al., 2013).

On the contrary, Stockfelt et al. (2013) and Bønløkke et al. (2014) found acute exposure to wood smoke to not cause systemic inflammation or increase the risk of thrombosis and atherosclerosis via endothelial dysfunction. Reductions in the coagulant, fibrinogen and platelets indicated an inactivation of the endothelium to induce haemostasis. Similarly, Forchhammer et al. (2012) found no apparent effect of four-hour wood smoke exposure upon microvascular function, oxidatively damaged DNA and biomarkers of inflammation and oxidative stress. Discrepancies in results may be attributed to the nature of exposure. Exercise enhances ventilation, as opposed to exposure at rest (Unosson et al., 2013). Furthermore, Jensen et al. (2014) suggests that acute durations of exposure are inadequate for establishing the events of systemic inflammation, oxidative stress and DNA damage. Dietary consumption of antioxidants may resist particulate-induced oxidative stress (Jensen et al., 2014). According

to Riddervold et al. (2012), humans undergo an adaptive period which further inhibits establishment of responses after only a few hours of exposure. Most studies compensated for this by incorporating an acclimatisation period before the PM concentration was sustained at the desired threshold.

According to Gold and Mittleman (2013), chronic PM exposure is more significant to cardiovascular health than acute exposure. Fine particulate exposure for durations of hours to weeks may provoke events of ischemic heart disease (IHD): myocardial infarction and heart failure. However, PM exposure over a number of years enhances the risk of cardiovascular mortality and reduces life expectancy by several years (Gold and Mittleman, 2013). The respiratory and cardiovascular ramifications of wood smoke exposure evaluated previously, predominantly pertain to individuals. Epidemiological studies articulate how such individual responses of clinical significance may be transformed into population-based statistics, representing trends of morbidity and mortality. McGowan et al. (2002) and Sanhueza et al. (2009) found daily ambient PM₁₀ concentrations to be associated with mortality and hospital admissions for respiratory and cardiovascular disease. Such observations were made within residential areas where domestic wood combustion is the main source of particulate air pollution in winter. In Christchurch, New Zealand, McGowan et al. (2002) found a 14.8µg/m³ increase in PM₁₀ to be associated with a 1.3% and 3.4% rise in hospital admissions for cardiovascular and respiratory conditions, respectively. Respiratory admissions were particularly elevated among children, and hospital admissions for pneumonia and COPD were particularly high among the elderly. Furthermore, in Temuco, Chile, Sanhueza et al. (2009) identified a $100 \mu g/m^3$ increase in PM_{10} to be associated with an 11.1% and 5.2% rise in respiratory and cardiovascular mortality, respectively. Hospital admissions for respiratory and cardiovascular conditions rose by 6.9% and 4.9%, respectively. Such an increase in PM_{10} yields even greater risk of mortality among adults aged over 64 years with a 14% rise in both causes compared to respective 9.5% and -13.6% admission rises among their younger counterparts (Sanhueza et al., 2009). Such trends between ambient particulate concentration and population mortality support the potential for interventions targeting wood smoke emissions to benefit public health.

1.4 Meteorological influences upon ambient air pollution

Respiratory and cardiovascular health risks of wood smoke are elevated during the winter due to local topography and seasonal variation of weather, coupled with the density and location of wood combustion sources (Su et al., 2013; Allen et al., 2011b). Thermal inversions, characterised by high relative humidity and low ambient temperature, wind speed and photochemical action, trap warm, polluted air at low altitude (Bergauff et al., 2008; McGowan et al., 2002). Air stagnation beneath the inversion layer accumulates pollutants at ground-level and inhibits dispersion from point source, thereby increasing the duration and intensity of residential exposure to ambient PM (Aymoz et al., 2007; Favez et al., 2010). Under conditions of high atmospheric pressure, coupled with an absence of vertical mixing and aerosol dilution, inversion layers may persist for several days and contribute to smog formation (Alfarra et al., 2007; Lanz et al., 2007). Primary organic aerosols (POAs) are emitted directly from point source in particulate or gaseous phase and undergo a series of atmospheric reactions to form secondary organic aerosols (SOAs) (Hennigan et al., 2011). Atmospheric ageing involves fragmentation and segregation of POAs via condensation, evaporation and oxidation (Heringa et al., 2011). Subsequent alterations in organic chemistry of primary aerosols imply that pure, immediately-generated wood smoke particulates do not accurately represent the toxicity of ambient wood smoke (Totlandsdal et al., 2014). Continuous atmospheric exposure and reaction with oxidants and hydroxyl (OH) radicals reduce their atmospheric stability (Hennigan et al., 2011). Nolte et al. (2001) detected differential PAH ratios between ambient and point source emissions as a consequence of atmospheric ageing. Higher concentrations of PAH have been detected in ambient air (Ding et al., 2012). Contrary to these findings, Danielsen et al. (2011) found PAH concentrations to be higher in pure wood smoke emissions compared to ambient air within a wood smoke-affected area. Pure particulates tended to produce more free radicals, DNA damage and gene expression *in vitro* in response to inflammation and oxidative stress, compared to particulates of ambient wood smoke-polluted air (Danielsen et al., 2011).

Ambient temperature is the most influential factor of PM concentration (Alfarra et al., 2007; Aymoz et al., 2007). Diurnal PM concentrations peak at midnight and fall to a minimum around midday due to alterations in meteorological conditions from low ambient temperature and absent photochemical action to increased temperature, optimal photochemical action and elevated wind speed (Jeong et al., 2008). In addition, low ambient temperature favours condensation of VOCs and positively corresponds with domestic wood heater usage (Lanz et al., 2007). Residential areas with high wood heater density are most vulnerable to the ramifications of wood smoke. Low elevation and hydrological catchment areas are equally susceptible to accumulating elevated pollutant concentrations (Su et al., 2013). During cold, calm nights, surface wind is influenced by drainage flow of wood smoke from up-slope sources (Su et al., 2013). During the day, vertical mixing exports pollutants from point source via elevated wind speed and photochemical action. During the night, pollutants remain close to the ground, with some dilution occurring from surface wind (Alfarra et al., 2007). According to Sanhueza et al. (2009) and Gold and Mittleman (2013), ambient temperature is associated with PM concentration and rates of mortality, hospital admission and incidence of respiratory and cardiovascular conditions, with peak rates likely to occur in winter when ambient temperature is minimal and PM concentration is elevated.

1.5 Chemical characterisation of PM

The chemical composition of PM depends upon the source. According to Camp et al. (2014), particulates of tobacco smoke are chemically similar to those of wood smoke. It is the differential nature of inhalation between both sources that influences the health outcomes induced. Inhalation of wood smoke is sustained at chronic, low levels while inhalation of tobacco smoke is acute and encompasses two phases: indirect inhalation through the mouth before a slight pause, and complete inhalation of particulates penetrating deep into the lung (Sood et al., 2010; Camp et al., 2014). Subsequent deposition of tobacco smoke particulates at the lung parenchyma impairs gaseous exchange and ultimately induces respiratory failure (Camp et al., 2014). Sood et al. (2010) describe a potential synergistic relationship between tobacco and wood smoke, whereby wood smoke instigates lung cancer among tobacco smokers via abnormal modification of lung cancer-related genes. In addition, wood smoke facilitates the development of COPD among tobacco smokers via impaired lung function, airflow obstruction and chronic bronchitis (Sood et al., 2010). Particulate deposition and inflammatory potential depend upon particulate size, structure, surface area and components adsorbed to the surface, in addition to host airway geometry (Shapiro et al., 2013). Camp et al. (2014) found that female COPD patients with exposure history to tobacco smoke express differential airway diameters when compared to patients exposed to wood smoke. Both PM sources induce different phenotypes of COPD. Subjects of the biomass-exposed group expressed symptoms of bronchiolitis, the airway-predominant phenotype of COPD associated with increased bronchial wall thickness and air trapping. Tobacco smoke was found to induce a combination of bronchiolitis and the emphysema-predominant phenotype, characterised by reduced expiratory flow, emphysematous destruction of lung parenchyma and bronchiole narrowing (Camp et al., 2014).

According to Sehlstedt et al. (2010), particulates derived from wood smoke and diesel exhaust have few compositional similarities and elicit significantly different host responses. The population-based study of Bui et al. (2013) found that ambient wood smoke and emissions of diesel exhaust are both associated with asthma severity via exacerbation of airway inflammation. However, Sehlstedt et al. (2010) suggest that wood smoke is relatively less toxic to the airways. Particulates derived from diesel exhaust are soot aggregates with a carbonaceous core coated with hydrocarbons, metallic ash and volatile organic particles; while particulates of wood smoke are carbon soot particles coated with PAHs and reactive oxygenated species (Totlandsdal et al., 2014; Sehlstedt et al., 2010). According to Totlandsdal et al. (2014), wood smoke particulates are more heterogeneous according to variations in fuel characteristics and combustion conditions. Louwies et al. (2013), Sandradewi et al. (2008) and Kocbach et al. (2008) found wood smoke particulates to contain higher oxygen and PAH content due to the organic compounds generated during combustion. However, Totlandsdal et al. (2014) found higher organic carbon (OC) content in diesel exhaust and similar PAH content between both sources. The inflammatory and cytotoxic potentials of particulates derived from wood smoke and diesel exhaust are significantly different (Kocbach et al., 2008). When exposed to monocytes and alveolar cells, traffic-derived particulates presented higher inflammatory potential, while those of wood smoke presented higher cytotoxicity, inducing a significant reduction in viable cell number (Kocbach et al., 2008). Contrarily, Totlandsdal et al. (2014) found that both the inflammatory potential and cytotoxicity of diesel exhaust particulates are significantly higher than those of wood smoke particulates when exposed to human bronchial epithelial cells. Variable fuel characteristics and combustion conditions may explain such discrepancies in results. Bølling et al. (2012) found that under specific combustion temperatures, wood smoke particulates were more toxic and exceeded the inflammatory potential of diesel exhaust particles when exposed to human pulmonary epithelial cells.

1.6 Chemical characterisation of wood smoke

Anhydrides, produced from thermal degradation of cellulose, are the most copious constituent of biomass combustion emissions and commonly used tracers for ambient wood smoke (Bergauff et al., 2008). However, the atmospheric instability of levoglucosan compromises its capacity to serve as a reliable tracer over time (Heringa et al., 2011). The chemical composition of wood smoke depends upon fuel characteristics and conditions of combustion air and temperature (Fine et al., 2004; Bølling et al., 2012; Riddervold et al., 2011).

Research has investigated the influence of wood species and moisture upon particulate emissions. According to Schmidl et al. (2008) and Weimer et al. (2008), wood species exerts minor influence upon the composition of wood smoke, despite variable ratios of levoglucosan and its derivatives detected between softwood and hardwood species. Combustion of highmoisture fuel produces emissions of CO, PAHs and VOCs, due to the reduced temperature and combustion efficiency initiated by the significant amount of energy required to vaporise water (Shen et al., 2012; Schmidl et al., 2008). Duration and conditions of firewood storage between harvesting and burning influence fuel moisture content. However, the rate of drying varies among individual species (Nord-Larsen et al., 2011).

According to Weimer et al. (2008) and Bølling et al. (2012), particulate emissions are predominantly associated with combustion phase and temperature. The combustion cycle is characterised by sequential variations in temperature, corresponding with dehydration, oxidation, pyrolysis and gasification (Weimer et al., 2008). The ignition or start-up phase reflects conditions of low temperature and elevated emissions whereby volatilisation of hydrocarbons, gasification of pyrolysis gases and evaporation of water occurs (Weimer et al., 2008). Steady-state refers to pyrolysis, or thermal decomposition of biopolymers under high

temperature and reduced emission concentration (Bølling et al., 2012). Smouldering burn-out phase is characterised by declining temperature, oxidation of carbonaceous material and the presence of glowing chars (Bølling et al., 2012; Hukkanen et al., 2012). According to Tapanainen et al. (2012), particulates emitted from the start-up phase of combustion are most genotoxic and potent activators of apoptosis in human bronchial epithelial cells. Oxidative and inflammatory potentials of wood smoke particulates decrease with increasing combustion temperature (Bølling et al., 2012). These findings are supported by Danielsen et al. (2011) and Hytönen et al. (2009) who observed significantly higher proportions of PAH emitted during low-temperature and low-oxygen, smouldering conditions, compared to high-temperature and high-oxygen, flaming conditions. Production of ROS induced by low-oxygen wood smoke particulates *in vitro* may be attributed to the relatively high proportion of PAH generated from incomplete combustion (Danielsen et al., 2011; Tapanainen et al., 2012). PAHs induce oxidative stress via metabolism of intracellular enzymes (Tapanainen et al., 2012). Assessment of emissions generated from a conventional wood heater under incomplete combustion temperature range of 700°C to 1000°C revealed that particulates collected during lower temperature combustion were more toxic and higher in PAH content than those generated under higher temperatures (Bølling et al., 2012). These findings support the inverse relationship between combustion temperature and particulate toxicity (Bølling et al., 2012). Similarly, Lanz et al. (2007) found that low-temperature combustion generates higher concentrations of organic aerosols and CO, compared to high-temperature combustion. As supported by Stockfelt et al. (2013) and Weimer et al. (2008), Bølling et al. (2012) concluded that the content of VOCs and PAHs are highest during the start-up phase of combustion when temperature is at a minimum. However, thermal decomposition of PAHs may be achieved by combustion temperatures exceeding 900°C (Bølling et al., 2012). As temperature rises, volatile compounds are combusted more efficiently (Tapanainen et al., 2012).

1.7 Wood combustion technology

Compared to modern combustion technology, conventional wood heaters may generate emissions of variable chemical composition and have reduced capacity to comply with national reporting standards for ambient air quality. Restricted air flow and unstable temperature control systems elevate smoke emissions and inhibit oxidation of VOCs and CO (Miljevic et al., 2010; Fine et al., 2004). Emissions of incomplete combustion are reduced by modern and automatic wood pellet burners designed to achieve more rapid and complete oxidation of hydrocarbons and CO with sophisticated airflow and heat convection systems (Heringa et al., 2011; Miljevic et al., 2010; Weimer et al., 2008; Tissari et al., 2008). Automatic loading of wood pellets maintains uniform combustion temperature (Heringa et al., 2011). Secondary combustion air chambers of modern wood heaters facilitate the ignition of combustion gases and soot to ultimately reduce particulate emission into the atmosphere (Tissari et al., 2008). Particulate emissions generated from pellet burners and conventional wood heaters are significantly different (Heringa et al., 2011; Weimer et al., 2008). As observed by Miljevic et al. (2010), temperatures attained by the former are significantly higher across all stages of combustion and the elevated surface area of wood pellets increases combustion efficiency (Weimer et al., 2008). Controlled airflow reduces the formation of thermal NO_x and provides oxidative conditions for combustion of VOCs (Weimer et al., 2008). According to Heringa et al. (2011), automatic pellet burners produce significantly lower emissions of PM and PAHs, compared to conventional wood heaters. In addition, the ratio of carbon monoxide (CO) to carbon dioxide (CO₂) is significantly reduced (Miljevic et al., 2010).

Catalytic oxidation of combustible compounds may provide an economical solution to residential wood smoke pollution (Hukkanen et al., 2012). According to Kaivosoja et al. (2012), catalysts act upon benzene rings of PAHs to ultimately reduce genotoxic and

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carcinogenic potential. Noble metals and transition metal oxides are two distinctive classes of catalysts, designed to facilitate combustion efficiency, generating heat from oxidation of hydrocarbons and CO (Liotta et al., 2010; Fine et al., 2004; Kaivosoja et al., 2012). Both catalyst classes have been found to reduce PAH, PM and CO emissions during all phases of combustion (Hukkanen et al., 2012; Kaivosoja et al., 2012). However, studies of Carnö et al. (1996) and Ferrandon et al. (1999) reported different catalytic capacities between both classes in relation to oxidation of methane, naphthalene and CO. As opposed to Ferrandon et al. (1999) and Carnö et al. (1997), Carnö et al. (1996) found platinum-based catalysts to be more stable with capacity to resist deactivation, while facilitating oxidation of combustible compounds. According to Kaivosoja et al. (2012), platinum-based catalysts also initiate chlorination of persistent organic pollutants (POPs).

Fine et al. (2004) contend that catalytic elements generate more PAH via aromatisation of natural wood components and ultimately produce more elemental carbon (EC) via further initiation of pyrolysis of organic compounds. Of the 22 wood species tested in the study, catalytic combustion only reduced PM emission concentration for one and ultimately generated higher emission concentrations of EC and PAH, compared to non-catalytic combustion. These results are contradictory to those of Hukkanen et al. (2012) who found that incorporation of a platinum-palladium catalyst reduced PM emissions by 30%, compared to non-catalytic combustion, in addition to a 21% reduction in CO emissions. Similarly, Kaivosoja et al. (2012) found catalytic combustion to induce a 24% and 26% reduction in emissions of PAH and CO, respectively.

Research indicates that catalysts are only effective under optimal conditions. Physical deposition and surface condensation of ash, soot and tar deactivate catalytic reactions, and water vapour inhibits oxidation of VOCs (Carnö et al., 1996; Yildiz et al., 2014). Inadequate combustion temperature and the presence of certain combustible compounds have been found

to impede catalytic activation. Methane has a very low oxidation rate and the presence of naphthalene impedes the conversion of CO to CO₂ (Ozil et al., 2009; Carnö et al., 1997). Furthermore, catalytic activation requires attainment of a certain threshold temperature that is not achieved during the start-up and burn-out, smouldering phases of combustion which generate the highest proportion of particulate emissions (Ozil et al., 2009). Ideal temperature thresholds are variable between studies. However, Carnö et al. (1996) recommend that combustion temperature should exceed 400°C in order to achieve catalytic oxidation of methane, naphthalene and CO. Various strategies have been researched to overcome the temperature-dependent activation of wood combustion catalysts. Ferrandon et al. (1999) found pre-heating the catalyst during start-up phase via injection of hot air to reduce CO emissions by 67% within the first ten minutes of combustion. Incorporation of an electric heating system in the study of Ozil et al. (2009) was found to reduce CO and VOC emissions during ignition by 50% and 17%, respectively.

1.8 Interventions to reduce residential wood smoke

Air quality influences population mortality (Johnston et al., 2013). Strategies to reduce PM emissions are therefore essential to alleviate the health burdens of wood smoke in residential areas where wood combustion is a significant source of ambient air pollution (Sandradewi et al., 2008). In order to achieve public attainment of desired outcomes, such strategies must be economical (Carnö et al., 1996). Bui et al. (2013) suggest clean burning practices and replacement of conventional wood heating appliances, while Hine et al. (2011) suggest the provision of educational material and catalytic devices to wood heater households. The two-week household implementation of high efficiency particulate air (HEPA) filters conducted by Allen et al. (2011a) found the filters to reduce mean indoor PM_{2.5} concentrations by 60% and to be associated with improved endothelial function. Reduced concentrations of inflammatory biomarkers and excretory products of oxidative degradation were detected in

the blood and urine among residents of a residential wood smoke-affected area. A wood heater replacement programme of Launceston, Tasmania was initiated during the winter of 2001 to replace conventional wood heaters with electric heaters as the primary source of domestic heating (Johnston et al., 2013). The intervention achieved almost a 50% reduction in wood heaters as the primary source of domestic heating. Comparisons between pre- and post-intervention periods reflected minor changes to all cause, cardiovascular and respiratory mortality rates during winter (Johnston et al., 2013). Throughout the period of 2005 through to 2007, a community-wide wood heater replacement program was implemented in Libby, Montana, a valley community of the United States, ultimately replacing 95% of conventional wood heaters with new, EPA-certified stoves or alternatives appliances (Noonan et al., 2012a). The intervention achieved a 36% and 30% reduction to indoor and ambient PM_{2.5} concentration, respectively, and more than 60% reduction to concentration of various PAHs (Noonan et al., 2012a). Childhood incidences of wheezing, bronchitis, mucosal irritation and influenza were also reduced, post-intervention, in addition to frequencies of illness-related school absences (Noonan et al., 2012b; Noonan et al., 2011).

Hine et al. (2011) found wood heater installation of Smartburn® catalytic devices to exert direct reductions upon individual household emissions based upon householder selfassessment of smoke plumes generated from 168 participating households within Armidale, a university city in New South Wales. Educational material informing appropriate wood heater operation was found to exert indirect influence upon emissions (Hine et al., 2011). However, results of education and catalytic combustion were limited to qualitative and intermittent assessment of emissions generated from individual households according to the apparent density of smoke plumes. In addition, the demographic profile of Armidale may not reflect those of susceptible wood-burning communities in terms of age and socioeconomic status (Hine et al., 2011). The potential for the catalytic device to quantitatively improve ambient air quality and protect vulnerable groups, via community-wide adoption, remains unknown.

1.9 Outcomes and limitations of research

The human dose-response relationship of wood smoke is ambiguous, due to the discontinuous and variable nature of biomass combustion (Miljevic et al., 2010). The relative toxicity of wood smoke is therefore difficult to determine, compared to other PM sources (Totlandsdal et al., 2014; Bølling et al., 2012). Atmospheric ageing transforms the composition of pure wood smoke particulates further, into complex heterogeneous mixtures of variable toxicity (Heringa et al., 2011). Thus, inconsistent results among human dose-response studies may be attributed to variations in sample size, fuel characteristics, combustion conditions and PM dose, nature and duration of exposure. For instance, acute, pure wood smoke exposure may elicit differential host responses to chronic, ambient exposure. Wood smoke exposure during intervals of exercise, as opposed to rest, increases ventilation and subsequent respiratory deposition of particulates (Riddervold et al., 2012; Kurmi et al., 2013; Unosson et al., 2013). Despite such differences in experimental design, common conclusions are that inhalation of fine particulates elicits immediate and long-term, adverse ramifications for cardiovascular and respiratory health. Systemic inflammation, endothelial dysfunction and oxidative stress are events documented in several studies (Riddervold et al., 2012; Painschab et al., 2013; Danielsen et al., 2011). Chronic exposure has been shown to impair lung function, exacerbate asthma and initiate the development of COPD (Steinvil et al., 2013; Bui et al., 2013; Fullerton et al., 2011; Camp et al., 2014).

Spatial monitoring has revealed elevated wood smoke concentrations beneath thermal inversion layers and within residential areas of high wood combustion density and low elevation (Aymoz et al., 2007; Su et al., 2013). However, the cytotoxic and inflammatory potentials of wood smoke particulates remain influenced by the combustion conditions under

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which they are generated. Adequate temperature and combustion air are required for thermal decomposition of PAHs, NO_x and VOCs, and oxidisation of CO, respectively (Fine et al., 2004). Significant variability in wood heater operation among individual households contributes to the uncontrollable nature of residential wood smoke pollution. Even high-efficiency wood combustion appliances have potential to generate pollution, especially when operated under suboptimal conditions (Noonan et al., 2011). Similarly, the potential for catalytic combustion to achieve significant emission reduction requires ideal fuel characteristics and combustion conditions for catalytic activation and sustained thermal oxidation of combustible compounds. Ideal conditions require adequate airflow, good design and careful, well informed operation.

1.10 Conclusions

Ambient air pollution generated from residential wood combustion is carcinogenic and clearly associated with respiratory and cardiovascular risks. Human exposure to ambient particulates is influenced by local topography and meteorological conditions, while the chemical composition of wood smoke alone depends upon fuel characteristics and combustion conditions. Conventional wood heaters remain popular in residential areas, despite the availability of alternative energy sources and wood combustion appliances of advanced efficiency. Replacement of conventional wood heaters has been found effective for reducing community-wide emissions. Furthermore, catalytic devices have been found to exert a direct effect upon wood smoke emissions. However, distribution throughout an entire community has not been evaluated with ambient PM concentrations quantified. There is potential for community-wide installation of a catalytic device to alleviate the public health burden of wood smoke.

1.11 Aims and Hypotheses

The aim of the present study was to evaluate the potential for community-wide firebox installation of a wood combustion catalyst to reduce ambient air pollution generated from domestic wood heaters in Perth, Tasmania. Ambient air pollution of Perth and three control towns, unexposed to the catalytic trial, were simultaneously monitored in order to establish an association between catalyst use and air quality. It was hypothesised that post-intervention mean ambient $PM_{2.5}$ mass concentration ($\mu g/m^3$) of Perth would be reduced, compared to that of the pre-intervention phase and compared to those of the control towns, Longford, Hadspen and Westbury.

Chapter Two

SCIENTIFIC PAPER

Community-wide distribution of a catalytic device for domestic wood combustion to reduce winter ambient $PM_{2.5}$ mass concentration

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Formatted for the Journal of ATMOSPHERIC ENVIRONMENT (Appendix A)

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Abstract

Residential wood combustion is the main source of winter air pollution in Tasmania. Inhalation of fine particulate matter (PM_{2.5}) of wood smoke is detrimental to respiratory and cardiovascular health and potentially carcinogenic. The aim of this study was to evaluate the potential for community-wide distribution of a firebox catalyst to reduce ambient PM2.5 concentration in Perth, Tasmania. In June 2014, residents of Perth were offered the device for placement in the firebox of their wood heater, in absence of receiving operational training in optimal use of their wood heater. Ambient air pollution was sampled from fixed air monitoring stations of Perth and three control towns, Longford, Hadspen and Westbury, throughout wood heater seasons of 2013 (pre-intervention) and 2014 (post-intervention). Participants were surveyed, post-intervention, to determine the conditions of combustion and catalyst treatment that were adopted throughout the intervention. A two-factor general linear model (GLM) was used to compare ambient PM2.5 concentrations among the sampled locations and between intervention phases. Almost 80% of Perth wood heater operators accepted a catalytic device. However, there was no significant post-intervention reduction to ambient PM2.5 concentration of Perth, compared to the control communities. Most participants failed to maintain ideal conditions for the catalyst within the firebox and observed no signs of catalyst function. Winter ambient air quality of Perth was ultimately not influenced by the catalytic trial. Poor maintenance of the catalyst and combustion appliance may have inhibited activation and function of the catalyst.

Key words: residential wood combustion, catalyst, ambient PM_{2.5}, Perth, Tasmania.

2.1 Introduction

Residential wood smoke is a significant source of winter air pollution in various parts of the world. In Christchurch and Temuco, direct associations have been reported between ambient wood smoke and population mortality (McGowan et al., 2002; Sanhueza et al., 2009). Chronic exposure has been associated with the development of respiratory and cardiovascular disease, particularly asthma, chronic obstructive pulmonary disease (COPD), lung cancer, pneumonia, heart failure and stroke (Camp et al., 2014; Sood et al., 2010; McGowan et al., 2002; Gold and Mittleman, 2013). The chemical constituents of wood smoke, responsible for these health effects, have become well known. Polycyclic aromatic hydrocarbons (PAHs), volatile organic compounds (VOCs) and particulate matter (PM) are carcinogenic and induce systemic inflammation and oxidative stress when inhaled (Danielsen et al., 2008; Stockfelt et al., 2013). In ambient air, the chemical profile of wood smoke becomes more complex and atmospheric aging increases toxicity (Hennigan et al., 2011; Ding et al., 2012). In Australia, ambient concentrations of fine PM (PM_{2.5}) exceeding a daily average of $25\mu g/m^3$ are unacceptable, according to Schedule 2 of the National Environment Protection (Ambient Air Quality) Measure (National Environment Protection Council, 2003). However, the national reporting standards are not legally enforced.

The toxicity of ambient wood smoke presents a threat to public health. Efforts to reduce wood smoke require ambient air quality monitoring to identify communities susceptible to pollution and to evaluate post-intervention effects. Implementation of air monitoring programs has become more affordable for government agencies around the world (Environment Protection Authority (EPA), 2014b; Environment Protection Agency (EPA), 2014c). Recent implementation of the Base Line Air Network of the EPA Tasmania (BLANkET) enables continuous, real-time and quantitative assessment of state-wide ambient air quality in Tasmania (EPA, 2012). As opposed to the three pre-existing regional stations, the network of

19 fixed air monitoring stations measures air pollution by town. This network enables comparisons to be made between communities exposed and unexposed to an intervention and between pre- and post- intervention air quality. Such comparisons may determine the influence of an intervention upon residential wood smoke pollution.

Interventions designed to reduce residential wood smoke tend to adopt three distinct approaches: reduce the number of wood heaters within a community, replace conventional wood heaters with cleaner alternatives and improve wood heater operation via education. In Launceston, Tasmania, a wood heater reduction of almost 50% was achieved in 2001, and in Libby, Montana, a community-wide replacement programme of conventional wood heaters reduced ambient PM_{2.5} concentration by 30% (Noonan et al., 2012a; Johnston et al., 2013). Burn Wise is an educational program, coordinated by the EPA (EPA, 2014c) of the United States, designed to promote combustion of dry wood under ideal conditions using certified heaters. A similar intervention, Burn brighter this winter, was coordinated by the EPA of Tasmania, where communities susceptible to elevated wood smoke concentration were advised of appropriate burning practices via several forms of media (EPA, 2014a). However, regular assessment of smoke plume densities and ambient PM2.5 concentrations revealed no significant post-intervention reduction to wood smoke (EPA, 2014a). Approaches to reduce ambient wood smoke via elimination and replacement of domestic wood heaters may not be economical or politically acceptable in many circumstances. Educational approaches may be less successful due to public resistance to behaviour change (Reeve et al., 2013).

Catalytic combustion is another, potentially more practical and economical approach to reducing residential wood smoke emissions. Wood combustion catalysts have been reported to reduce emissions of PM and PAH by up to 30% via thermal oxidation (Hukkanen et al., 2012; Kaivosoja et al., 2012). Simple firebox installation of a catalytic device, in the absence of altering wood heater operation, may be more acceptable and achievable by most members

of a community. A commercially available, catalytic device has been promoted in Australia to reduce accumulation of soot and creosote within the flue and firebox (Smartburn Pty Ltd, 2014). The steel tubular device encases a solid catalytic mixture. As combustion temperature rises, the mixture vaporises, escaping from open ends of the casing and facilitates combustion via ignition of gases and soot within the firebox (Smartburn Pty Ltd, 2014). The catalyst has been reported to reduce particulate emissions by up to 54% in the laboratory and reduce the density of smoke plumes generated from individual wood heaters (Smartburn Pty Ltd, 2014; Hine et al., 2011). However, the influence of this technology upon ambient pollution has not been quantified community-wide.

The present study was set in Northern Tasmania, incorporating the rural towns, Perth, Longford, Hadspen and Westbury, with respective populations of 2567, 3053, 2063 and 1476 (Australian Bureau of Statistics (ABS), 2011e; ABS, 2011c; ABS, 2011b; ABS, 2011d). As for most rural towns of Tasmania, wood combustion is the main source of air pollution, with approximately 50% of households operating a wood heater for domestic heating (Innis, 2012). Daily mean ambient $PM_{2.5}$ mass concentrations often exceed $25\mu g/m^3$, especially on winter nights with low temperature and low wind speed (Innis, 2012). The four towns are surrounded by catchments of the Tamar River and have similar topography and climate (Innis, 2012; Department of Primary Industries, Parks, Water and Environment (DPIPWE), 2012).

The present study was designed to evaluate the influence of community-wide distribution of a catalytic device upon Perth's winter ambient air quality. It was hypothesised that the post-intervention concentration of ambient $PM_{2.5}$ would be lower compared to the pre-intervention phase, and lower compared to the three control towns.

2.2 Methods

In June 2014, all Perth households identified as using firewood for domestic heating were offered a free catalyst. In order to raise community awareness prior to door-to-door distribution of the device, objectives of the study were promoted via Facebook and the website of the municipal Council, direct mail, television, newspaper and static display of posters in various local businesses of Perth.

2.2.1 Community-wide distribution of catalytic devices

On the weekend of June 14 and 15, 2014, 27 volunteers recruited from the University of Tasmania offered the catalytic device to as many wood heater operators of Perth as possible. Canisters were donated by Smartburn Australia and representatives instructed volunteers in inserting and maintaining the product. Company representatives were not involved in the design and implementation of the intervention, or the analysis and reporting of results. Volunteers' recorded residential addresses visited and indicated whether residents' were home and a wood heater was present, their willingness to participate and the number of canisters accepted per household (Appendix B). Residents were offered information sheets and written consent was obtained to contact them during September and evaluate their use of the device. Volunteers left flyers in mailboxes of houses that were visited twice with no response. The flyer directed residents to the Perth Roadhouse Service Station where it could be exchanged for a free canister (Appendix C). Sections of streets that were inadvertently missed were visited during the final week of June. Flyers were collected from the Roadhouse and new residential addresses were added to the database.

2.2.2 Fixed ambient air sampling

The BLANkET air monitoring stations of Perth, Longford, Hadspen and Westbury were used to measure hourly mean $PM_{2.5}$ concentrations and meteorological parameters. PM mass concentration was measured at 10-minute intervals via optical particle counters which are non-gravimetric, low-volume samplers that operate under the technique of light scattering (EPA, 2013a). Air particulates drawn into the monitoring chamber pass through a laser beam and scatter light which is then reflected to a detector. The amount of reflected light energy detected corresponds with a particular mass and diameter which may be converted to particulate mass concentration in micrograms (µg) per unit volume (m³) (EPA, 2013a). All samplers are calibrated against gravimetric samplers for $PM_{2.5}$. The wireless Davis Vantage Pro 2 weather station measured meteorological parameters of wind speed, atmospheric pressure, ambient temperature and relative humidity via an anemometer, a barometer and temperature and humidity sensors (Davis Instruments Australia Pty Ltd, n.d.).

2.2.3 Participant survey

A survey was designed to identify observed impacts of the catalyst among participants and any factors that may influence function of the device (Appendix D). Respondents reported the type and age of their wood combustion appliance, and typical durations that complete primary combustion airflow is maintained after ignition and reloading. Wood moisture was indicated by self-reported durations and conditions of firewood storage. Respondents also reported the duration and conditions of placement and maintenance of the canister within the firebox. The Smartburn® catalyst has an average lifetime of three months. Within the firebox, the canister must be placed level and to one side and remain elevated, maintaining the openings free for vapour escape and subsequent oxidation of combustible compounds (Smartburn Pty Ltd, 2014). Any changes to the wood heater observed throughout canister use were reported and intentions to purchase the catalyst in the future were indicated. Microsoft Excel 2010 was used to tabulate original PM and meteorological data obtained from BLANkET stations of the sampled locations. Mean hourly PM_{2.5} concentrations, across the entire sampling period, were graphically presented and the results of the participant survey and door-to-door distribution were tabulated. The entire study population was illustrated using QGIS version 2.6, to indicate proportions of wood heater houses that did and did not receive a catalytic device. Hourly PM_{2.5} concentrations were used to generate the mean 24-hour diurnal cycle of particulate pollution for all locations.

In order to detect an association between catalyst use and ambient air quality, three control towns were incorporated: Longford, Hadspen and Westbury. Westbury was the reference community, as it was unexposed to any wood smoke intervention throughout the entire sampling period. A public education campaign, *Burn brighter this winter* was carried out in Hadspen and Longford during winter seasons of 2013 and 2014, respectively (EPA, 2012).

Ambient air quality data of the four locations from 2013 and 2014 were analysed for the colder months only (i.e. excluding the seven months of October through to April). Data were allocated to one of three sampling phases as follows: pre-intervention, initiation and post-intervention (Table 1). Initiation was terminated at completion of door-to-door distribution and once the highest proportion (18%) of flyers had been exchanged for a free catalyst.

A generalised additive model (GAM) was generated with the response variable being mean hourly ambient $PM_{2.5}$ concentrations of Perth, Longford, Hadspen and Westbury, and the predictor variables being ambient temperature, wind speed, relative humidity and atmospheric pressure. Respective associations between the meteorological parameters and PM concentration were established using a non-linear, quasi-Poisson model. A two-factor general linear model (GLM) was then conducted using the residual mean hourly $PM_{2.5}$ concentrations to identify an interaction between location and intervention phase. Two-factor GLMs were conducted for the hours of 11pm, 7am and 6pm, when ambient smoke concentrations were highest throughout the day. The analyses compared pre- and post- intervention phases in order to identify a post-intervention change to Perth air quality that was absent for the control communities. Graphical data were presented as mean hourly $PM_{2.5}$ mass concentration ($\mu g/m^3$), partial plots of $PM_{2.5}$ mass concentration \pm the standard error of the mean (SEM) and 95% confidence intervals (CIs) of the mean. Significant results were defined as *p<0.05, **p<0.01 and ***p<0.001. Statistical analyses were conducted using R version 3.1.0. The research reported in this paper received official approval from the Tasmania Social Sciences Human Research Ethics Committee.

Table 1: Durations of ambient air sampling phases for all sampled locations

Sampling Phase	Duration
Pre-intervention	May 17, 2013 to June 14, 2014
Initiation	June 14, 2014 to June 30, 2014
Post-intervention	July 1, 2014 to September 30, 2014

2.3 Results

2.3.1 Sample population

Perth, Tasmania comprises 1,092 private residences (ABS, 2011a). Throughout the initiation phase, a total of 899 (82%) residences were visited, of which 46% were confirmed to have wood heaters. There was also identification of two open fires and one wood pellet burner. Distribution of catalytic devices was cumulative. Of the wood heater residences, 283 (approximately 68%) received a catalytic device during door-to-door distribution on June 14 and 15, 2014. The remainder were either not interested or not home and received a flyer in the

mail. Some residents reported to have replaced firewood with alternative heating sources. At the end of August 2014, flyer collection from the Perth Roadhouse raised the sample population to 326 households, representing approximately 78% of wood heaters in the Perth community (Figure 1). Wood heater presence was unknown for some houses, most of which received a flyer. However, a small proportion of residences (less than 1%) did not appear to have a mailbox or visitor access.

2.3.2 Ambient PM_{2.5} mass concentration for all sampled locations

Mean hourly $PM_{2.5}$ mass concentration, recorded for all sampled locations, followed a similar pattern throughout the entire sampling period (Figure 2). Figure 3 represents the observed correlation among the towns more closely, with two consecutive days of mean hourly $PM_{2.5}$ concentration extracted from the sampling period. All four sampled locations recorded similar mean particulate concentrations between pre- and post- intervention phases (Table 2). As for the proportion of sampling days in exceedance of $25\mu g/m^3$, $PM_{2.5}$ concentrations tended to increase slightly, post-intervention for all locations. The highest concentrations were recorded for Longford (Table 2).

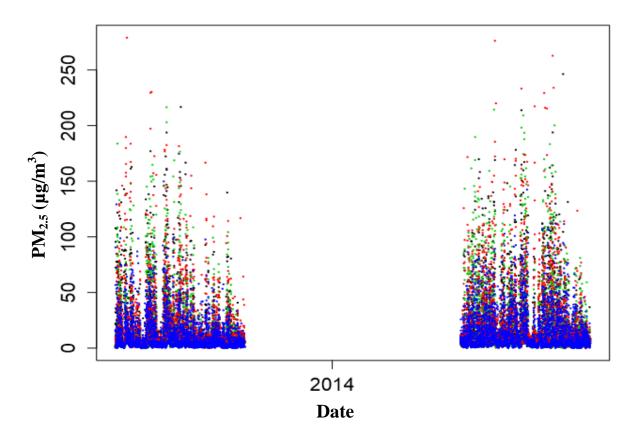


Figure 2: Mean hourly ambient PM_{2.5} mass concentration of Hadspen (black), Longford (red), Perth (green) and Westbury (blue) throughout the entire sampling period (May 17-September 30, 2013 and May 17-September 30,

2014).

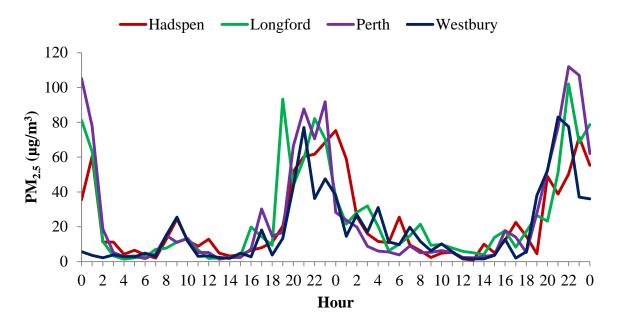


Figure 3: Mean hourly ambient PM_{2.5} mass concentration over two consecutive smoky days (June 29 and 30, 2014), showing the observed correlation among the four sampled locations.

Location (treatment)	Perth (catalytic trial)		Longford (education)		Hadspen (education)		Westbury (control)	
Intervention Phase	Pre	Post	Pre	Post	Pre	Post	Pre	Post
Mean PM _{2.5} (μg/m ³)*	17	20	19	22	17	19	12	15
Days Exceeding 25µg/m ³ (%)*	41 (26)	26 (29)	48 (30)	33 (37)	42 (26)	27 (30)	7 (4)	14 (16)
Total Days	160	90	160	90	160	90	160	90

Table 2: Mean PM_{2.5} concentration and total exceedance days of $25\mu g/m^3$ by location and intervention phase.

*values have been rounded to the nearest integer

The mean diurnal cycles of ambient $PM_{2.5}$ concentration were similar for Hadspen, Longford, Perth and Westbury, peaking at 11pm to $45\mu g/m^3$, $48\mu g/m^3$, $44\mu g/m^3$ and $27\mu g/m^3$, respectively; followed by secondary peaks at 7am ($15\mu g/m^3$, $21\mu g/m^3$, $19\mu g/m^3$ and $15\mu g/m^3$) and 6pm ($19\mu g/m^3$, $23\mu g/m^3$, $18\mu g/m^3$ and $16\mu g/m^3$), and minimum concentrations recorded at 1pm and 2pm ($4\mu g/m^3$, $5\mu g/m^3$, $3\mu g/m^3$ and $3\mu g/m^3$) (Figure 4).

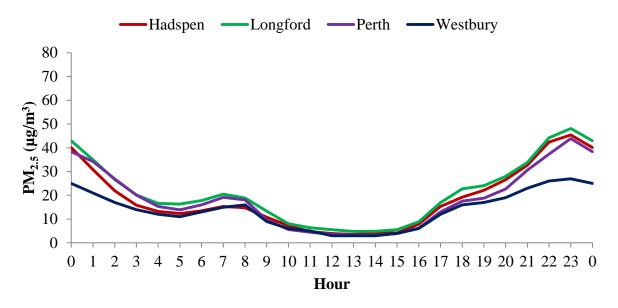


Figure 4: Mean diurnal ambient PM_{2.5} concentration for Hadspen, Longford, Perth and Westbury across the entire sampling period (May 17-September 30, 2013 and May 17-September 30, 2014).

2.3.3 Meteorological parameters for all sampled locations

According to the GAM, all meteorological parameters of ambient temperature, wind speed, relative humidity and atmospheric pressure accounted for 57% of variation in ambient $PM_{2.5}$ concentration for all locations. Thus, a combined quasi-Poisson model, encompassing all meteorological parameters, was adopted.

2.3.3.1 Ambient temperature

Mean ambient temperature was similar among the locations and between intervention phases; however, slightly lower in Westbury and post-intervention, for all locations (Table 3). Ambient temperature was inversely associated with $PM_{2.5}$ concentration (Figure 5). According to the GAM, 41% of deviation in $PM_{2.5}$ concentration was attributed to variation in temperature.

2.3.3.2 Wind speed

Mean wind speed was similar among the locations and between intervention phases; however, slightly lower in Longford and post-intervention, for all locations (Table 3). Wind speed was inversely associated with $PM_{2.5}$ concentration (Figure 5). According to the GAM, 43% of deviation in $PM_{2.5}$ concentration was attributed to variation in wind speed.

2.3.3.3 Relative humidity

Mean relative humidity was similar among the locations and between intervention phases; however, slightly lower in Perth and post-intervention, for all locations (Table 3). Relative humidity was directly associated with PM_{2.5} concentration (Figure 5). According to the GAM, 22% of deviation in PM_{2.5} concentration was attributed to variation in humidity.

2.3.3.4 Atmospheric pressure

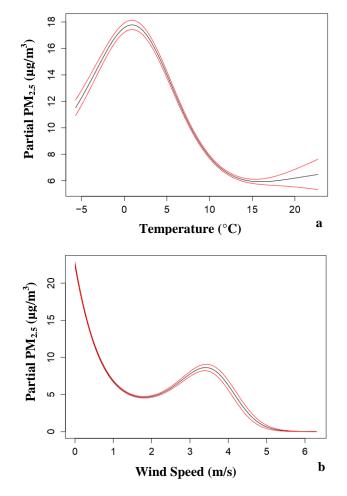
Mean atmospheric pressure was similar among the locations and between intervention phases; however, slightly higher in Perth and post-intervention, for all locations (Table 3). Atmospheric pressure was directly associated with $PM_{2.5}$ concentration (Figure 5). According to the GAM, 15% of deviation in $PM_{2.5}$ concentration was attributed to variation in pressure.

Table 3: Mean ambient temperature, wind speed, relative humidity and atmospheric pressure by location and

Location	Perth		Longford		Hadspen		Westbury	
(treatment)	(catalytic trial)		(education)		(education)		(control)	
Intervention								
Phase	Pre	Post	Pre	Post	Pre	Post	Pre	Post
Temperature								
(°C)	9.45	9.12	8.83	8.40	9.17	8.66	8.29	8.04
Wind Speed								
(m/s)	0.96	0.89	0.37	0.36	1.09	0.97	0.65	0.56
Relative								
humidity (%)	83.8	81.7	84.9	83.6	85.0	83.6	85.8	83.7
Atmospheric								
Pressure								
(hPa)*	1017	1020	1015	1018	1016	1019	1016	1019

intervention phase.

*values have been rounded to the nearest integer



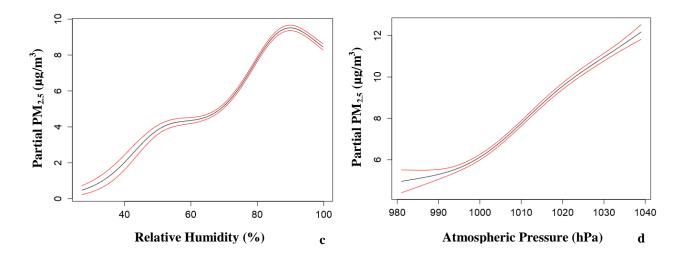


Figure 5: Partial plots of ambient PM_{2.5} mass concentration (mean ± SEM) attributed to ambient temperature (a), wind speed (b), relative humidity (c) and atmospheric pressure (d).

2.3.4 Mean PM_{2.5} concentration by location and intervention phase

2.3.4.1 Interaction between location and intervention phase

Hourly mean $PM_{2.5}$ concentrations among Westbury, Hadspen, Longford and Perth were not significantly different at both pre- and post- intervention phases (p>0.05) (Figure 6). There was no significant interaction between location and intervention phase upon mean $PM_{2.5}$ concentration (p>0.05).

2.3.4.2 Interaction between location and intervention phase at 11pm

At 11pm, mean $PM_{2.5}$ concentrations of Hadspen and Perth were significantly higher than those of Westbury, at both intervention phases (p<0.001 and p=0.0377, respectively) (Figure 6). However, there was no significant interaction between location and intervention phase upon mean $PM_{2.5}$ concentration at this hour (p>0.05).

2.3.4.3 Interaction between location and intervention phase at 7am

At 7am, mean $PM_{2.5}$ concentrations of Hadspen, Longford and Perth, were significantly higher than those of Westbury, at both intervention phases (p<0.001, p=0.0361 and p<0.001,

respectively) (Figure 6). However, there was no significant interaction between location and intervention phase upon mean $PM_{2.5}$ concentration at this hour (p>0.05).

2.3.4.4 Interaction between location and intervention phase at 6pm

At 6pm, post-intervention mean $PM_{2.5}$ concentrations were significantly higher, across all locations, compared to those of pre-intervention (p=0.00317) (Figure 6). Mean $PM_{2.5}$ concentrations of Hadspen and Perth were significantly higher than those of Westbury at both intervention phases (p<0.001 and p<0.001, respectively). However, there was no significant interaction between location and intervention phase at this hour (p>0.05).

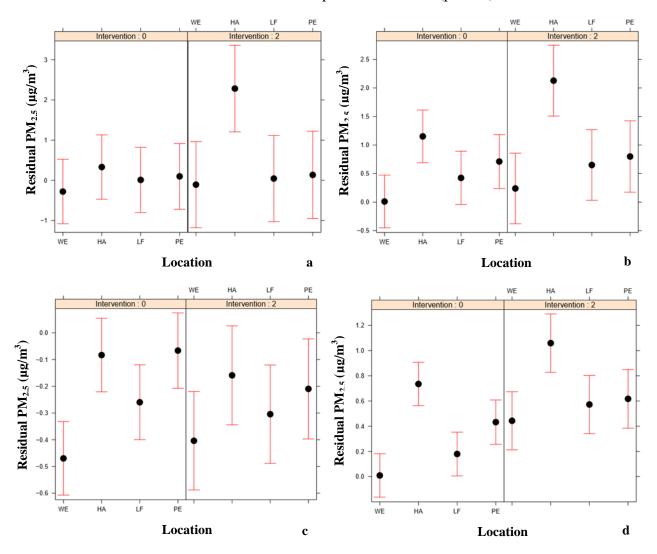


Figure 6: Mean PM_{2.5} mass concentration (95% CIs) of the sampled locations (WE=Westbury, HA=Hadspen, LF=Longford and PE=Perth) and intervention phases (0=pre-intervention and 2=post-intervention), across all hours (a), at 11pm (b), at 7am (c) and at 6pm (d).

2.3.5 Participant survey

Approximately 78% of the sample population provided consent for contact, via email or telephone (Appendix E). With a response rate of 52%, the survey reached 40% of the sample population. According to Table E.1, it may be concluded that throughout winter 2014, all respondents utilised a closed wood heater, as opposed to an open fire, wood pellet burner or any other appliance for domestic heating. Most (40%) wood heaters were relatively modern, aged less than ten years. As recommended by the EPA (2012), almost half of respondents reported to allow twenty minutes before reducing primary air intake after ignition and reloading. Firewood was stored in a dry, sheltered area by most (70%) respondents for up to six months (54%). Maintenance of the canister seemed ideal among 49% of participating residents, with the canister elevated and away from direct heat for more than two months (91%). However, 51% failed to maintain elevation within the firebox (Table E.1). About 41% observed combinations of clearance of soot from the flue and glass door, reduced smoke production from the firebox, elevated temperature and prolonged duration of combustion. However, most (51%) respondents observed no catalyst-related changes to their wood heater. Almost half (47%) of respondents intend to purchase the device in the future.

2.4 Discussion

Community-wide distribution of the Smartburn® device was not associated with a significant change to the ambient winter air quality of Perth. While the purpose of this study was not to evaluate educational interventions, Perth air quality data was compared with those of three nearby towns with different exposures to an EPA-coordinated public education campaign. There was no clear difference among the four communities that may be attributed to the catalytic trial.

Among the four sampled locations, there was little variation in hourly $PM_{2.5}$ concentration and meteorology throughout the entire sampling period (Figure 2; Figure 3; Table 2; Figure 4; Table 3). As reported by Bergauff et al. (2008) and Innis (2012), cold, calm, humid conditions were found to correspond with elevated $PM_{2.5}$ concentration (Figure 5). There was also a direct association established between atmospheric pressure and particulate pollution (Figure 5). This may be due to high pressure systems creating calm, clear conditions that are conducive to the formation of thermal inversions (Alfarra et al., 2007; Lanz et al., 2007).

Following adjustment for meteorological-induced variation in ambient pollution, there was no significant interaction established between location and intervention phase. This indicated that Perth air quality was not significantly different after the intervention, compared to the control communities. Post-intervention mean $PM_{2.5}$ concentration was not significantly reduced, compared to pre-intervention; nor was it reduced, compared to the post-intervention $PM_{2.5}$ concentrations of Longford, Hadspen and Westbury (Figure 6). Despite the significant influence of meteorology upon ambient pollution, similarities among the sampled locations and between intervention phases in ambient temperature, wind speed, humidity and atmospheric pressure do not explain the present findings. However, there are several possible explanations.

It is possible that the most polluting wood heater households of Perth were not reached by the catalytic trial (Figure 1). Within a community, only a small proportion of wood heater households generate the highest volume of particulate emissions (Todd 2013a). Wood heater operators among the 22% of Perth residents who declined participation may have represented such a proportion and therefore masked any significant influence that the trial may have induced upon ambient air quality. In absence of the intervention, the 78% of wood heater operators in receipt of a device may have contributed relatively little to community-wide emissions. Furthermore, close proximity to the air monitoring station of relatively smoky

chimneys may have disguised reductions to emissions achieved throughout the wider community. Future interventions to reduce ambient wood smoke may devise strategies to engage all residents of a community, especially those of the most polluting wood heater households.

Maintenance of the catalyst within the firebox may have been inadequate among participating households. The participant survey revealed that more than half of respondents failed to maintain the canister elevated and most respondents reported no observed catalyst-related changes to their wood heater (Table E.1). Such information indicates that appropriate maintenance of the catalyst was difficult to achieve, possibly leaving most devices inoperable and lacking ideal conditions within the firebox (Smartburn Pty Ltd, 2014). Future development of product design may be invaluable with 47% of respondents intending to purchase the device again (Table E.1). Various features may be introduced to compensate for the difficulty of maintaining level and elevated placement of the canister, away from excessive heat within the firebox. Incorporation of a small tripod-like device may be sufficient. With only few residents owning a fire brick, most participants indicated no means of preventing burial of the device in the ash-bed.

Poor and inconsistent conditions of combustion among participating households, from day to day, may have inhibited catalyst function. Subsequent alterations in temperature may have been particularly influential. Combustion of high moisture content-wood, insufficient provision of primary combustion air, and placement of logwood in configurations that prevent circulation of heat and oxygen within the firebox, create conditions of low-temperature smouldering combustion, under which catalytic activation is inhibited (Todd, 2013a; Ozil et al., 2009; Carnö et al., 1996). Catalytic performance may have been inhibited by durations of fast-burn combustion, under which sustained airflow and elevated combustion temperature increase the rate of vaporisation and exhaustion of the catalytic mixture (Smartburn Pty Ltd,

2014; Todd 2013b). Complete vaporisation of the catalyst may have occurred in less than three months. Most respondents reported that they allowed sufficient durations of complete combustion before reducing air intake and used firewood that had been stored in a dry, sheltered area for six months before burning (Table E.1). However, daily fluctuations in conditions associated with operation of both the combustion appliance and catalytic device were not objectively monitored throughout the intervention. Regular monitoring would have been impractical and potentially unacceptable to conduct among all participating households throughout the entire sampling period. As a consequence, participant self-reporting of combustion conditions risked misrepresentation of the true, day to day variability in fuel moisture and combustion air from household to household. Variability in these parameters would have influenced combustion temperature and subsequent activation and function of the catalyst (Todd, 2013b; Ozil et al., 2009; Smartburn Pty Ltd, 2014).

The catalytic device may have lacked capacity to influence residential wood smoke emissions. While this was not a published finding of the only other study to have trialled the device, differences in experimental design are important to note (Hine et al., 2011). As opposed to subjective assessment of smoke plume density generated from individual wood heaters, the present study invited all wood heater households of a community to install the device and quantified wood smoke concentration in ambient air. In the laboratory, a significant association has been quantified between installation of the catalyst and reduced particulate emission generated from a single wood heater (Smartburn Pty Ltd, 2014). However, establishment of the same association in the field, with community-wide firebox installation of the catalyst and operation of wood heaters under 'real-world' conditions with quantitative assessment of ambient pollution, remains absent. In addition to catalyst design, techniques of wood heater operation require improvement before the influence of catalytic combustion upon ambient wood smoke may be established in a residential setting. The willingness of most

respondents to purchase the catalytic device in the future provides incentive for further research and development.

Strengths of this study included the success of community awareness-raising strategies, with a large sample population encompassing almost 80% of Perth wood heater households. Continuous ambient air sampling at 10-minute intervals from BLANkET stations across four locations was almost complete with only a few hours of data missing due to temporary dysfunction. Limitations of the study included the possibility of inaccurate estimation of coverage. It was not confirmed that all distributed canisters were actually installed in the firebox, maintained under advised conditions and remained there for the duration of post-intervention sampling. In most circumstances, volunteers were not invited in by residents to ensure appropriate installation of the device. While the participant survey was designed to partially compensate for this limitation, sole reliance upon self-reporting from only 40% of combustion and catalyst maintenance among individual households. Researchers of future studies may redress these limitations by assessing combustion conditions at the time of catalyst installation and at regular intervals, post-installation.

2.5 Conclusions

Community-wide distribution of a catalytic device did not reduce winter ambient $PM_{2.5}$ concentration in Perth. Results of the community trial indicate that catalysed combustion is difficult to achieve in a residential setting, without effective control over factors associated with maintenance and operation of the catalyst and combustion appliance. Future research is required to improve catalyst design and simplify effective operation for consumers. Research may also identify typical conditions of residential wood heater operation that inhibit catalyst function and devise strategies to control them.

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2.6 Competing Interests

No competing interests exist.

2.7 Author's Contributions

All work was carried out by the author with mentoring and writing assistance from Dr Fay Johnston and Dr John Todd (Supervisors), statistical analyses advised by Dr Grant Williamson (University of Tasmania) and all experimental data provided by John Innis (Environment Protection Authority, Tasmania).

2.8 Acknowledgements

This research was supported by the National Health and Medical Research Council (NHMRC) via the Centre for Air Quality Research and Evaluation, the Heart Foundation, Menzies Institute for Medical Research, the University of Tasmania and the Environment Protection Authority, Tasmania. The author acknowledges Smartburn Australia for donating catalysts.

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Appendix B – Record Sheet

Address	Time	Name	Home (Y/N)	Participate (Y/N)	No. Smartburn	Home 2nd Visit? (Y/N Flyer)	Comments





FIREBOX PROJECT Sorry we missed you. To receive your <u>FREE</u> canister:

1. Write your street address below

2. Visit Perth Caltex Roadhouse and present this letter to staff Open 7 days 6am-8pm

The UTAS Firebox Project is being conducted by the Menzies Research Institute Tasmania. The research is funded by the National Health and Medical Research Council (NHMRC) via the Centre for Air Quality Research and Evaluation, and the Tasmanian Heart Foundation.

Fire.Box@utas.edu.au



Appendix D – Participant Survey

Firebox Project 2014

University of Tasmania

Menzies Research Institute Tasmania

Evaluation for Perth Residents

The purpose of this survey is for researchers to understand your techniques of wood heater operation and any observations made while using your free Smartburn canister.

Please answer the 9 multi-choice questions below. Feel free to provide additional comments in section 10.

1. For home heating, I use a(n):

Open fire

Closed wood heater

Wood pellet fire

Other

- 2. Which is:
- <10 years old
- 10-20 years old

>20 years old

3. a) After lighting the fire, air pockets are left completely open for:

<5 minutes 10-20 minutes

5-10 minutes >20 minutes

b) After re-loading the fire, air pockets are left completely open for:

- <5 minutes 10-20 minutes
- 5-10 minutes >20 minutes
- **4.** This question relates to fuel moisture.

My wood is stored in: and harvested:

A dry, sheltered area Winter 2014

Outside, unsheltered

Summer 2013/2014

Other

Other

5. After receiving a canister in June 2014, it was left in the firebox for:

- <1 month
- 2 months
- >2 months

6. Since placement in the firebox, the canister remained:

Elevated, out of ash

To the side, away from direct heat

Both of the above

None of the above

Comments:

While using the canister, I noticed changes in: (Please select <u>all</u> relevant answers)
 Appearance of the heater

Operation of the heater

I didn't notice any changes

Please specify any observed changes:

8. I <u>have</u> noticed changes to the outside air quality of **Perth this winter**, compared to previous years.

YES

Unsure

NO

9. I would **purchase a canister** for my firebox in the future.

YES

Unsure

NO

10. Additional Comments

Appendix E – Results of Participant Survey

	No. Respondents	% of Respondents [×]							
Type and Age (year) of Wood Combustion Appliance (9% unsure)									
Wood heater	131	100							
< 10	52	40							
10 to 20	45	34							
> 20	22	17							
Complete Aeration (minute) after Ignition, Reloading (3%, 9% unsure)									
> 20	65, 61	50, 47							
10 to 20	47, 34	36, 26							
5 to 10	12, 13	9, 10							
< 5	3, 11	2, 8							
Conditions of Firewood Storage									
Dry, sheltered	92	70							
Outside	20	15							
Both	19	15							
Duration of Firewood Storage (13% unsure)									
Up to 6 months	71	54							
1-2 years	35	27							
> 3 years	8	6							
Canister Placement Conditions in Firebox									
Elevated & Side	64	49							
Side	55	42							
None	12	9							
Canis	Canister Placement Duration (month) in Firebox								
> 2	119	91							
< 2	11	9							
Observat	Observations during Canister Installation (8% unsure)								
No change	67	51							
Specific change	53	41							
Cleaner glass*	40	31							
Less smoke*	15	11							
Cleaner flue*	12	9							
Hotter fire*	11	8							
Longer burn*	11	8							
Intend to Purchase the Catalyst in Future (21% unsure)									
Yes	61	47							
No	43	33							

 Table E.1: Self-reported conditions of combustion and catalyst maintenance by number and proportion of respondents. Email received 31 responses and telephone received 100 responses.

×values have been rounded to the nearest integer

*more than one response may have been provided by individual respondents