A review of particulate matter emissions and impacts on human health: A focus on Canadian agricultural and rural emission sources

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Submitted: 2018 July 19 Revision received: 2019 February 17 Accepted: 2019 April 18 Published online: 2019 April 18

ABSTRACT

Particulate matter (PM) has been documented in an increasing number of research studies as having a known or suspected negative impact on human health. The World Health Organization (WHO) estimates that 3.1 million deaths were caused by ambient fine particulate matter ($PM_{2.5}$) in 2010. While many Canadian studies focus on health impacts from $PM_{2.5}$, there is a gap with respect to rural sourced $PM_{2.5}$ and health impacts in these areas. This paper reviews the impact $PM_{2.5}$ has on Canadians' health, investigates where $PM_{2.5}$ data is being gathered, and outlines the sources of $PM_{2.5}$ reported. Secondary inorganic aerosols that are formed in and around animal production facilities due to the higher prevalence of ammonia gas is of particular interest. The conclusion drawn is that the reporting and gathering of rural sourced $PM_{2.5}$ data is lacking, leading to a gap in the data used to determine the impacts on Canadian human health.

RÉSUMÉ

L'impact négatif connu ou potentiel sur la santé humaine de la matière particulaire (PM) a été documentée dans un nombre croissant d'études de recherche. L'Organisation mondiale de la Santé (OMS) estime que les fines particules ambiantes ($PM_{2.5}$) ont causé 3,1 millions de décès en 2010. Bien que de nombreuses études canadiennes portent sur les effets de $PM_{2.5}$ sur la santé, il y a une lacune en ce qui concerne le $PM_{2.5}$ d'origine rurale et les effets sur la santé dans ces régions. Le présent article examine l'impact des $PM_{2.5}$ sur la santé des canadiens, analyse les endroits où les données sur le $PM_{2.5}$ sont recueillies et décrit les sources du $PM_{2.5}$ signalées. Les aérosols inorganiques secondaires qui se forment à l'intérieur et autour des installations de production animale en raison de la prévalence plus élevée de l'ammoniac à l'état gazeux est d'intérêt particulier. Nous concluons qu'il n'y a pas suffisamment de données sur le $PM_{2.5}$ d'origine rurale, ce qui entraîne une lacune dans les données utilisées pour déterminer les effets sur la santé humaine au Canada.

KEYWORDS

Particulate matter, $PM_{2.5}$, secondary inorganic aerosols, health, agriculture, ammonia.

MOTS CLÉS

La matière particulaire, $PM_{2.5}$, Les aérosols inorganiques secondaires, santé, agriculture, ammoniac.

CITATION

Spencer, J., and B. van Heyst. 2018. **A review of particulate matter emissions and impacts on human health: A focus on Canadian agricultural and rural emission sources.** Canadian Biosystems Engineering/Le génie des biosystèmes au Canada **60**: 6.9-6.21. *https://doi.org/10.7451/CBE.2018.60.6.9*

INTRODUCTION

Particulate matter (PM) is one of several pollutants that make up the collective category of Criteria Air Contaminants in Canada (CACC) that have an adverse effect on human health, the environment, and economy (Government of Canada 2017). PM is categorized into two size fractions, coarse and fine. Coarse PM is referred to as PM_{10} due to it having an aerodynamic diameter of 10 μm or smaller. Fine PM has an aerodynamic diameter of 2.5 μm or smaller, is commonly referred to as $PM_{2.5}$, and is responsible for a growing list of health problems afflicting humans (US EPA 2016a).

Particle size and type determines the location of deposition once inside the human respiratory tract. The four types of deposition include: interception, impaction, sedimentation, and diffusion. Larger particles (>PM_{10}) will often intercept or collide with the surfaces of the upper respiratory tract and eventually be expelled through physical means (coughing and/or sneezing). Particles smaller than PM_{10} can however travel to the lower regions of the respiratory tract and settle on the tissues of the bronchi and bronchioles or diffuse into the gas exchange areas of the alveoli (CCOHS 2018). When the smaller particles are deposited in the lower regions of the lungs, they are more difficult to expel and can remain in the system for extended periods of time resulting in the onset of problematic health conditions (Koenig 2000; CCOHS 2018).

The PM, both coarse and fine fractions, has been linked to a variety of health problems including: cardiopulmonary and cardiovascular diseases, diabetes, and various cancers (Brook et al. 2013; Crouse et al. 2012; WHO 2016a). In 2010, it was estimated by the World Health Organization (WHO) that 3.1 million deaths were caused by ambient outdoor $PM_{2.5}$ (WHO 2013) with 91% of the global population exposed to concentrations that are above the recommended limit (annual mean of 10 μg/m^3) (WHO 2016a). Further to this, in 2012 the WHO also reported that, although not specific to PM, 12.6 million deaths (in 2012 alone) were attributed to environmental pollution (Prüss-Ustüm et al. 2016). Lelieveld et al. (2015) approximated that premature deaths caused by $PM_{2.5}$ was 3.3 million annually, while Silva et al. (2016) similarly found that anthropogenic $PM_{2.5}$ was responsible for 2.23 million deaths annually. While there is variability around the exact global mortality estimate, the end result still suggests that $PM_{2.5}$ is a significant contributing factor responsible for human mortality (Valavanidis et al. 2008). In response to the growing body of evidence linking PM to human mortality the WHO listed outdoor air pollution and particulate matter as cancer-causing agents, further solidifying the dangers associated with exposure to pollution and particulate matter (WHO 2013).

The WHO has set air quality guideline limits for 24-hour and annual PM averages in an attempt to reduce human exposure and with it, reducing the negative impacts on health. National and regional governing bodies are responsible for setting their own regulatory concentration limits. Regardless of the limits established by the WHO, 83% of monitoring sites located in Europe and Asia, recorded annual concentrations that surpass the WHO PM_{10} limits (no comparison was given for $PM_{2.5}$) (WHO 2013). In Canada, approximately 30% of its inhabitants are exposed to concentrations of $PM_{2.5}$ that exceeded the national limit (Brook et al. 2014). However, what is less understood is contribution to this excess from the different $PM_{2.5}$ sources and species and the associated health impacts to susceptible populations.

Particulate matter standards

Currently, regulatory bodies of individual countries set standards for ambient PM concentrations (this includes PM_{10} and $PM_{2.5}$). These regulations are based on research and policy development, with the goal being to decrease emissions to reduce exposure and ultimately improve human health. However, even with such limits and regulations in place, global PM concentrations are still increasing (WHO 2016b). Table 1 below summarizes standards for PM_{10} and $PM_{2.5}$ that are recommended by the WHO as well as current regulations in Canada. Current regulations in Europe and the United States are also listed for comparison.

In Canada, the Canadian Ambient Air Quality Standards (CAAQS) limits, which replaced the Canadian Wide Standards in 2013 (CCME 2014), are more stringent to promote human health. Included in the new standards are future target limits for $PM_{2.5}$ (annual and 24-hour time scales) effective in 2020. In the United States, standards for total suspended particles (TSP), PM_{10}, and $PM_{2.5}$ are set by the United States Environmental Protection Agency's (US EPA) National Ambient Air Quality Standards (NAAQS). These standards take PM monitoring a step beyond what Canada and the WHO have dictated by incorporating limits with respect to PM size fractions: primary and secondary. Primary standards are established to protect against human health and sensitive populations, while secondary standards are established to protect environmental welfare in terms of visibility, crop protection, animals, and vegetation (not to be confused with the primary and secondary classifications based on formation mechanisms) (US EPA 2016b).

Based on Table 1, and focusing on the $PM_{2.5}$ fraction, the WHO has developed the most stringent standards for both the 24-hour and annual mean concentrations of $PM_{2.5}$. Canada, while currently matching the WHO annual standard, will have the most stringent standard starting in the year 2020. In Europe, the $PM_{2.5}$ standard has an annual average of 25 μg/m^3 which is significantly higher than the annual WHO guideline (European Commission 2017).

Sources and formation of $PM_{2.5}$

Sources of $PM_{2.5}$ are varied and include industrial practices, transportation, agriculture, household heating methods, and naturally occurring events (e.g. volcanic eruptions) to name a few. Across Canada, the most common sources of $PM_{2.5}$ in order of greatest contribution as reported by Environment and Climate Change Canada (ECCC), are: open sources (occurring over large geographic rural areas and generated primarily from construction operations and agriculture), burning of firewood to heat homes, and other sources that include (in order): ore and mineral industries, transportation, manufacturing, miscellaneous, oil and gas industry, and energy uses (ECCC Air Pollutant Emissions 2017). In these reports it is not specified whether the $PM_{2.5}$ emissions are primary or secondary.

Table 1. Summary of selected air pollution standards for PM.

Regulatory Body	PM_{10}	$PM_{2.5}$	Source
WHO	Annual mean: 20 $\mu g/m^3$ 24-hour mean: 50 $\mu g/m^3$	Annual mean: 10 $\mu g/m^3$ 24-hour mean: 25 $\mu g/m^3$	WHO 2006
Canadian Ambient Air Quality Standards (CAAQS) – Replaced the Canadian Wide Standards in 2013	N/A	Annual mean (2015): 10 $\mu g/m^3$ Annual mean (2020): 8.8 $\mu g/m^3$ 24-hour mean (2015): 28 $\mu g/m^3$ 24-hour mean (2020): 27 $\mu g/m^3$	Environment and Climate Change Canada (CAAQS) 2013
European Commission	Annual mean: 40 $\mu g/m^3$ 24-hour mean: 50 $\mu g/m^3$	Annual mean: 25 $\mu g/m^3$	European Commission 2017
US EPA - National Ambient Air Quality Standards (NAAQS)	Primary and secondary – 24-hour mean: 150 $\mu g/m^3$	Primary annual mean: 12.0 $\mu g/m^3$ Secondary annual mean: 15.0 $\mu g/m^3$ Primary and secondary 24-hour mean: 35 $\mu g/m^3$	US EPA 2016b

When a particle emission is classified as primary, it means that it has been emitted directly from the source and has been unchanged in its form. Examples of primary PM include: dust, animal dander, and soil particles. Secondary $PM_{2.5}$ is formed when a reaction occurs between basic and acid precursor gases, such as ammonia and sulphur or nitrogen gases. This results in the two gases undergoing a neutralization via a gas-to-aerosol reaction, producing an aerosol particle in the $PM_{2.5}$ size fraction. Behera and Sharma (2010) found that, in an urban environment, these reactions accounted for 30% of the mass of $PM_{2.5}$ measured.

Ammonia is a basic gas involved in the gas-to-aerosol reaction for secondary $PM_{2.5}$ formation and is often generated in its greatest quantity in animal production facilities (Behera and Sharma 2010; Roumeliotis et al. 2010; ECCC 2016). The greatest sources of ammonia in Canada are from agricultural operations that include both animal and crop production (use of synthetic fertilizers) and represent 94% of all emissions (APEI 2019). In Canada ammonia emissions have increased by 24% since 1990 (ECCC 2018) due to agricultural practices that can contribute to the secondary inorganic aerosol (SIA) $PM_{2.5}$ burden.

Agriculture and Agri-Food Canada (AAFC) has been measuring and analyzing particulate matter emissions data from cropping activities nationally since 1993. This endeavor is part of the environmental indicators reporting program (known as Agri-Environmental Indicators (AEI)) that includes ammonia emissions, soil erosion, and soil cover data. The analysis of $PM_{2.5}$ emissions (total mass) from cropping operations has shown a declining trend that is attributed to the adoption of less intensive tilling practices which reduces the amount of particulate matter becoming air borne, and reduced summerfallow. Since 1993, $PM_{2.5}$ decreased from 522 kilotonnes/year to 276 kilotonnes/year in 2011, a reduction of 46% (AAFC PMI 2016).

The design and function of modern animal production facilities lends itself to increased emissions of secondary $PM_{2.5}$. Enclosed animal production facilities, common in Canadian agriculture, creates an environment suitable for the generation and accumulation of ammonia and acid gases such as sulphuric acid (H_2SO_4), nitric acid (HNO_3), and hydrochloric acid (HCl) (Behera and Sharma 2010). Ammonia is formed during the microbial breakdown of nitrogen-rich animal excrement (Behera and Sharma 2010; Roumeliotis et al. 2010). When acid gases and ammonia are present, a neutralization reaction occurs forming a secondary inorganic aerosol (SIA), contributing to the total $PM_{2.5}$ burden. The formed SIAs include: ammonium sulphate ($(NH_4)_2SO_4$), ammonium bisulfate (NH_4HSO_4), ammonium nitrate (NH_4NO_3), and ammonium chloride (NH_4Cl) (Behera and Sharma 2010). In poultry production, it is known that the formation mechanisms of these reactions are dependent on environmental conditions within the animal housing unit, the environmental conditions outside the housing unit including seasonal variability, and the animal's stage of growth (Roumeliotis et al. 2010). While government bodies have yet to regulate emissions from animal production facilities in Canada, understanding the extent of $PM_{2.5}$ contribution from agriculture can help to strengthen the overall understanding of this pollutant's impact on human health. Currently, secondary $PM_{2.5}$ is not directly estimated by Canadian governing bodies and therefore has not been reported in pollution summary reports.

Particulate matter and health – The Canadian context

The PM studies, whether they are focused on human health effects, source-receptor relationships, or formation mechanisms, utilize data gathered primarily from urban areas (WHO 2016b). Studies focused on human health impacts use urban-based PM data correlated with population health data that is also obtained from urban-dwelling participants. In terms of the Canadian context, the majority of the population (81%) is classified as living in an urban dwelling (Statistics Canada 2011) while the remaining population is located in smaller rural/remote areas of the country.

Research related to health impacts associated with particulate matter exposure is considerable. Health effects caused by the exposure to PM primarily affect the function of the lungs and the heart. Several conditions such as respiratory diseases, cardiovascular and cardiopulmonary conditions, diabetes, and cancers are associated with mortality and morbidity caused by $PM_{2.5}$ (Pope III et al.

2004; Pope III and Dockery 2006). Table 2 presents a summary of study's findings pertaining to health effects associated with long and short-term exposure to fine particulate matter in Canada since the year 2000.

The studies highlighted in Table 2 demonstrates that increased exposure to $PM_{2.5}$ within Canada correlates to an increase the risk of mortality. It also demonstrates that the majority of studies focus on populations and pollution measurements from urban locations. What was not included in Table 2 are studies that highlight the health impacts of PM exposure on children. For example, Tetreault et al. (2016) looked into the effects of exposure to PM and the onset of childhood asthma in Montreal, Quebec, Canada and found that the onset of asthma in children was associated with exposure to $PM_{2.5}$, NO_2 and O_3. Lavigne et al. (2017) looked at ambient air pollution and its effects on adverse birth outcomes in Ontario and determined that exposure to ambient $PM_{2.5}$ and NO_2 during pregnancy was associated with the onset of astrocytoma (a form of brain cancer) in children. Further, studies focused on pregnant women and the health of their newborns include Erickson et al. (2016) and Brauer et al. (2008) who both determined that expecting mothers who were exposed to $PM_{2.5}$, in urban areas, gave birth to children with lower birth weights. These studies, like the majority of those outlined in Table 2, were based on data gathered from urban dwelling populations in Canada and urban $PM_{2.5}$ data. What is less understood are the $PM_{2.5}$ emissions from rural areas and the impacts on the health of Canadian residents living in those areas.

Canadian agriculture and rural particulate matter

Canada's large landmass and low overall population density (3.9 persons/km^2 in 2016 (Statistic Canada 2018)) with several large urban centers (Statistics Canada 2017b) results in there being a large expanse of rural agricultural and/or natural landscapes. This combination of landscape extremes presents a unique combination of factors when discussing PM in the Canadian context. In Canada, $PM_{2.5}$ emissions are measured from a network of monitoring stations that are then analyzed and reported by ECCC. Data tables made available by ECCC from 2013 show that there was a total of 385 monitoring stations situated throughout the country gathering hourly data. An assessment of the gathered data from this network has been summarized below in Table 3.

Table 3 above outlines the available $PM_{2.5}$ data from monitoring stations across Canada for 2013. Based on the summary provided, the majority of data is collected in urban areas, primarily within British Columbia, Ontario, and Quebec. Of the $PM_{2.5}$ data gathered from monitoring stations in rural areas, the averages are similar to urban areas, but from significantly fewer stations. There was no rural data reported from two of the three prairie provinces (Manitoba and Saskatchewan) that represent a substantial area of Canada's agricultural land mass. It should be noted that the three territories do not have land mass suitable for standard agricultural practices and are therefore not a concern in this context. Of the 31 agricultural stations, only 13 reported data for either peak or average $PM_{2.5}$ concentrations in 2013, while only 33 rural area stations in total had data to report (ECCC 2014). Due to the majority of ambient monitoring stations being located in urban areas, comparatively fewer PM studies focusing on emissions from Canada's rural areas have occurred as outlined earlier in Table 2.

Agricultural land in Canada accounts for 158.7 million acres (7.1% of the total landmass) and is operated by approximately 272 x 10^3 farmers (Statistics Canada 2017a; Statistics Canada 2017b). Agricultural production in Canada accounted for 6.7% of the total national Gross Domestic Product in 2016, a growth of 11% since 2012 (AAFC 2017) and is the 5th largest global exporter of agricultural products (AAFC 2018). The farmers, their families, agricultural operations, and the rural landscape is often overlooked when it comes to studying PM, specifically monitoring and quantifying the concentrations of $PM_{2.5}$ and linking it to potential health impacts. With a predominate agricultural sector in Canada, in terms of production and landmass, what impact do the subsequent $PM_{2.5}$ emissions of have on human health?

Agricultural practices result in the emission of primary PM (all size fractions) from land preparation, nutrient and pesticide applications, crop harvesting, animal husbandry operations, and fuel use. In the most recent annual report from ECCC's Air Pollutant Emission Inventory (APEI) Report, it was estimated that 1.7 x 10^6 tonnes of $PM_{2.5}$ were emitted in Canada during 2017 with agriculture accounting for 3.80 x 10^5 tonnes, or 22% of the total $PM_{2.5}$ emissions (APEI 2019). Ammonia emissions totaled 4.80 x 10^5 tonnes with agriculture contributing 4.50 x 10^5 tonnes or 94% of emissions (APEI 2019). Overall, it was noted that $PM_{2.5}$ and ammonia have either remained stable or fluctuated slightly since 2012 (APEI 2019). This summary does not directly identify if the $PM_{2.5}$ emissions were primary or secondary. Secondary inorganic aerosol formation attributed to ammonia (agriculture) was also not identified. Table 4 below gives a summary of PM emission quantities with respect to their size fraction, and ammonia emission quantities from Canada as of 2017 based on APEI (2019) reported data.

In addition to the emission from agricultural and rural sources as outlined by APEI (2019), emissions of PM and ammonia from 'other' rurally located sources were included to demonstrate the magnitude of emissions possibly occurring outside urban centers in Canada. Off-road diesel and gasoline vehicles, which can include agricultural equipment, contribute to $PM_{2.5}$ emissions in the form of dust and fossil fuel combustion however, this contribution of $PM_{2.5}$ emission quantity is not included as part of the agriculture sector contribution (APEI 2019). These vehicles play a significant role in agricultural operations with respect to the transportation of products to and from the farm, and on farm transportation. When considering the full impact of agriculture and rural sources of $PM_{2.5}$, the contributions of the "other" sources listed in Table 4 should be included in the conversation. When all sources of PM and ammonia in both sectors are added together, the total

Table 2. Summary of Canadian studies pertaining to particulate matter and health impacts (presented in reverse chronological order for long- and short-term exposure; R – Rural, U – Urban, AL – All Land).

Study Reference	Study Location	Study Participants	Pollutant of Interest	Health Effects/Impacts
Long-Term Exposure:				
Chen et al 2017	Ontario, Canada (AL)	Residents aged 55-85 by April 1, 2001 with no diagnosis of dementia (2.1 million persons) who were followed until March 31, 2013	$PM_{2.5}$ exposure – satellite and global atmospheric chemistry transport model data for all of North America	The 257,816 cases of dementia diagnosed between 2001 and 2013 were positively linked to an increase in dementia due to exposure to $PM_{2.5}$ (average concentration of 10.4 μg/m^3).
Pinault et al. 2016	Canada (AL)	Respondents to the Canadian Community Health Survey: 299,500 individuals followed from 2000 to 2008	Low concentration ambient $PM_{2.5}$ (mean 6.3 μg/m^3) – satellite data	26,300 non-accidental deaths (32.5% due to circulatory disease, 9.1% due to respiratory disease). An increase of 10 μg/m^3 in $PM_{2.5}$ was found to be associated with an increase in non-accidental, circulatory, and respiratory mortality.
Stieb et al. 2015	Canada (AL)	Census Data for adults of 25 years of age from 2000 to 2011	Ambient $PM_{2.5}$ from 2000 to 2011 – satellite data	Determined a decrease in $PM_{2.5}$ by 25% during the years studied resulted in an increase in life expectancy by 0.10 years. An increase in $PM_{2.5}$, up to 3.5 μg/m^3, was observed over the prairies.
To et al. 2015a	Ontario (AL)	Women who enrolled in the Canadian National Breast Screening Study from 1980 to 1985 (29,549 women) and linked to administrative data to determine the prevalence of chronic disease between 1992 and 2013	Ambient $PM_{2.5}$ from 1998 to 2006 – satellite estimates of surface concentrations (no mean was given)	Congestive heart failure, diabetes, ischemic heart disease and stroke were all associated with a greater than 20% prevalence rate ratio based on a 10 μg/m^3 increase in $PM_{2.5}$
Villeneuve et al. 2015	Canada (AL)	89,248 women enrolled in the Canadian National Breast Screening Study between 1980 and 1985	Ambient $PM_{2.5}$ from 1998 to 2006 - satellite observations – average of 9.1 μg/m^3	A 10 μg/m^3 increase in $PM_{2.5}$ exposure was associated with elevated risk of non-accidental and ischemic heart disease mortality.
Chen et al. 2014	Ontario (AL)	35,303 non-hypertensive adults who responded to a health survey – 65% in urban areas	Ambient $PM_{2.5}$ from 2001 to 2006 - satellite average at participants residences 10.7 μg/m^3	For every 10 μg/m^3 increase in $PM_{2.5}$, the hazard ratio for incident hypertension is 1.13, therefore supporting the claim that $PM_{2.5}$ is correlated with hypertension.
Brook et al. 2013	Canada (AL)	Adults over 25 years of age who were selected to complete the 1991 long form Census: 2.1 million adults	Ambient $PM_{2.5}$ from 2001 to 2006 – satellite data average 8.7 μg/m^3	Diabetes mortality was found to be positively associated with $PM_{2.5}$ exposure. When adjusted for a 10 μg/m^3 increase in $PM_{2.5}$, there was an increased risk for mortality associated with diabetes.
Chen et al. 2013	Ontario (AL)	62,012 non-diabetics that were followed from 1996 to 2005 – 66% in urban areas	Ambient $PM_{2.5}$ from 2001 to 2006 – satellite data average 10.6 μg/m^3	Long term exposure to $PM_{2.5}$ may contribute to the onset of diabetes in adults.
Crouse et al. 2012	Canada (R and U)	People eligible for the Canadian Census Mortality Study: 2.145 million people (Wilkins et al. 2008) – 73 % in urban areas	Ambient $PM_{2.5}$ from 1987 to 2001 - ground stations average 8.7 μg/m^3 $PM_{2.5}$ – rural/farm = 6.5 μg/m^3 $PM_{2.5}$ – urban = range 7.6 μg/m^3 to 11.1 μg/m^3	Strongest association was found to be with ischemic heart disease, having a Hazard Ratio (a comparison of events between two groups) of 1.31. Positive hazard ratios were also found for non-accidental, cardiovascular, and circulatory causes of death with a 10 μg/m^3 increase in $PM_{2.5}$.

Study Reference	Study Location	Study Participants	Pollutant of Interest	Health Effects/Impacts
Gan et al. 2011	Vancouver, British Columbia (U)	Individuals aged 45 to 85 years in metropolitan Vancouver: 445,868 people	Ambient $PM_{2.5}$ from 1994 to 1999– traffic and black carbon emissions $PM_{2.5}$ average 4.08 μg/m^3	Risk of coronary heart disease mortality increased by 6% when exposed to black carbon, but not $PM_{2.5}$
Elliot and Copes 2011	Rural Interior and Northern British Columbia (R)	Adults 30 years of age and older	Ambient $PM_{2.5}$ from 2005 to 2009– measured from near-by urban stations, average range 3.1 μg/m^3 to 7.4 μg/m^3 annual mean 5-year period	Anthropogenic $PM_{2.5}$ was responsible for 74 deaths per year for all-cause mortality.
Chen et al. 2004	Vancouver, British Columbia (U)	Elderly population (65 years of age and older) between 1995 and 1999	Ambient PM (all size fractions) from 1995 to 1999 – ground level monitors average $PM_{2.5}$ 7.7 μg/m^3	Low-concentrations of PM (PM_{10} = 13.3 μg/m^3, $PM_{2.5}$ = 7.7 μg/m^3) was found to be associated with COPD hospitalizations
<u>*Short-Term Exposure:*</u>				
To et al. 2015b	Ontario (AL)	Individuals (location not specified) who received either primary or secondary medical attention between 2003 and 2010 and resided in Ontario and suffered from: asthma, COPD, diabetes, hypertension, angina, acute myocardial infarction, ischemic heart disease, congestive heart failure, stroke, lung cancer, and non-lung cancers were included	AQHI scale - including $PM_{2.5}$, NO_2, and O_3	Incremental unit increase of the AQHI scale (equivalent increases: NO_2 = 10 ppb, $PM_{2.5}$ = 10 μg/m^3, and O_3 = 10 ppb) resulted in increased outpatient visits by 1% to 5% which equates to 15,000 patients. These increased visits continued for 2-days after the AQHI scale increased.
Szyszkowicz and Kousha 2014	Windsor, Ontario (U)	Emergency room visits between 2004 to 2010 associated with asthma - 6,697 patients in total	Ambient air pollution as determined by the AQHI ($PM_{2.5}$, NO_2, and O_3)	Exposure to ambient air pollution were positively correlated with ER visits for asthma. Younger people were affected on the same day as air quality deteriorated, while older people had a lagged response to the poor air quality.
Goldberg et al. 2006	Montreal, Quebec (U)	Deaths of Montreal residents who were diagnosed with diabetes and also had cardiorespiratory conditions between 1984 to 1993; 2,947 deaths occurred in persons over 65 years of age.	Daily concentrations of $PM_{2.5}$ and PM_{10} and gaseous pollutants (SO_2, NO_2, CO, O_3) were linked back to day of death– ground monitors.	Individuals with diabetes and cardiovascular disease have an increased risk for death when exposed to increased air pollution concentrations that include $PM_{2.5}$.
Jerrett et al. 2004	Hamilton, Ontario (U)	Mortality data from 1985 to 1994 within the city limits	PM as measured by the coefficient of haze (SO_2 also included) - ground monitors within Hamilton city limits	Mortality increases were associated with air pollution exposure city wide, especially areas with lower socioeconomic status
Villeneuve et al. 2003	Vancouver, Canada (U)	550,000 individuals between 1986 and 1999 and mortality	PM_{10} and $PM_{2.5}$ and gaseous pollutants (O_3, CO, SO_2, and NO_2) – daily continuous ground sampling	PM_{10} was associated with increased cardiovascular mortality, but $PM_{2.5}$ was not found to be correlated as a predictor of mortality

Table 3. Summary of Canadian monitoring station $PM_{2.5}$ concentrations – 2013 (ECCC 2014).

Region	Land Type	Peak $PM_{2.5}$ (µg/m^3)	Average $PM_{2.5}$	No. Stations (total)	No. Stations with Recorded Data	SD for average $PM_{2.5}$ (µg/m^3)
Summary	Rural agricultural areas	18.34	6.68	31	13	2.05
	Rural but not agricultural areas	15.36	5.26	61	20	1.18
	Urban areas	17.78	6.93	293	133	1.90
Alberta	Rural agricultural areas	14.48	5.18	12	5	2.16
	Rural but not agricultural areas	13.80	4.98	11	3	0.98
	Urban areas	22.29	7.66	34	20	1.83
British Columbia	Rural agricultural areas	N/A		1	0	
	Rural but not agricultural areas	11.60	5.20	6	1	N/A
	Urban areas	16.11	6.04	102	33	1.91
Manitoba	Rural agricultural areas	N/A		0	0	
	Rural but not agricultural areas	N/A		0	0	
	Urban areas	21.84	5.75	5	5	0.94
New Brunswick	Rural agricultural areas	N/A		0	0	
	Rural but not agricultural areas	17.80	7.00	9	1	N/A
	Urban areas	18.81	6.49	9	7	1.45
Newfoundland and Labrador	Rural agricultural areas	N/A		0	0	
	Rural but not agricultural areas	N/A		0	0	
	Urban areas	15.75	5.58	8	4	0.71
Northwest Territories	Rural agricultural areas	N/A		0	0	
	Rural but not agricultural areas	N/A		1	0	
	Urban areas	22.30	4.50	4	2	2.69
Nova Scotia	Rural agricultural areas	N/A		1	0	
	Rural but not agricultural areas	16.10	5.10	4	1	N/A
	Urban areas	16.83	6.45	6	3	0.21
Nunavut	Rural agricultural areas	N/A		0	0	
	Rural but not agricultural areas	N/A		1	0	
	Urban areas	N/A		0	0	
Ontario	Rural agricultural areas	19.30	7.90	4	1	N/A
	Rural but not agricultural areas	17.90	6.23	13	3	1.27
	Urban areas	18.09	7.15	69	35	1.19
Prince Edward Island	Rural agricultural areas	N/A		0	0	
	Rural but not agricultural areas	N/A		0	0	
	Urban areas	N/A		0	0	
Quebec	Rural agricultural areas	20.96	7.59	12	7	1.49
	Rural but not agricultural areas	14.95	4.81	12	11	1.20
	Urban areas	25.06	9.98	43	20	1.67
Saskatchewan	Rural agricultural areas	N/A		1	0	
	Rural but not agricultural areas	N/A		0	0	
	Urban areas	15.63	6.33	4	3	0.13
Yukon Territories	Rural agricultural areas	N/A		0	0	
	Rural but not agricultural areas	N/A		0	0	
	Urban areas	19.90	6.20	1	1	N/A

Table 4. APEI agricultural and rural emissions data (APEI 2019).

Emission Source	PM_{10} (tonnes)	$PM_{2.5}$ (tonnes)	Ammonia (tonnes)
	Agriculture and Rural Sources		
Animal Production	1.00×10^4	2.08×10^3	2.86×10^5
Crop Production	1.58×10^6	3.78×10^5	1.59×10^5
Fuel Use	629	377	43
Total Ag. and Rural	1.59×10^6	3.80×10^5	4.46×10^5
	Other (Rural) Sources		
Dust Unpaved Roads	2.40×10^6	3.55×10^5	-
Off-Road Diesel Vehicles	1.09×10^4	1.07×10^4	192
Off-Road Gasoline Vehicles	3983	3743	90
Total Other Sources	2.41×10^6	3.69×10^5	282
Total Agriculture, Rural, and Other Sources	4.00×10^6	7.49×10^5	4.46×10^5

amounts of the three pollutants combine to demonstrate the full picture of PM and ammonia emission potential from rural Canada. If agriculture and rural PM emissions were re-defined to include these "other" contributions, such as the transportation sector, the total contribution to $PM_{2.5}$ in 2017 would equate to 7.49×10^5 tonnes making the contribution of the agriculture and rural sector in Canada significantly larger. It was further noted that the burning of agricultural wastes is not yet included in the APEI reporting system and is therefore not reflected in the emission summary (APEI 2019).

The sources of $PM_{2.5}$ listed above in Table 4, are determined based on in-house estimates by the ECCC using emission factors and data from reputable sources. Estimates for PM are based off the research of Seedorf (2004), Takai et al. (1998), Van Heyst (2005), and Van Heyst and Roumeliotis (2007). Seedorf (2004) provides emission factors for PM based on the measurement of bioaerosols, while Takai et al. (1998) provides emission rates of respirable and inhalable dust measured through gravimetric analysis of filters. Both studies provided data gathered from inside several housing facilities of different animal species (beef, dairy, swine, poultry) located in Europe. The studies by Van Heyst (2005) and Van Heyst and Roumeliotis (2007) do provide estimates of secondary $PM_{2.5}$ emissions data, but only for a small sampling of poultry facilities in Ontario, Canada. PM data used for the General Inventory of animal production are based on Pattey and Qiu Guowang (2012) and Pattey et al. (2015) and are based on primary PM emissions.

Secondary $PM_{2.5}$ emissions in Canada are not yet clearly specified within the national inventory as emissions are typically calculated based on emission factors which, as outlined above, do not clearly outline the contribution from SIA formation for animal species other than poultry. While $PM_{2.5}$ data gathered by monitoring stations is not restrictive of SIA, it is important to understand what species of $PM_{2.5}$ are being measured and what sources they are being emitted from. For example, the formation of SIA, driven by ammonia precursor gas, would indicate an agricultural/rural source of $PM_{2.5}$. Knowing this would then allow researchers to determine what impact agricultural and rural emissions have on the concentrations measured in urban areas, and then subsequently understand how it is impacting human health. Without knowing the fractionation of $PM_{2.5}$, a true reflection of the impacts based on source is not well known and mitigation measures may be misallocated.

Ammonia's conversion to SIA is complex and varies based on environmental conditions (season, time of day, and environmental conditions) occurring at and in the vicinity of the source of the ammonia emissions (Roumeliotis et al. 2010). Few works (Aneja et al. 2006;

Table 5. Secondary $PM_{2.5}$ from ammonia – Referencing data presented in Table 4.

Emission Source	Reported $PM_{2.5}$ (tonnes)	Reported Ammonia (tonnes)	7% Ammonia to Secondary $PM_{2.5}$ (tonnes)	11% Ammonia to Secondary $PM_{2.5}$ (tonnes)	New Estimated $PM_{2.5}$ Total (7% - 11% conversion) (tonnes)
			Agriculture and Rural Sources		
Animal Production	2.08×10^3	2.86×10^5	2.00×10^4	3.15×10^4	$2.21 \times 10^4 – 3.36 \times 10^4$
Crop Production	3.78×10^5	1.59×10^5	1.12×10^4	1.75 x 10v	$3.89 \times 10^5 – 3.95 \times 10^5$
Fuel Use	377	43	3.01	4.72	380 - 382
			Other (Rural) Sources		
Dust Unpaved Roads	3.55×10^5	-	-	-	-
Off-Road Diesel Vehicles	1.07×10^4	192	14	21	1.07×10^4
Off-Road Gasoline Vehicles	3743	90	6	10	3749 - 3753
Total	7.49×10^5	4.46×10^5	3.12×10^4	4.91×10^4	$4.26 \times 10^5 – 4.44 \times 10^5$

Behera and Sharma 2010) report the percent conversions of ammonium to $PM_{2.5}$, due in part to the confounding conditions. Aneja et al. (2006) reported, based on samples collected in rural North Carolina, that 7 % to 11 % of ammonium was converted to $PM_{2.5}$ due to the neutralization reaction, and Behera and Sharma (2010) stated that these reactions could account for upwards of 30% of the mass of $PM_{2.5}$. This information, although limited, indicates that precursor gases, such as ammonia when considering the agricultural sector, are responsible for a portion of the measured $PM_{2.5}$ quantities. For demonstration purposes, the SIA $PM_{2.5}$ burden that could be attributed to agriculture is conservatively estimated in Table 5 below based on the reported conversion percentages from Aneja et al. (2006) and the amount of ammonia reported in Canada for 2017 (APEI 2019). The conversion presented here is based solely on the findings from Aneja et al. (2006) who found of the total $PM_{2.5}$ generated in a poultry house, 7% to 11% of it was due to SIA formation.

Based on the masses estimated above in Table 5, ammonia would account for approximately 3.12×10^4 to 4.91×10^4 tonnes of the total $PM_{2.5}$ burden annually in Canada from agriculture and rurally sources. While these contributions are variable, the reality is that while ammonia emissions from agriculture and rural sources in Canada continues to increase (APEI 2019), it will also continue to fuel the formation of SIA's, contributing to the overall $PM_{2.5}$ burden and continue to exacerbate the already increasing breadth of health impacts suffered nationally (and globally) due to fine particulate matter.

Health impacts from agricultural practices

Globally, air pollution is a significant health risk for both humans and animals (Prüss-Ustüm et al. 2016; WHO 2013). However, the impact that PM generated by agricultural practices has on human health is one that is being explored more recently. As there is a significant source of particulate matter emitted from Canadian agriculture, a study by Khan (2015) looked at the occurrence of respiratory disease leading to hospital admissions in the Prairie Provinces (Alberta, Saskatchewan, and Manitoba) due to $PM_{2.5}$. The statistical modelling performed by Khan (2015) outlined that there is a correlation with hospital admissions for respiratory disease and $PM_{2.5}$ emissions during 2009 and 2010.

Similar studies have been conducted in the United States using long term health cohort information. Weichenthal et al. (2014) looked at the health information presented in the US Agriculture Health Study Cohort. The 83,378 individuals enrolled in this cohort were farmers, commercial pesticide applicators, and their families from Iowa and North Carolina. This study is unique in that it focused strictly on individuals working in the agricultural sector but, like the studies discussed above, the results were similar. Specifically, a positive correlation between cardiovascular deaths and men was found as compared to their female counterparts only because they tended to work significantly longer hours outdoors. While not a Canadian specific study, agricultural practices and population demographics are similar between Canada and the United States.

CONCLUSIONS

To date, the major focus of $PM_{2.5}$ research in Canada has been conducted using data from urban dwelling populations and $PM_{2.5}$ data with few studies looking into the sources and health impacts occurring in rural/agricultural areas. The formation of SIA's, a known contributor to $PM_{2.5}$, is fueled in part by ammonia emissions that are linked to animal and crop production, and a contributor to the overall $PM_{2.5}$ burden. However, a current lack of understanding with respect to the source apportionment and fractionation of rural $PM_{2.5}$, restricts the understanding of the full impact of this pollutant on human health.

Across the globe, health impacts from air pollution and particulate matter are felt in a variety of ways. By 2050, premature death caused by air pollution is expected to double from current values (Lelieveld et al. 2015). Due to the lack of monitoring programs in place (WHO 2013), specifically in rural areas, it is still difficult to properly quantify emissions of fine particulates thus making it difficult to predict health impacts (Lelieveld et al. 2015) or long-term trends in data (Stieb et al. 2015). To further complicate this, the speciation between primary and secondary $PM_{2.5}$, has yet to be directly quantified in Canadian reporting systems. Given the surmounting long-term health impacts and increasing deaths linked to $PM_{2.5}$ and the significant, yet less understood quantification of rural based $PM_{2.5}$, gaining a deeper understanding of the impact of rural emission sources should be explored more holistically.

Discovered throughout this paper was that current $PM_{2.5}$ monitoring taking place in Canadian rural areas is significantly less as compared to urban. What is less understood is the fractionation of $PM_{2.5}$ that is directly attributed to agricultural production in these rural areas. Current $PM_{2.5}$ data is based on in-house estimates and emission factors that do not distinguish the contribution of secondary $PM_{2.5}$ to the overall $PM_{2.5}$ quantity. Knowing this will help build our understanding of the contribution that agricultural $PM_{2.5}$ has on the overall PM burden and, ultimately, the impact on human health.

ACKNOWLEDGMENT

The authors would like to acknowledge the supporters of this research: Ontario Ministry of Agriculture, Food and Rural Affairs (OMAFRA), the Canadian Poultry Research Council (CPRC), and the University of Guelph's School of Engineering.

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