QUANTIFYING FUGITIVE DUST EMISSIONS FROM LIMESTONE QUARRIES: DATA SELECTION AND UNCERTAINTY ASSESSMENT

by

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Abstract

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Accurate quantification of fugitive dust emissions from quarries helps maintain the integrity of the National Pollutant Release Inventory. Emissions from unpaved roads, material handling, and storage piles at three quarries were calculated using the AP-42 emissions factor method, and the error of using 30-year climate averages, the uncertainty introduced by typical material properties, and the limited availability of climate data were addressed. Using daily and hourly data predicted unpaved roads emissions between 38.95% and 42.50% higher, materials handling emissions 15.31% lower to 18.64% higher, and storage pile emissions 12.48% to 37.50% lower than calculations using 30-year averages. Employing Monte Carlo simulation, the confidence intervals attributable to typical material properties ranged from 87.50% below to 650% above the mean. Krige-interpolated climate data showed potential for being more accurate than observations at the nearest station. Using site-specific, temporally-relevant data and assessing uncertainty promotes calculations that better match the goals of the inventory.

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Lastly, I would like to thank friends and family who often accused me of working too hard, and my girlfriend who often accused me of slacking, for their continuous support and for pretending to be interested in my dry research area.

Dedication

This thesis is dedicated to the recession that began in the summer of 2008 and is continuing into the summer of 2009. Although the world is calling this a "time of economic uncertainty", I have yet to see any valid uncertainty assessment.

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1 Introduction

The creation of Canada's National Pollutant Release Inventory (NPRI) in 1992 reflected a change in the standards expected of Canadian industry. Rather than accepting industry's veil of secrecy, Canadians demanded to know which harmful pollutants were being emitted, by whom, and where emissions were concentrated. While this new scrutiny grew from the American "Right-to-Know" movement, the NPRI tackles fundamental issues, rather than the fear of catastrophic releases of toxic chemicals. The NPRI, by focusing on toxic pollutants emitted during *status quo* industrial operations, reflects the Canadian public's desire to know which chemicals enter their soil, water, and air as by-products of industrial operations.

Despite the public's growing need to know, surface mines and quarries were exempt from preparing an emissions inventory for the NPRI and thus their emissions went unreported to the public (Environment Canada, 2006). It is unclear why the exemption was established, but it was likely a combination of industry lobbying, and substantive issues including the difficulty of characterizing certain emissions sources at mines. But with the addition of the seven Criteria Air Contaminants (CACs) – including particulate matter (PM) - to the list of reportable substances in 2001, this exemption was reconsidered (Environment Canada, 2006). Because surface mines and quarries generate a large quantity of PM, keeping this exemption in place would have created inconsistencies in reporting requirements for the many diverse sectors of the Canadian economy. In 2005, Environment Canada hosted a multi-stakeholder workshop to revisit the mining

exemption, and it was ultimately removed for the 2007 reporting year (Environment Canada, 2006).

Particulate matter (PM) is the dominant airborne pollutant from surface mines and a substance notorious for its health consequences (Inyang and Bae, 2006, Kon, *et al.*, 2007). The true danger of PM is that substances stakeholders believe to be harmless – like the limestone extracted from quarries – can cause injury and death to humans, wildlife, and plants when in particulate form (Salvi and Holgate, 1999). For this reason, accurate quantification of emissions from surface mines and quarries is essential for maintaining the integrity of the NPRI and ultimately for building a trusting relationship with the local community. As summarized by Swart *et al.* (2007), policy-oriented data compilation like an emissions inventory is typically an exercise in fulfilling procedural requirements rather than scientifically valid quantification of the emissions values. Scientific validity, in this sense, means using an approach that will lead to accurate emissions reports, which, as will be exposed throughout the course of this study, is something that the current NPRI pits and quarries guidance does not stress.

1.1 Problem Statements

Environment Canada has published a guidance document for quantifying PM emissions from pits and quarries as part of its NPRI emissions inventory, based on accepted AP-42 predictive emissions factor equations developed by the United States Environmental Protection Agency (USEPA) (Environment Canada, 2008). Purposefully simple and commonly used, these AP-42 equations use meteorological data and material properties to estimate the amount of particulate matter emitted as a result of each quarry activity.

The quality of these equations is questionable, and USEPA admits (USEPA[3], 1995) that their completeness and detail is limited by the published references used to develop them. Even still, the USEPA and Environment Canada consider them to be appropriate for inventories and permit applications across a range of industries, as surface mines are just one of the hundreds of sectors that rely on AP-42 emissions factors for these purposes.

The emissions factors used in the NPRI pits and quarries guidance are taken from Chapter 11, "Mineral Products Industry", meaning that this study is directly relevant to other surface mines including coal, asbestos, and diamond mines, and to other related industries including cement manufacture, phosphate rock processing, and glass fibre manufacture.

1.1.1 Temporal Resolution of Climate Data

Climate averages from the Canadian Climate Normals Online website are typically inputted to the AP-42 emissions factor equations, which notably ignores annual, seasonal, daily, and diurnal fluctuations in meteorological variables (Countess, n.d.). These normal values have been averaged between 1971 and 2000, which too ignores any effects related to climate variability or change. Choi and Fernando (2008) state that dust models that do not consider episodic, high wind periods perform poorly, and Neuman *et al.* (2009) suggest that temporal variations are inadequately considered in calculations using emissions factors, and that intermittent releases should not be ignored. Accordingly, some studies have used field meteorological measurements in calculating emissions, but they too have only sampled a few days of data which cannot be extrapolated to calculate yearlong emissions totals.

The first goal of this research was to develop a modification to the established emissions quantification methodology to allow for the use of temporally relevant daily and hourly climate data, and then compare the results of the new calculations to the current method based on 30-year average climate data.

1.1.2 Uncertainty in Material Properties

Average values from published documents are also used to represent material properties like silt and moisture contents, rather than conducting any on-site sampling to get true values. According to Neuman *et al.* (2009), these average values may bear little or no resemblance to the materials on site and result in over- or under-estimations of emissions by several orders of magnitude. Simon *et al.* (2008) assert that the silt contents of unpaved roads listed in AP-42 – the main resource for NPRI pits and quarry emissions calculations - are not well characterized and are not suitable for use in many areas of the United States. Both climate data and material property oversights have the potential for introducing considerable inaccuracy and uncertainty to the calculated quantities of PM emitted from pits and quarries.

Countess (n.d.) states that many emissions inventories have been inaccurate in the past due to use of inappropriate emissions factors. Additionally, both Countess (n.d.) and Frey (1997) suggest that uncertainty in activity rates are an additional source of inaccuracy. While there has been research to characterize the uncertainty in emissions inventories, no publications have been uncovered to provide a solution that is more accurate and which does not involve time-consuming or expensive dust sampling.

Houyoux *et al.* (2003) describe uncertainty as unavoidable in emissions inventories, and that best practice is to describe it quantitatively using statistical techniques. The Nobel prize-winning Intergovernmental Panel on Climate Change (IPCC) has advocated the use of Monte Carlo methods for quantifying the uncertainty of carbon emissions inventories, an opinion reflected by its frequent application in predictions of global climate change (Intergovernmental Panel on Climate Change, 2000). Frey (1997) and Sax and Isakov (2003) have used Monte Carlo methods for assessing the uncertainty of emissions from smaller-scale sources: highway vehicle emissions and local air quality, respectively. Other authors have used, scrutinized and verified the USEPA emissions factor equations for pits and quarries, yet they have all used the equations deterministically, to generate a point estimate of particulate matter emissions (Veranth *et al.*, 2003; Ghose, 2007). According to Gardner and O'Neill (1979), representing such complex, variable, and uncertain systems with a deterministic estimation of PM emissions is mathematically incorrect. This sentiment is echoed by Watson *et al.* (2000), who state that fugitive dust estimates have a high amount of variability, attributable more to systematic than random error.

The second goal of this research is therefore to estimate emissions on a probabilistic basis using Monte Carlo simulation which will be an indication of the uncertainty introduced by use of average material properties.

1.1.3 Spatial Data Availability

A third problem relates to the availability of climate data. The NPRI pits and quarries guidance explains the use of the Canadian Climate Normals On-line website to collect

average wind speeds and precipitation values, yet many of the quarries are located far from an Environment Canada weather station. In cases where a site is located near a weather station, many of the stations do not report the average wind speed values needed for calculations.

The problem of data availability can be approached using kriging, a geographic spatial interpolation technique once used by Nalder and Wein (1998) to interpolate climate data within the Canadian Boreal forest. With the use of Geographic Information Systems, kriging has become readily available. Its appropriateness for NPRI applications, however, has not been reported.

2 Fugitive Dust in the Context of the NPRI

The Right-to-Know movement has its origins with the modern conservation movement in 1952 when a thick smog settled over London, England, killing an estimated 12 000 people and sickening 100 000 more. Bell *et al.* (2004) call the London smog one of the most important air pollution events in history, as the link between air pollution and human became engrained in the public's awareness, and was met with significant environmental regulation and research efforts. By 1984, a pair of catastrophic incidents involving pesticide manufacturer Union Carbide strengthened the resolve of right-to-know activists (Jobe 1999; Jackson, 2000). The first of these incidents was the catastrophic release of methyl isocyanate from its plant in Bhopal, India that resulted in over 2000 deaths (Jobe 1999; Jackson, 2000). This was an important event, but it was not until 100 West Virginians were hospitalized by fumes from a Union Carbide plant that the North Americans began asserting that citizens have the right-to-know about toxic substances used by private industry in their communities that may harm their health or the environment (Jobe, 1999; Stephan, 2002). In 1986, as Right-to-Know groups were becoming more organized and vocal, the US congress took action and established the Toxics Release Inventory (TRI) which formally recognized this right (Howes, 2001). The establishment of this publicly-accessible pollution database forced accountability upon firms, and empowered citizens by turning costly or legally-restricted private information into a public good (Stephan, 2002). Canada soon followed suit with the establishment of the National Pollutant Release Inventory (NPRI) in the early 1990s, with the first reports published in 1995 (Jackson, 2000).

Forcing firms to report to a publicly-accessible database about their environmental performance, as both the NPRI and TRI do, applies both political and economic pressure on industry as a whole to strive for cleaner production or to evade reporting of substance emissions which firms have, in some cases, managed to do (Howes, 2001). It is worth noting, however, that neither inventory bans the use or production of any substances, nor attaches fines for threshold exceedances, making the only true financial threat that from poor public perception (Howes, 2001).

2.1 Successes and Failures of Pollutant Inventories

Although there is a large gap in the published literature about the effectiveness of the NPRI, the TRI from which it was modelled is considered one of the most successful pieces of environmental legislation in the history of the United States (Fung and O'Rourke, 2000). However, Howes' (2001) study comparing the Australian National Pollutant Inventory suggests that the both inventories have had noticeable industry resistance, and problems with the accuracy of reported emissions. He suggests that industry's reluctance to participate is primarily due to the resources needed to tabulate emissions (Howes, 2001).

In contrast, some firms are eager to cooperate, and use their annual emissions reports as an opportunity to find inefficiencies – particularly wasted raw materials (Howes, 2001). Gottlieb *et al.* (1995), as cited in Howes (2001), claim that the reduction of the inefficiencies discovered in emissions inventories can lead to savings that offset compliance costs. Despite the reluctance of industry, Fung and O'Rourke (2000) maintain that the TRI has been more successful in reducing toxic pollution than any other

piece of American legislation, and at only a fraction of the cost to government. Because of this success, public disclosure of environmental information is touted by some as a valuable replacement for traditional regulation (Stephan, 2002).

2.2 Accuracy of Emissions Reports

Like the TRI, the data collected and published by the NPRI is self-reported, and receives only limited quality control. In fact, only 3% of TRI reports are ever scrutinized meaning that companies may choose to under-report their emissions (Fung and O'Rourke, 2000). Similar figures are unavailable for the NPRI, but the similarities between the two programs suggest that few Canadian reports ever get scrutinized. Under-reporting is a significant problem for the TRI, largely related to poor enforcement practices (Wolf, 1996). The EPA discovered that nearly 30% of all companies required to report to the TRI fail to do so; mostly small- and medium-sized businesses that are unaware of the requirement to report (Wolf, 1995). Although limited by a lack of published statistics for the NPRI, it would not be unexpected to discover that many businesses fail to submit an inventory.

2.3 Quarries in the NPRI

A search of the NPRI database shows that several quarries across Canada had been reporting since 2002 – even while they were exempt from the program. This proactive reporting was likely prompted by their visibility in the community and to avoid the shame and fear associated with bad environmental performance, as explained by Stephan (2002). Yet, questions must be raised about which emission sources were quantified, and how values were calculated, as the NPRI pits and quarries guidance for reporting was first published in 2007. The particulate matter (PM) emissions values in the 2002 reports

are fairly low compared to 2007 values; however it must be considered that 2007 was the first year that fugitive dust from unpaved roads – a significant source of PM - had to be calculated for all industrial sites (Environment Canada, 2007; Ghose, 2007).

2.4 Fugitive Dust Defined

Under the framework of the NPRI, PM is a solid or liquid that remains suspended in the air for any length of time (Environment Canada, 2007). Two classes of PM exist: primary and secondary. Primary PM has been directly emitted to air, like mineral and soil particles generated during quarry processes. Secondary PM results from chemical reactions in the atmosphere; common reactions involving nitrates and sulphates (Reed, 2003). The NPRI does not distinguish between the two categories, however Chang's (2004) study of the chemical composition of PM from a quarry reports that particulates contained no nitrogen or sulphur, suggesting that the majority of PM emissions were primary.

Dust is defined by the Western Regional Air Partnership (WRAP) as "particulate matter of a geologic, organic or synthetic origin that is, or has the potential to be, suspended in the atmosphere as a result of mechanical abrasion, wind erosion, or explosive activities" (Western Regional Air Partnership, 2006). NPRI reports require the division of particulate matter emissions into two categories: point-source and fugitive. Point-source dust emissions are those having a defined flow from a specific location, for example from a stack or chimney (Abdul-Wahab, 2005; Ghose, 2007). Contrasting these are fugitive dust emissions that encompass the remaining emissions from other sources that "could not reasonably pass through a stack, chimney, vent, or other functionally equivalent

opening" (Abdul-Wahab, 2005; WRAP, 2006). This definition which echoes that of non-point or distributed emissions applies to the majority of quarry activities as will be detailed in Chapter 3. In the literature, the terms fugitive dust and particulate matter are often used interchangeably. Accordingly, the same convention is applied to this report despite the important distinctions.

2.5 NPRI Reporting Thresholds

The particle size of the emitted dust is of far greater regulatory importance than its classification as fugitive or point-source since an inverse relationship between particle size and toxicity has been reported (Salvi and Holgate, 1999). Three size fractions of PM are individually reportable to the NPRI after threshold emissions have been surpassed:

1 TPM: Total particulate matter with an aerodynamic diameter less than 100 microns (μm)
2 PM_{10}: Particulate matter with an aerodynamic diameter less than or equal to 10 μm
3 $PM_{2.5}$: Particulate matter with an aerodynamic diameter less than or equal to 2.5 μm (Environment Canada, 2007)

The reporting thresholds are strikingly different for each size fraction which is not explained by Environment Canada, but is likely related to the differences in potential health effects. Emissions of PM_{10} and $PM_{2.5}$ are reportable if annual emissions exceed 0.5 tonnes (T) and 0.3 T respectively; much lower than the 20 T threshold for TPM (EC, 2007). It is worth noting that the aerodynamic diameter of a particle is used, instead of

the true geometric diameter of the particle, due to difficulties in establishing this value for PM_{10} or $PM_{2.5}$ using traditional methods (Watson *et al.*, 2000).

Although health effects were likely the main influence in setting the emission thresholds, the NPRI documentation does not explain how they were established. One reason may be that the majority of the mass exists in the TPM fraction, but that the greatest numbers of particles are in the smaller PM_{10} and $PM_{2.5}$ ranges (Salvi and Holgate, 1999). PM_{10} and $PM_{2.5}$ are the focus of regulation because they have the potential for entering deep into the lungs and resulting in serious damage (Godoi *et al., 2008).*

2.6 Mechanism of Health Impacts

Salvi and Holgate (1999) reviewed over 60 studies from around the world that linked particulate matter in ambient air to health effects such as asthma, heart disease, impaired lung function and other respiratory illness. Recently, the focus has shifted to ultrafine particles in urban airsheds, where increased hospitalization has been correlated with elevated ambient concentrations of $PM_{2.5}$ (Godoi *et al.*, 2008). There is a direct correlation between the size of particulate matter and health impacts; smaller particles pose a significantly higher risk than larger ones, regardless of composition (Natusch and Wallace, 1974). A brief description of how particulate matter affects the respiratory system assists in this discussion.

On a typical day, the human respiratory system is exposed to approximately 10 000 to 20 000 litres of ambient air containing particulate matter of varying sizes (Salvi and Holgate, 1999). Air enters the body through the nasopharyngeal region of respiratory system;

through the nose or mouth of exposed individuals towards the tracheobronchial region via the pharynx and larynx (Natusch and Wallace, 1974; Reed, 2003). The tracheobronchial region, reaching from the trachea to the terminal bronchioles, is where the majority of large particles – diameter greater than 10 um – are deposited (Salvi and Holgate, 1999; Reed, 2003). Deposition is enhanced at bifurcations, or branching points, in the bronchioles which may explain why many lung diseases occur at these locations (Natusch and Wallace, 1974). In healthy lungs, however, low doses of small particles do not pose a problem as they are captured in the trachea or bronchi, are removed by mucociliary clearance into the throat (Salvi and Holgate, 1999).

Damage to the alveolar region is most severe as this is where oxygen absorption and exchange occurs, but only the $PM_{2.5}$ is fine enough to accumulate in this area (Beckett *et al.*, 1998; Salvi and Holgate, 1999; Reed, 2003). Even though only sufficiently small particles reach the alveolar region, $PM_{2.5}$ is far more toxic than the other size fractions. This phenomenon was observed experimentally as rats exposed to smaller, chemically-identical, particles sustained worse injury than those exposed to an equivalent mass of large particles (Salvi and Holgate, 1999). Salvi and Holgate (1999) explain that as a mass of small particles has a much higher surface area than an equivalent mass of large particles, the particles have a higher capacity to carry the toxic substances and free radicals that damage lung tissue.

Natusch and Wallace (1974) support this claim, suggesting that particles in the $PM_{2.5}$ range tend to host a higher concentration of heavy metal and volatile organic compounds

(VOCs) on their surfaces than PM_{10} and TPM. Their study focused on urban PM from sources like automobile exhausts which are more likely to contain heavy metals and VOCs than the fugitive dust. However, if a quarry is located near a processing plant or other industries that burn coal, trace elements, like mercury, may be available to bind with the fugitive PM (Natusch and Wallace, 1974).

2.7 Evidence of Health Impacts

Inspired by an association between dolomitic lime production and morbidity in a town centred on limestone production, Godoi *et al.* (2008) studied lung deposition of PM in residents of Colombo, Brazil. A simulation model of lung deposition based on Monte Carlo methods was built to probabilistically determine which parts of the respiratory system would be most adversely affected (Godoi *et al.*, 2008). The results showed enhanced alveolar deposition during resting breathing versus light physical activity, which is consistent with the findings of Salvi and Holgate (1999). As expected, elemental analysis of the PM uncovered high concentrations of calcium and carbon, but the authors stated that the high proportion of dolomitic limestone particles (MgO.CaO) in the $PM_{2.5}$ fraction was somewhat unexpected (Godoi, *et al.*, 2008). Godoi *et al.* (2008) also found several noteworthy limestone impurities in the PM, including titanium, chromium, and manganese which can all harm human health (Abdul Wahab, 2005). These impurities are related to local geological conditions, and would not be expected to be present in the PM generated at all quarries. Godoi *et al.* (2008) also found significant concentrations of sulphur and chlorine associated with lime kiln fuels, and elevated concentrations of copper and zinc attributable to wood-burning operations in the area.

Cement plants are another large emitter of PM as dust is generated during every stage of the process (Abdul-Wahab, 2005). The dust from these plants has caused illness in nearby communities by inhalation, and dermal and eye contact; however, the risk of injury depends on duration and intensity of exposure (Abdul-Wahab, 2005). Abdul-Wahab (2005) stated that cement dusts can contain crystalline silica, gypsum, nickel, cobalt, chromium, and lime which can all be detrimental to human health. Silica dust can cause silicosis and other fatal lung diseases, and chromium is a known carcinogen (Abdul-Wahab, 2005). These latter two substances are unlikely to be found in high concentrations in limestone as these impurities would likely prevent the exploitation of the deposit.

Limestone quarries tend to be more isolated from communities than cement plants; however, the dust still poses occupational risks to workers on site. Thompson and Visser (2001) report that 74% of accidents in South African surface mines occurred during transportation of ore by haul truck, and that dust generation was a significant contributor to many of the incidents. One specialized task in limestone production is disposal of furnace residues which involves dumping lime kiln dust (LKD) from a hopper into the back of a haul truck. These trucks are often completely covered with dust, and Organiscak and Reed (2004) claim that these types of operations are among others in quarries that often exceed respiratory dust standards.

2.8 Cost of Health Impacts

Much research has been devoted to health impacts of PM and other air pollutants, but only recently has this focus shifted to studying the economic consequences of these

health effects. One such study is the definitive "Illness Cost of Air Pollution" (ICAP), published in 2005 by the Ontario Medical Association, where the health effects of smog were quantified in terms of both number of incidents and monetary burden. Although the most recent version does not isolate the impacts from each smog component, the only pollutant considered in the preceding year 2000 version was PM_{10}, and it was concluded that it alone caused 1900 premature deaths that year (Ontario Medical Association, 2005). The 2005 version of ICAP included ozone, sulphur dioxide, nitrogen dioxide, carbon monoxide and $PM_{2.5}$ in the model, which increased the number of premature deaths attributable to smog by 4 000 (Ontario Medical Association, 2005). Besides the approximately 5 900 premature deaths reported in 2005, the OMA (2005) estimates that nearly 17 000 hospital admissions, 60 000 emergency room visits, and 29 million minor illnesses were caused by smog. An Iranian study about the illness cost of air pollution in Tehran showed lower numbers of deaths and hospital admissions, although the methodology and assumptions were substantially different (Karimzadegan *et al.*, 2008). Unlike the OMA study, the annual number of deaths attributable to each smog component was determined using hospital records; PM_{10} was blamed for 786 of the total. Tehran is considered one of the most polluted urban areas in the world, and the population in the Greater Tehran Area is slightly greater than that of Ontario. The number of deaths and hospitalizations attributable to air pollution might therefore be expected to be higher in Iran if the same methods were used in both studies.

The number of medical incidents caused by air pollution is shocking in both studies, but when the economic value of lost productivity, health care costs, pain and suffering, and

loss-of-life are estimated, the costs of air pollution become truly staggering. Lost productivity in Ontario during 2005 – the time lost for treatment and recovery of illnesses induced by smog – was estimated have cost nearly $375 million (Ontario Medical Association, 2005). The OMA (2005) estimates that $507 million was spent treating these afflicted individuals, and that the value of pain and suffering of these individuals is $537 million. It should be noted that putting a value on pain and suffering was estimated by using established models that assess how much an individual would be willing to pay to avoid the illness, which has inherent limitations according to Karimzadegan *et al.* (2008).

2.9 Environmental Impacts

Besides health effects, the inclusion of PM in the NPRI also reflects the environmental harm that the emissions can cause. According to Lodge *et al.* (1981), the most prevalent impact of PM emissions – even more common than the health effects – is reduced visibility due to absorption and scattering of visible light. In this case, visibility relates to the haze seen over cities during smog days, rather than the reduced visibility for drivers on unpaved roads, as discussed by Mohamed and El Bassouni (2007) and Thompson and Visser (2001). Another problematic impact, specifically near quarries, is dust loading on vegetation that block stomata and inhibit gas exchange (Lodge *et al.*, 1981; Beckett *et al.*, 1998). Beckett *et al.* (1998) explain that the phytotoxic effects of heavy metals bound to PM, can disrupt physiological processes, alter the genetic structure, and ultimately make trees more vulnerable to pathogenic infection.

The most noticeable local impacts, like visibility and noxious odours, are periodic, aesthetic inconveniences; Drew *et al.* (2002) go as far as calling these complaints ironic as the communities who host quarries would never have developed without the existence of the quarry. However, paleoclimatic researchers have also connected atmospheric dust to long-term global phenomena, such as glacial periods and climate change (Miller and Tegen, 1998; Arimoto, 2001). Atmospheric dust may not be just a response to climate change, but also a cause through the chemical and physical changes it makes to the atmosphere (Arimoto, 2001). In fact, climate change cannot be explained solely by the radiative forcing of atmospheric CO_2, suggest Miller and Tegen (1998), and that other forcing agents – like dust – are contributors.

Polar ice cores from past Ice Ages have shown 2 to 20 times greater dust deposition rates during glacial periods, but the degree to which dust is responsible for climate change is uncertain (Harrison *et al.*, 2007). Dust in the atmosphere influences the surface temperature both by reflecting incoming solar radiation, and absorbing outgoing terrestrial radiation (Miller and Tegen, 1998; Harrison *et al.*, 2001; Mohamed and El Bassouni, 2007). Because the temperature decrease caused by reflected solar energy is offset by the increase caused by trapped terrestrial radiation, dust-induced radiative forcing is considered by Miller and Tegen (1998) to be marginal.

The particle size distribution, concentration, vertical distribution, and mineralogy of the dust determine whether there will be a heating or cooling effect, as does the albedo and the temperature of the underlying surface material (Harrison *et al.*, 2001, Mohamed and

El Bassouni, 2007). As expected, there is limited agreement about how, exactly, PM emissions are influencing anthropogenic global climate change. As discussed by Harrison *et al.* (2001), some studies suggest that atmospheric dust produces negative forcing, resulting in cooling between 1 and 3°C, while others conclude that it contributed to the positive forcing that melted the glaciers of the last Ice Age.

The dust emitted from a single quarry is insignificant on the scale of climate change, but roughly half of the worldwide atmospheric dust has resulted from anthropogenic activities, which justifies accurate quantification in emissions inventories like the NPRI (Miller and Tegen, 1998). In the United States, about 90% of all fugitive dust emissions originate from agricultural sources, unpaved roads, industrial processes, and surface mines (Watson *et al.*, 2000). The following chapter outlines the lime industry and the mechanisms of fugitive dust generation at limestone quarries.

3 Fugitive Dust in the Lime Industry

Natural aggregates, including sand, gravel, and stone, are the most valuable nonfuel minerals in the world according to Drew *et al.* (2002), and are an important part of the Canadian economy. Current social attitudes promote resource conservation and some advocates would insist that natural resource extraction can and should be slowed. However, unlike energy production where alternative fuels exist, ending aggregate extraction would prove infinitely more difficult as few substitutes for basic aggregate materials, such as concrete, exist. Drew *et al.* (2002) provide a vivid account of the importance of aggregates in sustaining the economies of the developed world, and expanding those of developing nations. The infrastructure on which these economies depend, including roads, bridges, dams, airports, and locks, are 85% (by volume) natural aggregate – dominated by Portland cement, of which lime is a key ingredient. This chapter focuses on the lime industry, but many of the fugitive dust generation mechanisms and mitigation techniques are relevant to other surface mining settings.

3.1 Lime Industry

The lime industry produces several products, each with vastly different properties, and many uses, but with similar chemical formulae depending on the geochemistry of the limestone deposit. High-calcium quicklime (CaO) is produced from calcium-rich limestone, and dolomitic quicklime (CaO.MgO) is produced from magnesium-rich dolomitic limestone found in deposits scattered around the world (United States Environmental Protection Agency[1], 1995). Other hydrated lime products and production by-products are also commercially available.

According to the National Lime Association, the single largest use of lime is in steel production for the removal of impurities such as silica, phosphorus and sulphur (National Lime Association, n.d.). Lime products (hereafter called "lime") are also used to treat mining wastewater, to remove sulphates from industrial emissions, and as a soil amendment (National Lime Association, n.d.). Each of these uses requires that the lime meets purity specifications, specifically that they have high carbonate content and few impurities such as heavy metals (United States Environmental Protection Agency[1], 1995; Tang, 2002). Consequently, only a small fraction of limestone deposits is appropriate for extraction and conversion to lime (United States Environmental Protection Agency[1], 1995). When limestone is not sufficiently pure, it is used mainly as construction stone to stabilize roadbeds, airfields and dams (National Lime Association, n.d.).

3.2 Lime Quarrying and Processing

Production of lime begins with the physical extraction limestone or dolostone from a deposit using surface mining techniques. It begins by clearing overburden using a grader or bulldozer. With the deposit exposed, blasting takes place to loosen the stone (USEPA[2], 1995). Blasting involves drilling dozens of boreholes, packing each one with several kilograms of explosives, and detonating the rock face. The most commonly used explosive in quarries is a mixture of ammonium nitrate and fuel oil, a granular solid called ANFO; other less common explosives include pure ammonium nitrate, and gelatine dynamite (Donoghue, 1998; Environment Canada, 2008). After blasting, the limestone is loaded into large haul trucks using front-end loaders and is transported to the processing plant which is often – but not necessarily - located on the same property (USEPA[2], 1995). It is common practice to use waste rock to refill sections of the

excavation or to create sound or visual barriers around the site, but oftentimes it is sent off-site for other uses.

Each of these quarry operations generates fugitive dust; however, the most significant source is haulage truck traffic on unpaved roads, which represents about 50% of the site's total particulate matter emissions (Ghose, 2007). Material handling operations are also significant, being responsible for approximately 25% of the total (Ghose, 2007).

Raw stone delivered to the processing plant is generally too large for efficient conversion to lime in the kilns, therefore it must first be sent through a series of screens and crushers to increase its surface area (USEPA[2], 1995). When heated in the kiln, carbon dioxide is liberated from the limestone or dolostone which increases the calcium concentration in the finished products. High-calcium lime or dolomitic lime is produced according to the following calcination reactions:

$$CaCO_3 + \text{heat} \rightarrow CO_2 + CaO \text{ (high-calcium lime)}$$

$$CaCO_3.MgCO_3 + \text{heat} \rightarrow CO_2 + CaO.MgO \text{ (dolomitic lime)}$$

(USEPA[1], 1995)

3.3 Unpaved Roads

As the single biggest source of fugitive dust from quarries and other surface mines, particulate matter emissions from traffic on unpaved roads is arguably the most important to assess correctly in NPRI reports (Ghose, 2007). Sanders *et al.* (1997) claim that 34% of all particulate matter in the atmosphere originates from traffic on unpaved roads, and Williams *et al.* (2008) report that up 10 million tonnes of dust originated from this source

in the United States alone. Although the mass emitted is high, dust from this source is predominantly in the coarser PM_{10} and TPM fractions according to Watson *et al.* (2000), but this is contested by Williams *et al.* (2008) who describe the knowledge of particle sizes emitted from unpaved roads as incomplete.

Unpaved roads are typically graded and compacted road beds or are composed mainly of the underlying parent material (William *et al.*, 2008). Dust is generated by the pulverization action of the wheels on road surface materials, with the dust discharge increasing linearly in proportion to the amount of traffic (Western Regional Air Partnership, 2006). Williams *et al.* (2008) add that some uplift is also attributable to air movement in the vehicle's wake.

In addition to a positive correlation with the amount of traffic, Watson *et al.* (2000) cite two studies suggesting that dust emissions are negatively correlated with silt content of the road surface. This is in direct conflict with the formula adopted by the NPRI for calculating emissions from unpaved roads, as it positively correlates emissions with the siltiness of the road. These two studies suggest that roads with higher gravel content will have greater abrasive action and therefore have higher emissions (Watson *et al.*, 2000)

Veranth *et al.* (2003), like others tabulating fugitive dust emissions, have taken field measurements of dust caused by traffic on unpaved roads to measure the horizontal flux – a measure of dust migration. Dust flux is the product of dust concentration multiplied by wind speed, but this calculation is problematic because both concentration and wind

speed are continuous variables that have been measured at discrete sampling points in this study (Veranth *et al.*, 2003). Venkatram (2000), as cited in Veranth *et al.* (2003), states that the time-averaged flux is not equal to the product of average concentration and average wind speed due to temporal and spatial variability, which is an important consideration as the emissions calculation methodology advocated by the NPRI (see Chapter 5) uses average wind speeds. It must, however, be emphasized that the NPRI reports only require the total mass of emissions and not a calculation of the flux.

To combat this difficulty, Veranth *et al.* (2003) used several models to interpolate values for wind speed and concentration at points that were not physically sampled. These authors concluded that a significant quantity of dust suspended from traffic on unpaved roads is redeposited near its origin, posing little risk to nearby communities. Because traffic in quarries is intermittent, the energy needed to lift these particles high into the atmosphere is not constantly applied, making off-site migration problematic only during windy periods (Countess, n.d.).

Organiscak and Reed (2004) conducted a similar study, but in addition to characterising the concentrations and dispersive activity of dust, they also examined the size fractions of the particulate matter. Unpaved roads on two sites were studied: one in a limestone quarry, and the other at an open-pit coal mine. An acknowledged limitation was that dust sampling equipment for individual exposure assessment was used as an area sampler; however, the authors justify the decision by calibrating the air intake to take a more appropriate sample (Organiscak and Reed, 2004). Wind speeds were recorded at 30

second intervals using data loggers, allowing for more accurate modelling than possible using regional meteorological databases reporting climate averages (Organiscak and Reed, 2004). The results show that 80% of the dust was in the non-respirable fraction and that the remainder of respirable dust posed little risk to human health. Interestingly, the road surfaces at each site had silt contents well over 20%, which is more than double the average value given in the USEPA AP-42 tables of typical material properties which are provided in the NPRI pits and quarries guidance document. Like Veranth *et al.* (2003), Organiscak and Reed (2004) have found that dust from unpaved roads diffuses rapidly after being suspended and decreases to background levels at approximately 30 meters from the emissions point.

The AP-42 predictive emissions factor method is advocated by the NPRI as the preferred method for estimating fugitive dust discharge from quarries. Martin *et al.* (2008) compared an experimentally-determined emissions factor for a "dusty road" to one calculated using the AP-42 method, and it was found that the emission factor under-reported emissions by 20%. The finding is significant, because, as the major source of PM emissions from quarries, it can be argued that accurate quantification of this source is most important. These methods are discussed in greater detail in Chapter 5.

3.4 Material Handling

Despite being a significant source of fugitive dust, little research specifically targets emissions from material handling operations. Artinano *et al.* (2007), in a study of emissions from Tarragona harbour in Spain, attribute the lack of research to the discontinuity and randomness of emissions, as well as the diversity of materials. At

quarries, regular operations include the loading and unloading of material from haul trucks, additions to and removal from material storage piles, and the dumping of lime kiln dust and other wastes to on-site landfills (Western Regional Air Partnership, 2006). The description of the operations at Spanish harbours by Artinano *et al.* (2007) suggest similarities with a quarry setting, as a large amounts of raw materials – including coal - are loaded, unloaded, transported and stored, often releasing plumes of dust to the atmosphere. Like unpaved roads, silt content of the material and wind speed are important factors for fugitive dust generation, however the moisture content proves more significant as it dictates whether fine particles will aggregate or cement together (Western Regional Air Partnership, 2006).

Martin *et al.* (2007) conducted two field studies at the same harbour in association with the work of Artinano *et al.* (2007) to measure PM_{10}, $PM_{2.5}$ and PM_1 (particulate matter less than 1 micron) using laser spectrometers placed more than ten meters from materials handling operations. Dispersion and concentration of the fugitive dust were measured in order to determine emissions based on a Lagrangian puff model that involved various meteorological data. In each of three trials, the experimental emission factor was compared with the site-specific emission factor calculated using the AP-42 predictive emissions factor equations. Two of three trials showed no significant difference between the mass of PM estimated using emissions factors and the laser spectrometry, however the third revealed that AP-42 equations under-predicted the true emissions rate. It should be noted that the moisture content of the material used in the calculations was not

experimentally determined, so the average value found in the AP-42 tables was used (Martin *et al.*, 2007).

The AP-42 predictive emissions factor methodology is identical to that recommended by the NPRI for estimating emissions from pits and quarries, and Martin *et al.* (2007) show that the equations can be as accurate as field methods in some cases, but that this assumption is not necessarily true. It is also interesting to see the material moisture content from the AP-42 tables being used in the calculation without any on-site sampling. Although in this study it appears to have been successful, not measuring the specific material's moisture content is an invitation to error.

3.5 Wind Erosion of Storage Piles

The third important source of fugitive dust emissions at quarries is wind erosion of storage piles. In practice, the degree of erosion is related to the speed, direction, and gustiness of the wind, the elevation of the pile, particle size properties, the bulk density of the material, pile geometry, and the presence of a crust on the pile (Badr and Harion, 2007; Torano *et al.*, 2007). Storage piles at quarries may consist of waste rock, limestone waiting to be processed, or coal or coke used as kiln fuel at the processing plant. For wind erosion to occur, WRAP (2006) has established through experimentation that a storage pile must face gusts surpassing a threshold velocity of 5 m/s, as measured 15 cm above the surface of the pile. It should be mentioned that this threshold velocity was tested in the field using a portable wind tunnel on coal storage piles at a coal mine, but has been adopted for all materials (WRAP, 2006).

The emission rates of large particles in piles decreases over time as the loose material is lost, however any piles of material sand-sized or smaller have effectively unlimited erosion potential and thus the emission rates do not decline over time (WRAP, 2006). WRAP (2006) has published a very complex method for calculating wind erosion from storage piles that considers wind speed profiles and precise field measurements which make it very challenging to use. Fortuitously, the authors also make reference to the simpler AP-42 formula that calculates emissions based on the silt content of the material in the pile, precipitation, and average wind speed (WRAP, 2006; Environment Canada, 2008).

Badr and Harion (2007) have studied the configuration of storage piles as it relates to dust emissions, hypothesizing that the layout could be optimized to reduce wind erosion. Because the angle of repose for a material in a pile is fixed, the main variable of concern was the height (Badr and Harion, 2007). And since piles are obstructions to the flow of wind, they change the capacity of wind to erode other piles. For this reason, reputable Computational Fluid Dynamics (CFD) software was used to run a comprehensive numerical simulation of the turbulent flow over and around the pile surfaces. It is noteworthy that Badr and Harion (2007) use the same AP-42 predictive emissions factor equations recommended by the NPRI. The authors conclude that piles with an intermediate height of 12-14m and with a flat top – as opposed to the conical piles for which the equations were developed - resulted in the lowest fugitive dust emissions, and that pile configuration, turbulence, and complex terrains should be included in analyses of wind erosion from storage piles (Badr and Harion, 2007).

3.6 Dust Suppression

While mitigating fugitive dust emissions is motivated by environmental regulation and public perception, quarry operators also approach the problem for economic reasons. When aggregate material is lost from unpaved roads, costs related to grading, pothole refilling, and maintenance increase (Sanders and Addo, 1997). Wind erosion of storage piles has a financial cost, as limestone is lost before it can be sold, and fuel is eroded before it can be used to fire the kiln (Badr and Harion, 2005).

Several strategies have been used to mitigate dust emissions from unpaved roads including road wetting, reduced vehicle speeds, chemical dust suppressants, and physical barriers, all of which stabilize the road surface (Sanders and Addo, 1997). Neuman *et al.* (2008) state that road wetting is the most common method of suppression, but that few guidelines exist regarding that volume of water to be applied, nor the frequency of application. Tibke (1988) suggests roughening the land surface with small ridges which reduces wind speeds and thus erosion. Quarries do often grade their unpaved roads, which adds roughness, but it is unclear if their motivations are environmental or economic in nature.

Sanders and Addo (1997) conducted a quantitative study into the effectiveness and environmental impacts of various chemical dust suppressants on five sections of municipal unpaved roads. Test substances included hygroscopic chloride and magnesium salts, and lignin derivatives. Other effective chemical suppressants include soil cements, surfactants and bitumens, but these were not included in the study (Organiscak and Reed,

2004). Organiscak and Reed (2004) caution that no suppressant is 100% effective, and that the effectiveness degrades over time due to dry climatic conditions and traffic (Organiscak and Reed, 2004).

According to Sanders and Addo (1997), topical suppressants such as calcium and magnesium salts are easy to apply, but have a short-lived effectiveness and require multiple applications during the dry season. Contrasting these are suppressants that are mixed into the road surface using a grader; while more costly and labour intensive, they usually remain effective for the entire season (Sanders and Addo, 1997). Ultimately, the effectiveness of any suppressant is site-specific and depends on the type and volume of traffic, and the condition of the unpaved road (Sanders and Addo, 1997). The results from Sanders and Atto (1997) show that lignin reduced emissions more effectively than $CaCl_2$, $MgCl_2$, and a special ultra-pure $CaCl_2$ salt, but that all treatments resulted in significantly less fugitive dust than the untreated control. In terms of environmental impacts, all treatments were detected in runoff sampled after rain, however the implications were not discussed in any detail (Sanders and Atto, 1997)

3.7 Windbreaks

Windbreaks are a physical barrier against wind that reduce the uptake and suspension of dust by lowering its energy or by redirecting its flow (Brandle and Finch, 1991). According to Brandle and Finch (1991), barriers of this sort are typically made of trees, shrubs or grasses, but functionally-comparable barriers have been made of waste rock to mask both noise and the unsightliness of the site area. Baldauf *et al.* (2008) studied the air quality behind noise barriers surrounding roads and found that they reduced air

pollution by 20% on average. A consequence of reducing off-site pollution using a physical barrier is that pollutants become trapped on-site and increase occupational exposure (Baldauf *et al.*, 2008).

Height is the most important characteristic of a windbreak, as wind speed reductions are noticeable on the windward side from 2 to 5 times the height of the barrier; up to 30 times in the lee (Brandle and Finch, 1991). Barrier density is also an important characteristic as a barrier built of waste rock will inhibit wind flow and will reduce wind speeds more effectively than less dense vegetation barriers (Brandle and Finch, 1991; Cleugh, 1998). However, with all the wind being forced over and around dense barriers, a zone of low pressure develops on the leeward side which pulls the wind downward, increasing wind turbulence (Brandle and Finch, 1991; Cleugh, 1998). Therefore, dense windbreaks have a greater wind speed reduction, but a smaller protected area than a more porous windbreak would provide (Brandle and Finch, 1991). The issue, suggests Cleugh (1998), is that while turbulence is reduced in the vicinity of the windbreak, it is greatly enhanced in the wake. Therefore, a windbreak may reduce turbulent suspension of PM in some areas, and increase it in others (Cleugh, 1998).

Much of the research into windbreaks relates to increasing agricultural productivity. In that setting, windbreaks are used to create a more benign microclimate by providing shade and shelter to reduce land degradation (Cleugh, 1998). Cleugh (1998) suggests that shielding from wind reduces erosion, and that lower air temperature in the wake reduces moisture loss, however he cites several studies contending that the air temperature

actually increases in the wake. Overall, windbreaks profoundly affect the microclimate, which is important as microclimatic variables are fundamental influences on suspension, transport, and deposition of PM as is explained in next chapter (Cleugh, 1998; Baldauf *et al.*, 2008).

4 Particulate Matter in the Atmosphere

4.1 Particle Size Distribution

With the given size definitions of TPM, PM_{10} and $PM_{2.5}$, one might expect a simple mathematical relationship between the relative masses of each fraction in quarry emissions. This ratio between the fractions is termed the particle size distribution. Lodge *et al.* (1981) claim that the particle sizes from a given source will have a lognormal distribution, but Cowherd *et al.* (1998) explain that this is not the case at quarries due to the highly variable and site-specific physical and chemical properties of the PM emitted from each operation. Recent research by Choi and Fernando (2008) generalized that the fugitive dust from wind erosion of soils in the United States will have a $PM_{2.5}/PM_{10}$ ratio of 0.1, oversimplifying the processes that affect the particle size distribution. The size distribution of aeolian sediments is well known to be polymodal, reflecting its transport and deposition processes, but the degree to which this applies to dust generation processes was not uncovered in the literature (Sun *et al.*, 2002). Petravratzi *et al.* (2007) suggest that mineral hardness, moisture, density, and process intensity can all influence the dustiness of an operation, and the size of particles emitted.

Moisture is an important factor, as high moisture content facilitates adhesion between particles, increasing the relative proportion of large particles (Watson *et al.*, 2000). Unfortunately, complications like moisture are not reflected by ratios used to calculate relative quantities as was done in Choi and Fernando (2008). Presently there are no other generally-accepted, mathematical ways of predicting site-specific particle size distribution, strengthening our need to conduct physical sampling.

Establishing the spectrum of PM in each size classification is essential for assessing emissions, as the diameter of the particles directly correlates with its atmospheric transport and deposition; a characteristic necessary to predict how far it will travel off-site, and if the fugitive dust from a quarry threatens nearby communities (Countess *et al.*, n.d.). Harrison *et al.* (2001) cite Pye (1987) and Duce (1995) who both suggest that the largest fraction of dust subjected to long range transport is between 1μm and 2μm in diameter, a part of the $PM_{2.5}$ fraction. Chang (2004) provides confirming evidence, finding that airborne concentrations of TPM and PM_{10} decreased significantly at 100m and 300m from a source quarry, but that $PM_{2.5}$ did not exhibit this relationship. Generally speaking, fugitive dust with a higher diameter will result in lower off-site deposition because large particles settle more quickly than small ones (Chang, 2004; Grantz *et al.*, 2003).

4.2 Particle Interactions

Particulate matter, explained by Tecer *et al.* (2008), is a complex mixture of dry particles and particles with liquid coatings that have very different structures, shapes, and sizes. Structure, shape and size are all influenced by surface interactions including adsorption, absorption, nucleation, and adhesion (Lodge *et al.*, 1981). At the molecular level, PM with high surface roughness will tend to adsorb gaseous molecules to its surface (Lodge *et al.*, 1981). Absorption, the dissolution of the gas into the particle, also occurs, but is of much less importance (Lodge *et al.*, 1981). At the particle scale, PM will adsorb well to moist, rough, or electrically charged surfaces – this last principle employed by electrostatic precipitators used for industrial emissions control (Beckett *et al.*, 1998).

Grantz *et al.* (2003) suggest that adsorption reactions threaten plant life as phytotoxic organic compounds and herbicides may bind to the surfaces of particulates. Because adsorptive capacity is a function of surface area, $PM_{2.5}$ can carry a higher load of cations relative to mass, and become more problematic than PM_{10} or TPM fractions; the same principle that makes $PM_{2.5}$ most dangerous to humans (Grantz *et al.*, 2003). As more molecules become bound to the surface of the PM, the diameter and mass increase, inhibiting its transport (Giri *et al.*, 2008). The increase in diameter also results in a lower surface area to mass ratio, decreasing the capacity of the PM to adsorb other molecules.

Lodge *et al.* (1981) state that $PM_{2.5}$ is also the most active fraction in nucleation and adhesion reactions. Nucleation (also called 'hygroscopic growth') is the condensation of a vapour on a particle's surface, and occurs only while the vapour is present in sufficient quantity that it equals or exceeds its saturation vapour pressure (Lodge *et al.*, 1981; Beckett *et al.*, 1998). Vapours will continue to condense on the nucleated layer if their atmospheric concentration is sufficiently elevated. Problems arise when the PM is coated with condensate contaminated with toxic trace elements, volatile organic compounds, and products of incomplete combustion (Natusch and Wallace, 1974).

Like adsorption, nucleation has the beneficial effect of increasing the diameter of PM, limiting its travel distance. Besides condensed water vapour, nucleation can occur with volatilized nitric or sulphuric acids (Lodge *et al.*, 1981; Beckett *et al.*, 1998). These acids will not typically be emitted as a result of quarry operations themselves, but may be available in the atmosphere of quarries located near their processing plant, power plants,

or other industries that emit sulphur oxides (SO_x) or nitrogen oxides (NO_x). Interestingly, the high carbonate content of the PM generated at limestone quarries may scavenge some of the SO_x and NO_x, as well as smog precursors like ground level ozone (O_3) and the hydroperoxyl radical (HO_2) (Arimoto, 2001; Harrison *et al.*, 2001; Mohamed and El Bassouni, 2007). These alkaline emissions have also been known to neutralize hydrochloric acid, one of the major pollutants emitted from lime processing plants (Godoi *et al.*, 2008).

Particles with a diameter less than approximately 2 μm will typically adhere to one another after a collision, resulting with a larger particle (Lodge *et al.*, 1981). Adhesion does not usually occur between larger particles as they will tend to rebound elastically during a collision (Lodge *et al.*, 1981). A humid atmosphere increases the adhesive forces between particles, facilitating the formation of larger particles and the enhanced deposition of the suspended PM (Beckett *et al.*, 1998; Hann and Strazisar, 2007; Giri, *et al.*, 2008).

4.3 Deflation

According to Harrison *et al.* (1998), the cycling of dust in the atmosphere can be described in terms of three operations: deflation, transport, and deposition. Deflation occurs when particles are eroded from their underlying geological material by wind, and occurs when aerodynamic lift and drag forces overpower the materials' cohesive forces (Harrison *et al.*, 1998). Considering deflation as the only origin of fugitive dust, however, is inconsistent with the definition of fugitive dust, as it ignores the human-induced dust from abrasive and explosive activities. At the quarry, deflation is relevant

to the erosion of the quarry faces, flat surfaces, and storage piles; this PM becoming suspended in the atmosphere. Of these sources, the NPRI guidance for pits and quarries only requires the quantification of deflation from storage piles, and is therefore missing two major erodible surfaces.

The threshold wind velocity at which deflation occurs is called the wind shear velocity, and is dependent on many factors, including the material's grain size, aggregation, and moisture content, as well as the land surface roughness and vegetation (Harrison *et al.*, 1998; Hagen, 2004). Hagen's (2004) study of soil erodibility suggests that these factors are not independent, and modelling each one separately - as has been done, he claims, in many erodibility studies - can overstate the importance of that factor. Despite Harrison *et al.* (1998), who claim that moisture has a stabilizing effect on surface material; Hagen (2004) offers that the reduction in roughness caused by rainfall may increase deflation. Typically, however, low rainfall and low moisture has been found to result in more dust generation (Miller and Tegen, 1998).

Surfaces subject to deflation processes are limited reservoirs that become depleted over time without any abrasive action adding to its PM load (Watson *et al.*, 2000). A wind tunnel study by Neuman *et al.* (2008) into the wind erosion of mine tailings confirms this behaviour, as airborne PM concentrations degraded over time while wind speed was held constant. At a quarry where surfaces are continually disturbed by vehicle traffic, grading, and blasting, these reservoirs essentially become unlimited, and are eroded whenever the

wind shear velocity is surpassed (Watson *et al.*, 2000). Particles deflated from surfaces at a quarry are then suspended in the atmosphere and subject to transport mechanisms.

4.4 Transportation

Once PM is suspended at a quarry as a result of some combination of deflation, mechanical actions and explosive activities, it is subject to aeolian transport as dictated by weather conditions (Petravratzi *et al.* 2007). Harrison *et al.* (2001) describe dust transport as a function of wind strength and particle size, slightly different than the explanation of Countess (n.d.) who suggests that it is a product of vertical and horizontal mixing, and deposition rate. In a study by Tecer *et al.* (2008), it was found that surface-level wind speed was the governing factor in the transportation of $PM_{2.5}$ and PM_{10} in a Turkish coal mining region, which supports observations made by Badr and Harion (2005). In general, dry and windy conditions will result in more suspended PM, and further travel distances (Ghose, 2007).

Supporting the suggestion of Harrison *et al.,* (2001), Etyemezian *et al.,* (2004) state that as dust plumes travel, they spread in both vertical and horizontal directions; and elaborate that this diffusion causes a drop in airborne concentrations as a function of distance (Etyemezian *et al.*, 2004). Gaussian models are commonly used to predict transport and diffusion of PM because the degree of spread can be modelled as a function of time in three dimensions (Etyemezian *et al.*, 2004). The assumptions inherent in such models are that the concentration of PM will be normally distributed from its source, which Etyemezian *et al.,* (2004) suggest is too simple to account for variations in atmospheric stability. Gaussian models also assume that the concentration of PM in a plume heads

toward uniformity over time, implying a constant mixing ratio (Venkatram, 1993). According to Venkatram (1993), having a constant mixing ratio and uniform concentration in the vertical direction is a physical impossibility because air density varies with elevation.

During periods with calm winds, particles are subject to limited transport from the source of emissions, but with sufficient turbulence, will remain in the atmosphere for longer times and have the potential to travel thousands of kilometres before deposition (Watson *et al.*, 2000; Arimoto, 2001). Much of the research into long-range transport is related to dust from large deserts, like the Sahara, that has been detected across the Atlantic Ocean (Lodge *et al.*, 1981; Miller and Tegen, 1998; Harrison *et al.*, 2001). In order to travel these vast distances, PM must reach elevations of approximately three to five kilometres which can occur when intense daytime heating creates a thermally mixed layer, or when lifted by a cold front (Harrison *et al.*, 2001). On a smaller scale, ground heating can cause turbulent eddies, which in turn will increase the load of suspended particulate in the local airshed (Countess *et al.*, n.d.).

Ghose (2007) states that PM with a diameter of a 100μm or less – encompassing all three size fractions – is subject to Brownian motion and will remain in the air for a long time. However, evidence presented by Dyck and Stukel (1976) and Watson *et al.* (2000) suggests that dust plumes from unpaved roads and wind erosion of storage piles generally reach elevations between 1-2m, and are not subject long-range transport. Countess (n.d.) provides a different figure, claiming that the plume behind vehicles on unpaved roads

extends approximately 1-2 times the vehicle height, which suggests plume heights of up to 6m. When plumes remain low to the ground, particles collide with vegetation and other obstacles and are removed from suspension.

4.5 Deposition

Two mechanisms facilitate the removal of dust from suspension: wet deposition and dry deposition (Harrison *et al.*, 2001). Dry deposition is essentially gravitational settling of particles, and is thus a function of particle size and density (Harrison *et al.*, 2001; Countess, n.d.). Dry deposition is therefore enhanced by any processes that result in larger, heavier particle sizes, like adhesion, nucleation, adsorption and absorption (Countess, n.d.). Watson *et al.* (2000) generalize that PM greater than 30um in diameter is deposited near the emissions source, unless intense vertical mixing propels it high into the atmosphere. The particulate matter generated mechanically on roads or by other non-natural modes tends to remain closer to the ground and thus settle quickly (Countess, n.d.).

Wet deposition, on the other hand, is the removal of particles by precipitation and atmospheric moisture (Grantz *et al.*, 2003). Particles may be absorbed into water droplets in atmospheric moisture, and subsequently washed by precipitation, or alternatively by physical washing without being absorbed. Grantz *et al.* (2003) identify a third type of deposition as "occult", which is the interception by fogs and mists. Occult deposition is essentially a mixture of wet and dry deposition, but this distinction is not made by other authors.

4.6 Effects of Topography

Topographical features, like hills, share many common characteristics with manmade windbreaks (as discussed in section 3.6). Similar principles apply, as these natural features alter wind speed, wind direction and turbulence, and ultimately, the uptake and dispersion of fugitive dust (Arya and Gadiyaram, 1986). With respect to pollutant transport and dispersion, Castro and Apley (1997) suggest that the main effects of topography on pollutant dispersion are changes to the plume path, spread, and the possibility of the pollutants entering a recirculating region, which Gong (1990) attributes to changes in flow turbulence, and the convergence and divergence of wind caused by hills. Arya and Gadiyaram (1986) state that hills and other obstacles, such as buildings, disturb the flow of wind over a distance that is equal to or greater than the height of the obstacle. The interaction of wind with topographical features may result in significant changes to upwind and downwind concentrations of PM, but such a relationship cannot be predicted easily. A basic understanding of the effect of local topography is important for choosing a quarry's location, deciding which hills should be levelled or kept, and equally for locating storage piles to minimize erosion.

Jackson and Hunt (1975) were the first to study flows over complex terrain, but other authors have revised and improved the theory since that inaugural publication. Studies sprouting from their research are unique, as wind is characterized in the scale of relatively low hills, rather than the effects of large mountains (Arya and Gadiyaram, 1986). Arya and Gadiyaram (1986) conducted one such study, where the structure of the wake of conical hills was characterized in a wind tunnel. On undisturbed, flat terrain the

wind velocity profile was observed to be relatively constant at height, and to decrease logarithmically as it approached the ground surface; an effect of surface roughness (Arya and Gadiyaram, 1986). This observation confirms the well-known logarithmic law employed by some PM emissions factors (WRAP, 2006).

Flow measurements over the two conical hills showed accelerating wind velocities on the upslope and that the velocity profile at the crest was uniform rather than logarithmic (Arya and Gadiyaram, 1986). Interestingly, the mean velocity was higher at the crest of the more gently sloped hill, however Arya and Gadiyaram (1986) explain that the "speed-up factor" is dependent on both initial wind velocity and hill shape. Another possible explanation is that when hills reach a critical slope, flow separation occurs and hinders speed-up (Arya and Gadiyaram, 1986). Despite the acceleration on the upslope, wind speeds measured at different positions on the horizontal axis are reduced in the wake zone for between 15-25 hill heights depending on slope; higher slope having a more pronounced reduction (Arya and Gadiyaram, 1986). On the vertical axis, however, the reduction in velocity only affects winds within two hill heights (Arya and Gadiyaram, 1986). Unfortunately, the reduction in speed comes at a cost of greater turbulence in the wake zone.

With respect to turbulence, measurements by Arya and Gadiyaram (1986) show that it is maximized approximately 0.5 hill heights downstream, and across a width of 1.6 hill heights. It was, however, more intense downstream of the steeper hill (Arya and Gadiyaram, 1986). Because both turbulence and wind speeds are factors in the uptake

and transport of particulate matter, levelling a hill in one location or placing a storage pile in another is not the simplest of choices. However, beyond acknowledging the complexity of the problem, the way in which such choices should be made is beyond the scope of this review

Another important study is Gong (1990) where plume dispersion over hills was studied in a wind tunnel using a tracer gas, an approach mimicking Arya and Gadiyaram (1986). They found that on flat terrain, lateral concentrations followed a Gaussian distribution from the emission source (Gong, 1990). Lateral spread, under these conditions, was approximately equal at all heights (Gong, 1990).

One hill studied by Gong (1990) was a ridge, and expectedly it was found that the Gaussian distribution closely fit lateral pollutant concentrations. The only differences between the ridge and flat terrain were the rates of horizontal spread: at the top of the ridge the plume had lower spread than that measured at the top of the hill; and it was 35% higher at the foot of the hill that at the foot of the ridge (Gong, 1990). These phenomena were caused by differences in wind velocity and turbulence across the hill (Gong, 1990). It was also shown that, relative to the flat terrain, vertical dispersion was increased at the hill foot and suppressed at the hill summit, and that the increase and suppression were greater when the plume originated from a lower elevation (Gong, 1990). In the wake of the hill, concentrations return to their flat terrain values.

Another hill, a circular hill, showed a similar relationship: lateral spread was increased by the presence of hills near the emission source, and reduced at the summit of the hill (Gong, 1990). In this case, however, the effects of spread expanded the plume width to approximately twice that of the flat terrain case. This was due to flow divergence on the upslope, and flow convergence on the downslope (Gong, 1990). With respect to vertical dispersion, the effects were identical to two-dimensional ridge, however in this case, the elevation of the emission source was not a significant factor to the magnitude of plume spread. These observations suggest that considering topography of quarries and the shape of storage piles are important for accurately predicting emissions, the NPRI procedures do not mandate this depth of analysis.

Gong (1990) and Arya and Gadiyaram (1986) both used point sources of emissions and only few scenarios. Fugitive dust emissions at a quarry are emitted from various, unpredictable heights, complicating the development of simple relationships between wind speeds, turbulence, plume spread, and downwind concentrations. Additionally, quarries may have many topographical obstacles, and others like buildings and machinery that may counteract or synergize dust generation or suppression activities. Despite the difficulties, it is essential to understand that the topography of the land can significantly influence to both the quantity of dust generated, and its potential to reach nearby populations; this emphasizes the importance of using site-specific parameters when modelling fugitive dust emissions.

5 Emissions Calculation and Uncertainty Assessment

In preparing a quarry's emissions inventory for the National Pollutant Release Inventory (NPRI), Environment Canada offers the USEPA AP-42 emissions factors methods to quantify the mass emitted during the course of a year in their NPRI guidance document for pits and quarries.

An emissions factor (EF), in this context, is a statistical average of the rate at which a pollutant is released to the atmosphere as a result of some quantity of an activity (Ghose, 2007). For example, an emissions factor might have units of kilograms of PM_{10} per kilometre travelled by a vehicle, so multiplying the emissions factor by the annual kilometres traveled in a year would yield the PM_{10} emissions for that vehicle during that year. The AP-42 methods are used in place of the more complex mass-balance techniques and expensive dust sampling methods that exist, however they come with the cost of decreased accuracy.

The general form of an emissions calculation involving an EF is:

Emissions = Emissions Factor x Activity x Abatement Factor

5.1 Emissions Factors Equations

Establishing the most accurate emissions factors while ensuring that they apply to the widest variety of sites and settings remains a challenge. Occasionally the AP-42 methodology assumes all quarries are the same, for example, assuming that a specific mass of PM will be emitted per kilogram of blasting explosive used, without considering

the rock material or climatic factor (Environment Canada, 2007). Fortunately there has been an effort by several researchers to develop relatively basic equations to calculate an emissions factor based on local climate and material properties: predictive emissions factor equations.

According to Ghose (2007), Sendleim's (1983) predictive emissions factor equations are one of the better methods of determining emissions factors for particulate matter generated from the lime industry. Rather than using generic emissions factors and then multiplying them by site-specific activities, Sendleim's equations are desirable as they incorporate site-specific information. As Petavratzi *et al.* (2007) assert, local conditions are critical because each limestone deposit has a different propensity for generating dust. The statements made in Petavratzi *et al.* (2007) are suggestive that on-site sampling of materials is necessary, rather than the current practice of arming the equations with default values for siltiness and moisture content of materials.

Sendleim's predictive equations are given by Ghose (2007), and are very similar to the AP-42 predictive emissions factor equations. According to Ghose (2007), the emissions factors generated using predictive equations are only rules of thumb, and the site-specific nature of dust emissions is not reflected in these equations. This claim, while insightful, refers to the emissions factors equations supplied in an outdated, 1973 version of AP-42 that do not incorporate any site-specific data, only the activity rates. In the 1995 version, a series of more advanced equations is provided that included site-specific material properties and climate information as parameters, in addition to activity rates.

The AP-42 equations are slightly different from those of Sendleim (1983) as cited by Ghose (2007), and will be referred to as AP-42 predictive emissions factor equations for the remainder of the paper. Using Sendleim's predictive equations, it was shown that the calculated emissions were significantly different, and more accurate, than using the outdated 1973 AP-42 emissions factors equations (Ghose, 2007). Veranth *et al.* (2003) explain, however, that published predictive emissions factor equations are empirical, and based on only a small number of field studies, suggesting these equations are still approximations at best.

5.2 AP-42 Predictive Emissions Factor Equations

Despite limitations, the USEPA AP-42 predictive emissions factor equations remain a cornerstone of emissions calculations for the NPRI and other applications. Because calculating emissions from unpaved roads, material handling operations, and wind erosion from storage piles involves the input of climate data and material properties, these three major sources have been the focus of this study. Calculations of PM from other sources, such as overburden removal, grading, drilling, and blasting, are still only activity-based equations that do not involve either climatological or material parameters, and for this reason were excluded from the study.

5.2.1 Unpaved Roads

$$\textbf{Emissions Factor } (EF_{\text{unpaved roads}}) = k(s/12)^a \times (W/2.72)^b \qquad \text{(Equation 1)}$$

Where, $EF_{\text{unpaved roads}}$: Emission factor (kg PM/vehicle km travelled)
s: Surface material silt content (%)
W: Mean vehicle weight, (tonnes)
k, **a**, **b**: Numerical constants

To calculate PM emissions, the EF is multiplied by the total number of kilometres travelled by all vehicles on the unpaved roads. The numerical coefficients found within this equation are likely empirical constants discovered from experimentation and curve fitting, although the calibration of the equations is not made available for scrutiny by users.

Notes: This formula does not include the corrections for precipitation, snow depth, or dust suppressants added to unpaved roads.

Surface silt content (s) is ideally determined by on-site sampling, however in section 13.2.2 of AP-42 there are tables with typical values.

Mean vehicle weight (W) is an average of the loaded and unloaded weight of the vehicle travelling on the unpaved road.

k, a, and b are correction factors that are not well explained in section 13.2.2 of AP-42.

5.2.2 Materials Handling

$$\mathbf{EF}_{\text{materials handling}} = \mathbf{k} \times \mathbf{0.0016} \times \mathbf{(U/2.2)^{1.3}/(M/2)^{1.4}} \qquad \text{(Equation 2)}$$

Where, $\mathbf{EF}_{\text{materials handling}}$: Emission factor (kg PM/tonne material dropped)
U: mean wind speed (m/s)
M: material moisture content (%)
k: particle size multiplier (Note: this *k* is different from the numerical constant in Equation 1.)

To calculate PM emissions, the EF is multiplied by the total mass of material dumped during material handling operations, and must be calculated separately for each different material. All of the numerical coefficients and exponents found within are likely empirical constants discovered through experimentation and curve fitting, although the calibration of the equations is not made available for scrutiny by users.

Mean wind speed (M) is most accurate when measured on site using meteorological equipment, however weather data can be collected from a local weather station if site-specific measurements are unavailable. Section 13.2.4 of AP-42 does not give definite instructions on how the mean value is to be derived

Material moisture content (M), is the percentage of water in the material being handled. Ideally this value is a known material property determined through laboratory testing; however section 13.2.4 of AP-42 contains typical values. Like

the case of surface silt content, using these default values may introduce a lack of specificity

Particle size multiplier (k) is a factor that is applied to the calculation in order to isolate the masses of PM_{10} and $PM_{2.5}$ from the total particulate matter emissions. The assumption using these multipliers is that the *k* values in section 13.2.4 of AP-42 are representative of any material being handled.

5.2.3 Wind Erosion of Storage Piles

$$\mathbf{EF_{wind\ erosion} = 1.12 \times 10^{-4} \times J \times 1.7 \times (s/1.5) \times [365 \times (365\text{-}P)/235] \times (I/15)}$$

(Equation 3)

Where, **$EF_{wind\ erosion}$**: Emission factor (kg PM/m^2 surface area)
J: Particulate aerodynamic factor
s: Average silt loading of storage pile in percent (%)
P: Average number of days during the year with at least 0.254 mm of precipitation
I: Percentage of time in the year with unobstructed wind speed >19.3 km/h in percent (%)

To calculate PM emissions, the EF is multiplied by the total surface area of the storage pile, and unique EFs must be calculated for each material. However, if there are several piles of the same material their surface areas can be combined for the calculation

Particulate aerodynamic factor (J) is the same factor as the particle size multiplier used in Equation 2.

Average silt loading of storage pile, in per cent (P) is the percentage of silt in the material in the storage pile. Like moisture content, it is ideally determined through laboratory testing. Section 13.2.4 of AP-42 has a set of typical values.

Average number of days during the year with at least 0.254 mm of precipitation (P) is a troublesome quantity to as the closer value available from the Environment Canada Climate Normals web site is the number of days during the year with precipitation over 0.200 mm.

This value is included in the calculation as a mitigating factor against wind erosion. The "**[365 x (365-P)/235]**" portion of the equation is essentially saying that wind erosion is negligible every day during the year when precipitation is greater than 0.254 mm. If it rained every day of the year – if *P* was 365 - the annual PM emissions from this source would be zero. No explanation is available for the division by 235.

Percentage of time in the year with unobstructed wind speed >19.3 km/h in per cent (I) - Environment Canada instructs the user to count the number of months with average monthly wind speed greater than 19.3km/h, which, except in rare cases, will almost certainly be zero.

5.3 Uncertainty Assessment

When calculating emissions for an emissions inventory, the Intergovernmental Panel for Climate Change (IPCC, 2000) asserts that quantitative estimates of uncertainty are an essential in any complete inventory. The IPCC (2000) recommends including confidence limits on estimates as a way of identifying inaccuracies, and as a way of guiding future choices of emissions calculation methodology.

Poulter (1998) states that including confidence limits on calculations in the context of environmental risk assessment is desirable because it provides insight into the high and low ends of risk, while highlighting the central tendency. It is under this framework that quantitative uncertainty assessment is explored, with a particular focus on the simple yet robust Monte Carlo methods advocated by the IPCC and a variety of other studies where complex systems are modelled.

When preparing emissions inventories, direct measurements of emissions are rarely made, yet increasingly stringent regulatory demands require improving the accuracy of estimates (Winiwater, 2007). Because the uncertainty portion of the assessment is reducible – and thus is an opportunity to improve inventories – understanding its types and sources is crucial for prioritizing areas needing improvement (Winiwater, 2007). For these reasons, uncertainty assessment using Monte Carlo simulation has already been

used to assess national green house gas (GHG) emissions inventories in Australia, Norway, and the United Kingdom; uncertainties of plus or minus 4-21% were found which are large compared to emissions reductions targets (Monni *et al.*, 2004). Monni *et al.* (2004) have applied similar methods to Finland's GHG emissions inventory, and have estimated 95% confidence limits of -5 to +6% around total emissions. In Finland, the national level emissions are much more certain than those of specific sectors of the economy; the industrial sector having 95% confidence limits of -27% to +43% (Monni *et al.*, 2004).

Uncertainty in a model's predictions can be separated further into model uncertainty, relating to model construction; parameter uncertainty, relating to the uncertainty in a point estimate of outcome, and input uncertainty; relating to the quality of input data (Sax and Isakov, 2003). Model uncertainty can be assessed statistically or by validation with true values. A detailed discussion of quantifying parameters and input uncertainties follows, as the quality of data used in calculations, and the degree of uncertainty in quarry NPRI reports, is under investigation.

5.4 Input and Parameter Uncertainty

In the context of both site- and national-scale emissions inventories, measures of uncertainty contain two components: uncertainty and variability. Uncertainty is imprecision that results from poorly characterized processes, model assumptions, unrepresentative data, and measurement error, but unlike variability, it can be reduced with further study or having a more representative sample (Gardner and O'Neill, 1979; Finley *et al.*, 1993; Frey and Burmaster, 1999; IPCC, 2001). Variability is the

component of the uncertainty measure that cannot be reduced, because it comes from natural heterogeneity in a population (Frey and Burmaster, 1999). Even in an ideal scenario where the model describe phenomena perfectly, and true values of inputs were known, variability would still remain. Gardner and O'Neill (1979) describe variability "as the population of possible behaviours", which is further explained Monni *et al.* (2003) who explain that this is a function of data being heterogeneous with respect to time and space.

Input uncertainty occurs when data introduced to a model have measurement error, estimation error, or is not representative due to a small sample size (Sax and Isakov, 2003; Intergovernmental Panel on Climate Change, 2001). Besides the challenges of characterizing data accurately, it is this natural variability that suggests even with high certainty in an input, a single value can never completely describe that parameter; this phenomenon is called parameter uncertainty (Sax and Isakov, 2003). All the different sources of uncertainty and variability fed into a model are interdependent, but as New and Hulme (2000) explain, not in a predictable additive or multiplicative way. Kelly and Campbell (2000) assert that it is a necessity to separate uncertainty from variability, however this is only possible with a large, representative data set.

While this preceding explanation focused on the numbers inputted to a model, Gardner and O'Neill (1979) strengthen the need for uncertainty assessment from the output side. Specifically, they suggest that it is incorrect to have a deterministic, point estimate model output when the purpose is to represent a variable system. Vose (1996) calls

deterministic modeling of these types of systems "best guess". The conclusion of Gardner and O'Neill (1979) stems from the fact that the expected value of a nonlinear function with a set of randomly variable inputs is not equal to the value of the function when the expected value – typically the mean – is used in the equation (Gardner and O'Neill, 1979).

Using an average point value for wind speed and precipitation values that exhibit variability on a daily, even hourly (or less), basis is not mathematically correct in the context of Gardner and O'Neill (1979). Using an annual mean value in EF equations, as is advocated by the NPRI, removes the effect of temporal fluctuations. Frey (1997), in his assessment of uncertainty found within emissions factors themselves, states that the type of averaging used to aggregate spatial and temporal differences is, in itself, a contributor to uncertainty. Vallack and Shillito (1998) agree with this idea, suggesting that fugitive dust from quarries tends to be episodic with large intermonth variation. This makes it clear that using climate values averaged over the course of a day would better incorporate temporal fluctuations in meteorology. Ultimately, having values in the scale of minutes or seconds would be an improvement still, but this type of data is not readily available.

The AP-42 predictive emissions factor equations for major sources like unpaved roads and materials handling are nonlinear, both having inputs like silt and moisture contents raised to exponents. Environment Canada instructs quarries to use published mean values for these two inputs which introduces considerable parameter uncertainty because

these averages are based on small sample sizes, have not been measured on site, and therefore do not adequately describe the parameter for the intended use. Even in the case of linear equations – like that for wind erosion from storage piles – the inevitable natural variability in equation parameters would seem to make quantifying uncertainty a necessity.

Finley *et al.* (1993) and Thompson (1999) suggest that correlations between input variables should be explored and accounted for to prevent impossible combinations, but Hanna *et al.* (1998) suggest that correlating data in a simulation model introduces unnecessary uncertainty. Hanna *et al.* (1998) used Monte Carlo simulation to quantify the uncertainty in photochemical grid models which are used to demonstrate the effectiveness of emissions control strategies. In this study, they claim that if the correlation coefficient between variables is less than 0.30 then the correlation effects will be insignificant (Hanna *et al.*, 1998).

5.5 Monte Carlo Simulation

As alluded to earlier, the most accepted technique for including uncertainty measures in model inputs, and quantifying them in the outputs is Monte Carlo (MC) simulation models (Burmaster and Anderson, 1994). The concept was originally used to solve statistical physics problems – notably at the Los Alamos Laboratory where the first nuclear weapons were produced - but has since been applied by chemists, epidemiologists, ecologists, and climate change scientists (Metropolis and Ulam, 1949). Essentially, suggests Vose (1996), it is a computationally intensive version of a "what if" analysis.

MC simulation involves building a computer model with multiple uncertain variables describing each variable by a function that relates possible values with their likelihoods of occurring, having software randomly select and input plausible values for each variable into the model, calculating the results, and repeating the process until the probability distribution of output values does not change significantly from one iteration to the next (Metropolis and Ulam, 1949; Vose, 1996). MC simulation weights each possible outcome by its probability of occurrence by running the model with different combinations of values for each variable, with each value randomly selected from the predetermined distribution (Vose, 1996). The biggest advantage of this technique, however, is the relative simplicity of combining multiple uncertainties compared to difficult and laborious analytical techniques (IPCC, 2000; Monni *et al.*, 2004;).

Although an application to quarry emissions calculations has not been found in the literature, one related, common application of MC methods is analyzing impacts of climate change. This phenomenon is a good candidate for MC simulation because there is incomplete knowledge of emissions, poor understanding of atmospheric physics, and computing limitations (New and Hulme, 2000). New and Hulme (2000) quantified uncertainty in climate change scenarios by conducting a Bayesian Monte Carlo simulation, which is a slightly more complex method that accommodates changes in a variable's probability of occurrence over time. Modifying probabilities over time is necessary in climate change simulation as emissions scenarios result in different greenhouse gas levels, and consequently different values for radiative forcing. Despite

computational differences of Bayesian Monte Carlo simulation, the basic methodological framework exists and the model was used to account for uncertainty, climate sensitivity, strength of carbon sinks, global atmospheric circulation, among other variables (New and Hulme, 2000).

New and Hulme (2000) made predictions of global temperature change at 30 year increments using MC methods, but unlike New and Hulme (2000), estimates of technological change and economic growth were included, as were emissions-capping policy scenarios (Webster *et al.*, 2003). By 2100, Webster *et al.* (2003) predict with 95% certainty that annual CO_2 emissions will be between 7 and 36 gigatonnes (Gt) per year, with a mean of 19 Gt. This range is not presented for contrast with other climate change simulations, but to demonstrate that like the Finnish emissions inventory of Monni *et al.* (2003), the confidence limits range between -36% to +52% of the mean value is acceptable. Ramirez *et al.* (2008), in assessing the uncertainty of the Netherlands GHG inventory, found that the uncertainty was similar to the Finland study by Monni *et al.* (2003) as well as Belgian and Austrian values. As expected, the uncertainty related to the type and strength of emissions-reduction policy – specifically that related to sulphur emissions – has high uncertainty.

While the two previous examples have focused on national and global emissions, Rodriguez *et al.* (2007) have used Monte Carlo simulation paired with a 3D air quality model to predict ozone and $PM_{2.5}$ concentrations attributable to distributed power generation in several regions of southern California. Their model had 11 input variables

including boundary concentrations and emissions rates of O_3, NOx, VOCs, NH_3, and 6 more representing photochemical reactions (Rodriguez *et al.*, 2007). It is worth noting that $PM_{2.5}$ is not one of the input variables, as only the secondary particulate matter resulting from chemical reactions is quantified (Rodriguez *et al.*, 2007).

Rodriguez *et al.* (2007) used the variance as a measure of uncertainty, and in the case of ozone emissions, found uncertainty of 42% in Central Los Angeles which has high automobile traffic. Estimates of $PM_{2.5}$ were more certain, having variances in the range of 11 to 17%. This higher certainty is related to ozone and $PM_{2.5}$ having different sensitivities to each model input (Rodriguez *et al.*, 2007). Interestingly, the authors only ran 50 iterations of the model to come up with these uncertainty estimates. It is claimed that this is sufficient due to the small number of variables, however most authors use at least 1 000 iterations (Gardner and O'Neill, 1979; O'Neill *et al.*, 1980; Frey and Burmaster, 1999)

Site-scale emissions inventories are commonplace in Canadian industry due to NPRI reporting laws, but the uncertainty in smaller scale inventories – like that of a quarry site – has been poorly studied. In fact, no published research has been found that attempts to simulate climate conditions with respect to emission factors. Sax and Isakov (2003) present one of the few attempts to characterize uncertainty at the local scale in they study of hexavalent chromium – Cr(VI) - emissions estimates from a shipbuilding facility in California. Emissions were calculated by multiplying the average annual process rate by an emissions factor describing the expected fume emission rate, by multiplying by the

Cr(VI) content in the fume, and then inputting the values into a Gaussian dispersion model (Sax and Isakov, 2003). Because there is variability in process rates due to fluctuations depending on the type of project, MC simulation was used to estimate the possible range of annual emissions (Sax and Isakov, 2003).

The Gaussian dispersion model introduces additional uncertainty because, as Sax and Isakov (2003) state, the model cannot perfectly describe a physical process of such complexity. Like emission rates, site-specific meteorology has a large degree of variability, so a simple average values is not appropriate for use (Sax and Isakov, 2003).

5.6 Probability Density Functions

Central to MC simulation are probability density functions (PDFs), mathematical functions describing the plausible values that a variable may have, and the probabilities of assuming those values (Thompson, 1999). PDFs are commonly continuous functions, with an infinite number of possible values, however discrete functions may be used when there are only distinct values that a variable may assume (Copeland *et al.,* 1993; Thompson, 1999). When the distribution is continuous, they are frequently modelled as normal or lognormal curves, but a variety of other distribution shapes exists (Copeland *et al.,* 1993). Besides normal and lognormal functions, many parametric distributions have been developed to describe natural systems, including beta, exponential, gamma, logistic, triangular, and uniform – among countless others (Vose, 1996).

Thompson (1999) has developed a comprehensive set of guidelines for developing univariate distributions for use as PDFs in human health risk assessment; but the same

principles apply to any situation. Having good data is the basis for developing distributions, as understanding about the mean and variance of a population are usually needed as parameters in the equations (Thompson, 1999). If there is insufficient data, as is often the case, it must be adjusted or extrapolated using best judgment to adequately represent the variables in context of the assessment goals (Thompson, 1999).

In cases where only the range of possible values is known about a variable, Copeland *et al.* (1993) recommend a uniform distribution be used, giving each possible value an equal probability of occurring. If only the range and mode is known, a triangular distribution is recommended by Copeland *et al.,* (1993). This distribution is a continuous function with the only parameters being the lower limit, the mode, and the upper limit, and in practice begins to approximate a normal distribution (Copeland *et al.*, 1993). Finley *et al.* (1993) also recommend using this distribution as a more conservative version of a truncated normal or lognormal distribution; more conservative because extreme values in the range have a higher probability of being selected. In New and Hulme (2000), IPCC climate sensitivity parameters which fall within a range of 1.5°C to 4.5°C were assigned a triangular probability distribution in order to deemphasize high and low values. It should be understood, however, that using a triangular distribution – or any other distribution – does not mean that the true distribution has that shape, but that it is an acceptable way to represent the data at hand (Finley *et al.*, 1993). As implied, judgment must be used when picking a distribution, but even an imperfect PDF has been shown to introduce less than 10% error (Copeland *et al.*, 1993; Thompson, 1999).

With the basis of emissions factors, the theory of MC simulation, and probability density functions covered, it is now appropriate to apply these concepts in quantifying the PM emissions from three quarries in Quebec, as is covered in the following chapter.

6 Methodology

The shortcomings of the NPRI pits and quarries PM emissions calculations can be isolated into three issues:

1. The potential for error when calculations are done with 30-year climate averages
2. The uncertainty introduced by using typical material properties from AP-42
3. Limited availability of spatially-relevant climate data needed for the calculations

A case study approach was used to examine these three problems, which have each been addressed individually as proofs-of-concept. It should be emphasized that the issues are not distinct in practice. Validation of the emissions calculations would have been desirable; however, this was beyond the scope of this research.

Problem #1: Error of Using 30-Year Climate Averages

Because of the limitations of using average values in nonlinear equations like the predictive emissions factor equations (see Chapter 5), it was hypothesized that the effect of peak wind and precipitation periods are dulled when 30-year climate averages are used in PM emissions calculations and that the use of climate data with higher temporal resolution would drastically change results. To test this hypothesis, emissions were calculated twice; once using 30-year climate averages, and a second time using a combination of daily and hourly climate data. No attempt was made to validate the accuracy of the two methods, but to expose differences and establish a procedure that can be used for including data with higher temporal resolution in the calculations.

This approach using daily and hourly data is not unique, as the NPRI provides the Unpaved Industrial Road Dust Calculator that incorporates daily precipitation and snow depth values in the calculation. Concepts from this spreadsheet were reproduced for unpaved road calculations, and expanded to the calculations for materials handling and wind erosion from storage piles.

Problem #2: Uncertainty of Material Properties

In preparing emissions inventories for the NPRI, quarries are not required to collect site-specific information regarding the silt and moisture contents of their unpaved roads and other on-site materials. Instead, the NPRI provides a series of typical values for material properties needed for the calculations. These typical values reproduced by the NPRI for use by quarries were originally published by the USEPA in sections 13.2.2 and 13.2.4 of their AP-42 documentation, and are a collection of minimum, mean, and maximum from government studies and other literature. Unfortunately, the sample sizes and number of sites sampled are too small to be representative or termed "typical". Therefore, the uncertainty introduced into the emissions calculations when single point values are selected from these tables is assessed using Monte Carlo simulation. Typical values are discussed in further detail in Section 6.3.

Problem #3: Poor Availability of Spatially-Relevant Data

Some quarries are located in towns that do not have an Environment Canada weather station that reports the climate averages needed to calculate emissions following the NPRI guidelines. It was hypothesized that spatial interpolation using geographical

information systems (GIS) would be a viable solution to the problem of obtaining the necessary climate averages for quarries in remote locations.

6.1 Climate Data

The NPRI guidance instructs reporters to consult the National Climate Data and Information Archive (NCDIA), an online repository operated and maintained by Environment Canada to collect the climate data necessary to complete the pits and quarries PM emissions calculations. As mentioned in Problem #3, quarries are often located far from Environment Canada weather stations that publish to this database. In these cases, the simplest solution is to use a nearest station approach and assume that the nearest weather station reporting the required climate data is representative of the site (Nalder and Wein, 1998). This method is defensible when weather stations are located relatively close to the quarry, and according to Nalder and Wein (1998), occasionally proves more accurate that many of the more complex interpolation methods.

Rather than struggle with the nearest station method which might involve selecting data from several stations and applying them to one site, a "What If" approach was adopted to better demonstrate the difference in the choices of climate data. The approach was therefore to consider what the emissions would from each quarry had they been located elsewhere where the full array of average, daily, and hourly climate data was available.

6.1.1 Thirty-Year Climate Averages

Thirty-year climate data were collected from the "Climate Normals" section of the NCDIA. Necessary data included:

- Annual average wind speed
- Annual number of days with precipitation greater than 0.254mm, and
- Average monthly snow depths

It should also be noted that the number of days with precipitation greater than exactly 0.254mm is not reported in Climate Normals, so the number of days with precipitation greater than 0.2mm was used, as it is the closest value available. The minute difference was not expected to significantly affect calculations.

Another exception was made in determining the percentage of time with wind speed greater than 19.3km/h, which is not possible following the NPRI guidance. Users are instructed to consult the NCDIA "Climate Normals and Averages" to find the average wind speeds for each month, to count the number of months with average wind speed greater than 19.3km/h, and finally to divide this number by 12 months to get the annual percentage of time. These instructions are problematic as wind speeds averaged over a month will rarely approach 19.3km/h. Therefore, following this method will yield zero per cent, and thus zero wind erosion from storage piles.

To circumvent this problem, hourly wind speeds from 2007 were collected from the bulk data spreadsheets available from the "Climate Data Online" section of NCDIA and were

used to calculate a more realistic percentage of time. One limitation of this resource is that the wind speeds are published as integers, meaning that getting a percentage of time with wind speeds greater than exactly 19.3km/h was not possible. Therefore, the number of hours with wind speed values greater than 19km/h were counted for each month and divided by the total number of values collected during that month to generate a percentage. The twelve monthly percentages were averaged to generate an annual average. Calculating averages on a monthly basis and then averaging them for the year is the approach that EC takes to calculating annual averages for the other variables on their "Climate Normals and Averages" database and thus was mimicked for this portion of the analysis.

6.1.2 Daily and Hourly Climate Averages

Daily and hourly data were obtained from the bulk data spreadsheets available in the "Climate Data Online" section of NCDIA, and the data with highest temporal resolution was used for the calculations. Collected data included:

- Hourly wind speeds
- Daily precipitation
- Daily snow depths

For the determination of the percentage of time with wind speed greater than 19.3km/hr, the same approach as described in Section 6.1.1 was followed, but in this case the data from all twelve months were aggregated into a single spreadsheet and an annual percentage was directly calculated without averaging monthly percentages. This was

considered more accurate than preparing 12 monthly averages and then averaging those to get a yearly average.

6.2 Quarry Operational Data

With the assistance of a Canadian lime producer, data from three quarries in southern Quebec which reported to the NPRI in 2007 were available for this study. As a condition for using the data, the company requested that their name and the exact locations of the quarries be kept confidential. The quarries were not intended to represent typical Canadian quarries, but rather to be a source of realistic data that can be used to demonstrate some of the concepts and issues under investigation.

Production data from 2007 needed to calculate each quarry's PM emissions were extracted directly from corporate production spreadsheets. Production data include the masses of rock blasted, high quality limestone, waste rock, and lime kiln dust (LKD). Quarries often contained LKD landfills which were suspected to be subjected to wind erosion, but because the AP-42 calculation methodology does not address this source, these emissions were not considered in any calculations.

Other operational data needed for the calculations, including vehicle mileages, vehicle empty and loaded weights, unpaved road dust control practices, and storage pile sizes which were taken from the spreadsheets prepared for the quarries' 2007 NPRI report by an external consultant. The data in these spreadsheets were originally collected through phone conversations and electronic correspondence with quarry managers at each site during May of 2008 before the completed report was submitted to the NRPI. There was

no additional correspondence with the consultant or the quarry management during this research to confirm or revise values.

6.3 Material Properties

Because no on-site sampling was conducted at any quarries to establish the silt and moisture contents of materials, a weighted average was calculated using the "typical values" published in the NPRI guidance. These typical values maintained in AP-42 by the USEPA, are a collection of minimum, mean, and maximum values. While convenient, the values are based on few samples and a small number of sites which casts doubt on whether the values are truly "typical". According to Neuman *et al.* (2008), the values in these tables may bear little or no resemblance to the materials on site and result in over- or under-estimations of emissions by several orders of magnitude. Simon *et al.* (2008) assert that the silt contents of unpaved roads listed in AP-42 are not well characterized and are not suitable for use in many areas of the United States. Rather than assume one minimum, mean or maximum value from the table was correct for a material, a weighted average of all values for the material was calculated, and assumed to be the most representative average value for that material.

An attempt was made to review the primary data used to populate the typical values table, however the referenced studies were either unavailable for review or did not provide any additional insight. Without having individual data points, each minimum, mean, and maximum value was treated as a unique data point. The weighted means and standard deviations are shown in Table 1; full detail can be found in Tables B.1-B.7 in Appendix B.

6.3.1 Lime Kiln Dust

The moisture content for LKD was not listed in the AP-42 documentation, therefore an alternative source was sought. Data were only found at the Turner Fairbanks Highway Research Center (TFHRC), an institute connected to the U.S. Department of Transportation. Much like the AP-42 tables, insufficient detail and the small sample size limits confidence in the data. Two values for loss-on-ignition (LOI) are given for fresh LKD; 14.2% and 37.4%. Stockpiled LKD only has a single LOI value, 27.9% (Turner Fairbanks Highway Research Center, n.d.). The mean of these three moisture content values is 26.5%, and is quite close in value to the LOI for cement kiln dust, a comparable material, as reported by Sreekrishnavilasam *et al.* (2006).

The silt content for LKD was not collected for this research as it was not needed for any of the calculations.

Table 1: Summary of weighted average material properties

Material	Property	Weighted Mean (%) (μ_w)	Weighted Std. Dev. (σ_w)
Unpaved Roads	Silt Content	9.3	7.7
Limestone	Silt Content	4.46	5.37
	Moisture Content	2.12	1.95
Coal	Silt Content	3.88	3.62
	Moisture Content	5.48	1.36
Coke	Silt Content	4.9	0.71
	Moisture Content	7.8	1.98
Lime Kiln Dust	Moisture Content	26.5	11.66

6.4 Emission Calculation Using Thirty-Year Climate Averages

The calculations of particulate matter emissions using 30-year climate averages followed the NPRI guidance, using the unmodified AP-42 methodology described in Chapter 5.

The climate data used were the 30-years averages collected from the NCDIA as described in section 6.1.1. The material properties used were the weighted means from AP-42 as listed in Tables B.1 through B.7 in Appendix B.

6.4.1 Predictive Emissions Factor Equations

To calculate emissions using 30-year climate averages, $EF_{unpaved\ roads}$, $EF_{materials\ handling}$ and $EF_{wind\ erosion}$ were used. Consult Section 5.1 for a complete description of the equations and necessary operational parameters.

For the unpaved roads calculations, an abatement factor of 100% was applied based on number of days with precipitation greater than 0.254mm. For example, if 50% of the days during the year met this threshold, the annual emissions total was halved. This is in accordance with the NPRI instructions within the Simplified Industrial Unpaved Road Dust Calculator (Environment Canada, 2007) and the Western Regional Air Partnership (2006). Additionally, an abatement factor of 100% was applied based on the number of months with average snow depths greater than 15cm following guidance of the Industrial Unpaved Road Dust Calculator – see Table 2 (Environment Canada, 2007) and Western Regional Air Partnership (2006). Although snow depths are not used in the simplified calculator, it was applied in this study in order to maintain comparability between the different calculations.

6.5 Emission Calculation Using Daily and Hourly Climate Data

In order to use more temporally-relevant daily and hourly climate data, the AP-42 predictive emissions factor equations were calculated in slightly different way, following

an approach based on the NPRI Industrial Unpaved Road Dust calculator (Environment Canada[2], 2007. Daily and hourly data were obtained from the bulk data spreadsheets available in the "Climate Data Online" section of NCDIA, and the data with highest temporal resolution was used for the calculations. Like the 30-year climate averages calculations, the material properties were the weighted means from AP-42 as shown in Table 1.

The daily/hourly approach involved modifying the equations to calculate daily or hourly emissions factors, depending on data availability.

6.5.1 Calculation Using Daily Data – Unpaved Roads

In situations where daily data were the most frequent data available, the procedure was based on the concept that:

Daily Emissions Factor = Annual Emissions Factor / 365 days per year,

Or equally that

Annual Emissions Factor = 365 days per year *** Daily Emissions Factor**

This was substituted into the standard emissions calculation of

Annual Emissions = Annual Emissions Factor * Annual Activity

Leading to

Annual Emissions = (Daily Emissions Factor * 365 days *** Annual Activity** (Equation 4)

The multiplication by 365 days was carried out by adding the 365 daily emissions values. This technique using daily data was only employed in calculating emissions from

unpaved roads, as the only meteorological data needed are precipitation values, which are published at daily intervals. The following was the predictive emissions factor equation for daily emissions from unpaved roads:

$$\textbf{Daily Emissions Factor (DEF)} = k(s/12)^{a} \times (W/2.72)^{b} / 365 \qquad \text{(Equation 5)}$$

Where, **DEF**: Daily emission factor (kg PM/vehicle km travelled)
s: Surface material silt content (%)
W: Mean vehicle weight, (tonnes)
k, **a**, **b**: Numerical constants

Following the guidance of the NPRI's Industrial Unpaved Road Emissions Calculator (Environment Canada, 2007) and WRAP (2006), the abatement factors in Table 2 were applied to the calculations based on the daily precipitation and snow depth values:

Table 2: Precipitation and snow depth abatement factors

Precipitation in a day period	Adjustment for precipitation	Assumed Efficiency
< 0.2 mm	No Control	0%
>= 0.2 mm And < 5 mm	Minimal control	10%
>= 5 mm And < 10 mm	Moderate control	50%
>= 10 mm And < 25 mm	Near maximum control	80%
>= 25 mm	Maximum control	100%
Snow Depth >= 15 cm	Maximum control	100%

(Environment Canada, 2007)

6.5.2 Calculation Using Hourly Data – Materials Handling

In the more desirable case where hourly data were available, the calculations took a similar form:

Hourly Emissions Factor = Annual Emissions Factor / 8760 hours per year,

or conversely that

Annual Emissions Factor = 8760 hours *** Hourly Emissions Factor**

This was then be substituted into the standard emissions calculation of

Annual Emissions = Annual Emissions Factor * Annual Activity

which led to

Annual Emissions = (Hourly Emissions Factor * 8760 h**) * Annual Activity** (Equation 6)

This technique using daily data was employed in calculating emissions from materials handling, as the only meteorological data needed are average wind speeds, which are published at hourly intervals. The multiplication of hourly emissions factor by 8760 hours was done by adding the 8760 hourly emissions factors to generate an annual EF and then this was multiplied by the annual activity; a slightly different order than the unpaved road calculation due to different parameterization and abatement factors.

$$\mathbf{DEF} = \mathbf{k} \times 0.0016 \times (\mathbf{U}/2.2)^{1.3}/(\mathbf{M}/2)^{1.4} / 8760 \qquad \text{(Equation 7)}$$

Where,
EF: Emission factor (kg PM/tonne material dropped)
U: mean wind speed (m/s)
M: material moisture content (%)
k: particle size multiplier (Note: this *k* is different from the numerical constant in Equation 1.)

6.5.3 Calculation Using Daily and Hourly Data – Wind Erosion of Storage Piles

Calculating fugitive dust from wind erosion of storage piles involved a combination of daily and hourly data, but no modification to the formula. The values for the number of days with precipitation greater than 0.254mm were determined using daily climate data from 2007. The percentage of time with wind speed greater than 19.3km/h was calculated as described in section 6.2.3 using hourly data from 2007.

6.6 Uncertainty Assessment: Material Properties

The NPRI pits and quarries guidelines permit calculating emissions using the average values for silt and moisture content of a material from the AP-42 data presented in Tables A.1-A.9 in Appendix A. When choosing an average from AP-42, a point value is selected from a large spread of potential values, or from so few data points that the values cannot be considered representative. Therefore, when using typical values instead of analyzing site-specific samples, it is logical to analyze the uncertainty of the choice, and attach measure of confidence attached to the PM emissions calculations.

6.6.1 Monte Carlo Simulation

To assess uncertainty, a Monte Carlo simulation was conducted using Decisioneering's Crystal Ball 7 software, termed "Crystal Ball" for the remainder of the document. Crystal Ball is recommended by both the International Panel on Climate Change (2000) and by Monni *et al.* (2004) for assessing the uncertainty of emissions inventories. Each simulation involved 1000 iterations of the model, which is the number recommended by O'Neill *et al.* (1980), Gardner and O'Neill (1979), and Frey and Burmaster (1999).

Using this software, each silt and moisture content values in the calculation using climate averages was retrofitted with a probability density function representing the possible values and likelihoods of occurrence, as will be described in section 6.6.2. From these distributions, simple random sampling (SRS) - as opposed to Latin Hypercube sampling – was used to select a value for each material property to be assessed. LHS was not used as it removes randomness by forcing the software to choose an equal number of values from all parts of the PDF. This approach is only necessary when computational power or model complexity limits the number of iterations that can be run (Hammonds *et al.*, 1994).

6.6.2 Probability Density Functions

Selecting a probability density function (PDF) to adequately represent the potential values for a variable and likelihood of their occurrence involves both objective statistics and subjective choices. The statistical portion of developing PDFs involves calculations of means and standard deviations necessary to parameterize the distribution, but choosing the shape of the distribution involves a degree of educated subjectivity, particularly when data are sparse (Baraldi and Zio, 2008). Tests for choosing the best-fitting distributions are available, including Kolmogorov-Smirnov, Anderson-Darling, and Chi Square, but they are not foolproof and require the assessor to use best judgment (Hammonds *et al.*, 1994; Thompson, 1999; Romano *et al.*, 2004). Following the principles of good practices outlined by Burmaster and Anderson (1993), each PDF used in this uncertainty assessment, the summary statistics, and the assumptions made, are detailed in the results.

6.7 Spatial Interpolation

Even under the assumption that using 30-year climate averages is an accurate way of calculating emissions, following the NPRI guidance remains problematic for quarries in remote locations or located near a weather station that has not published averages for all climate parameters in the equations. Therefore, the inability to acquire of spatially-relevant climate data forces NPRI reporters to seek the missing data from the nearest station which may be upwards of 100 kilometres away (Nalder and Wein, 1998). Weather stations are discrete locations, and as recommended by Hartkamp *et al.* (1999), the use of rigorous spatial interpolation techniques can be an accurate way of estimating climate values at unsampled locations.

Confronted by an identical problem of data scarcity in the Canadian climate records, Nalder and Wein (1998) used spatial interpolation techniques in a geographic information systems (GIS) environment to determine climate normals for locations in Canada's boreal forest. Several interpolation methods were compared in this study as the authors assert that no interpolation technique is optimal in all circumstances (Nalder and Wein, 1998). Kriging techniques were emphasized in the paper, as it is as a well-proven method that limits bias and is capable of providing error estimates for interpolated values (Nalder and Wein, 1998). Using known values, kriging weights each value based on its spatial arrangement which is then used to determine the value at unknown points (Hartkamp *et al.*, 1999).

Following the advice of Nalder and Wein (1998), kriging - using ArcMap 9 software - was chosen for the interpolation of number of days with precipitation greater than 0.254mm, average wind speed, snow depth, and percentage of time with wind speed greater than 19.3km/h at the exact location of the three quarries. Data were available from 67 Environment Canada weather stations located in an area between 45°01' N and 46°48' N in latitude, and between 71°04' W and 74°38' W in longitude. Of these 67 stations, all reported precipitation, 54 reported snow depth values, only 12 reported average wind speeds and none reported a meaningful percentage of time with wind speed greater than 19.3km/h values, the latter two cases reinforcing the magnitude of the data scarcity problem.

Two important assumptions have been made in using kriging. Firstly, it is assumed that both average wind speed and the number of days with wind speed greater than 0.254 mm are stationary variables, meaning that their values are dependant solely on the distance between measurements and are not a property of their location (Hartkamp *et al.*, 1999). Because the 67 weather stations are located at elevations between 9m and 564m above mean sea level it is likely that this assumption is not perfectly satisfied. Secondly, it is assumed that the semivariogram depends only on the magnitude of the variable's value, and not on its direction (Hartkamp *et al.*, 1999). This assumption is reasonably valid as, in this case study, the weather stations are all located within the bounds of a small geographic area, and with small numbers of data points; no meaningful trends were expected.

7 Results and Discussion

This chapter discusses the results of the fugitive dusts emissions calculations for three subject quarries. The emissions values as calculated using 30-year average climate data are compared to those calculated using daily/hourly climate data to determine the effects of increasing temporal resolution of the calculations. Next, the results of the uncertainty assessment of the material properties are discussed, centred on the implications of using typical values in the calculations. The final section focuses on the emissions calculated with interpolated site specific climate averages, and discusses whether using spatially-relevant climate data is an improvement to using the "nearest station" approach to select data.

7.1 Quarry Data and Descriptions

Quarry descriptions and operational data collected from three quarries in southern Quebec operated by a major Canadian lime producer are presented in subsections 7.1.1-7.1.3. It was assumed that all operational and production data were accurate; however, as Marchi and Hamilton (2006) explain, there might be considerable bias and optimistically low values when plants self-report pollutants to the Toxics Release Inventory. While a legitimate issue in this research, Marchi and Hamilton (2006) focused on the final pollutant emission values which may negatively affect public perception of the company rather than biases in the primary and intermediate values used to calculate the emissions. Because the quarry managers who provided the data likely had little knowledge of the emissions calculation methodology, it is improbable that the values reported were purposely altered to lower their emissions.

One exception may be in unpaved road dust control practices, as the road-wetting frequency may have been exaggerated had the quarry been out of compliance with corporate standards. Although each of the three sites has distinct local management and similar operations, the reported value for road watering at Quarry J was lower than the other two sites; it was unknown whether the company has standard procedures, and it was beyond the scope of this research to evaluate internal compliance. Without having conducted a site visit, it is unclear whether any embellishment occurred. This exposes an important limitation in the data, as it is entirely second-hand, and was not verified with a site visit. It is clear that having verified, accurate operational data is ideal, but it is not necessary for the exploration of the material properties and climate data choices which is the primary goal of this research.

7.1.1 Quarry B

Quarry B is the furthest south of the three sites, located near the Vermont border, roughly between Montreal and Sherbrooke. During 2007, 1 360 000 tonnes of stone were quarried. Of this total, 800 000 tonnes were of sufficient quality to be processed to lime and 200 000 tonnes were shipped as construction stone. The remaining 360 000 tonnes of waste rock were added onto two sound and sight barriers – the only two storage piles at the quarry. The two barriers had approximate surface areas of 27 000 m^2 and 88 000 m^2 respectively.

The high quality limestone is hauled to the on-site crushing facilities by a pair of haul trucks which are filled by one of two loaders. The site also employs two lighter haul

trucks, a supervisor vehicle and a transport truck for lime kiln dust (LKD). See Table A.1 in the Appendix A for the vehicle fleet specifications and mileage during 2007. 30 000 tonnes of LKD – a by-product from the processing plant on-site – was dumped into a pit as backfill during the year. The roads at this site were reported watered "several" times per day, so it was assumed that this meant more than twice per day.

7.1.2 Quarry J

Quarry J is located between Montreal and Trois Rivières, and is the northernmost of the three quarries. In 2007, 1 014 057 T of stone were quarried, of which 333 231 T were of sufficient quality to processed into lime, 620 826 T were used as construction stone, and the remaining 60 000 T were too large for processing and put aside. 22 786 T of coal and 8 159 T of coke were used to fuel the kiln of the on-site processing plant. Two conical coal piles and four conical coke piles are located outdoors at the quarry with surface areas ranging from 426 m^2 to 1223 m^2. 17 123 T of LKD from the processing plant were dumped into an on-site pond with an estimated surface area of 35 000 m^2.

The site operates with two haul trucks, three loaders, a supervisor vehicle, a service truck for the drills, and a water truck. According to management, the unpaved roads are watered twice per day. The mean vehicle weights and estimated vehicle kilometres traveled during 2007 are reported in Table A.2 in the Appendix A.

7.1.3 Quarry M

Quarry M is located between Sherbrooke and Trois Rivières, and is the furthest west of the three quarries investigated. It is the largest site, with three active quarry faces, and

two others that were operational in the past. During 2007, 1 428 146 T of stone were quarried, of which 425 244 T were processed to lime, 236 946 T were used as aggregate, and 765 956 T of waste rock that were loaded onto seven barriers located around the active quarries. These barriers have estimated surface areas ranging from 1856 m^2 to 21 750 m^2. 14 775 T of LKD were dumped into two on-site LKD ponds.

The vehicle fleet at this site consists of one haul truck, four loaders, one mobile drilling vehicle, one 18-wheel transport truck, one maintenance truck, and a supervisor vehicle. Site management claimed water was sprayed "continuously". It was assumed that this meant more than twice per day. The mean vehicle weights and vehicle kilometres traveled during 2007 are reported in Table A.3 in Appendix A.

7.2 Climate Data Sources and Descriptions

The majority of climate data used in this study was collected from the National Climate Data and Information Archive (NCDIA). Unfortunately, none of the quarries were located in towns with weather stations that reported the 30-year climate averages - nor daily or hourly values - for the variables needed for the predictive emissions factor equation.

Because data are sparsely available for southern Quebec, populating variables using the nearest station method would have involved making daring assumptions that urban weather stations upwards of 100km away from rural quarries would be an adequate representation of wind speeds and precipitation. Thus applying the "What If" approach

described in Chapter 6, the PM emissions from each quarry were calculated as if each had been sited in Montreal, Sherbrooke and Trois Rivières rather than their small rural towns.

7.2.1 Thirty-Year Climate Averages

All 30-year climate averages (average wind speed, number of days with precipitation greater than 0.254mm, and average monthly snow depth) were collected from the "Climate Normals" section of the NCDIA, with the exception of average wind speed in Trois Rivières. This value was collected from Windfinder.com – a real-time wind monitoring website - as the 30-year average was unavailable from Environment Canada. It was measured between March 2007 and January 2009. Although not ideal, it is conceivable that a quarry may seek other sources for climate data when Environment Canada does not suffice.

Using the modified method for calculating the percentage of time with wind speed greater than 19km/h, this variable was found to be much greater than zero at all three quarries as shown in Table 3. This finding indicates that the method advocated by the NPRI is flawed, and following it would result in a drastic under-calculation of emissions from storage piles. Snow depths and average wind speeds were readily available.

Table 3: Thirty-year average climate data used for the calculations

Station	Av. Wind Speed (m/s)	# Days precip > 0.254mm	% Time wind speed > 19.3km/hr	# Months w/ snow depth > 15cm
Montreal	3.97	163	35.2	2
Sherbrooke	2.61	192	16.6	4
Trois Rivières	4.12	156	26	4

7.2.2 Daily and Hourly Climate Data

The array of daily precipitation values, daily snow depths, and hourly wind speeds was nearly complete from NCDIA, with the exception of Sherbrooke where daily snow depths were unavailable. In that case, the number of months with average snow depth greater than 15cm from the "Climate Normals and Averages" was substituted into calculations. The percentage-of-time values calculated with the modified methodology in section 6.1.2 are shown in Table 4.

Table 4: Percentages of time with wind speed greater than 19.3km/hr used for daily/hourly calculations

Station	% Time wind speed > 19.3km/hr
Montreal	35.2
Sherbrooke	16.6
Trois Rivières	26

It is not practical to include a table of all daily and hourly climate data used; readers are referred to the 2007 daily and hourly bulk data spreadsheets available from "Climate Data Online" of NCDIA for "Montreal/Pierre Elliott Trudeau Intl A", "Sherbrooke A", and "Trois Rivières Aqueduc."

7.3 Increasing Temporal Resolution

In this research, the effects of increasing the temporal resolution of fugitive dust emission calculations were tested by comparing the NPRI/AP-42 methodology that used 30-year climate averages with an altered methodology that incorporated daily and hourly (hereafter called "daily/hourly") climate data. Both calculations followed a "What If" approach as discussed in Chapter 6, where data from three different weather stations were applied to three quarries in southern Quebec. Only emissions from traffic on unpaved

roads, material handling operations, and wind erosion from storage piles were included in calculations.

The emissions values using 30-year climate averages are presented in Table 5, and the values using daily/hourly climate data are in Table 6. The material properties were held constant for the two analyses as the weighted means of the typical material properties.

Table 5: Emissions calculated using 30-year climate averages

			Emissions in Tonnes		
Site	**Climate Data**	**Size Fraction**	**Unpaved Roads**	**Material Handling**	**Storage Piles**
Quarry B	Montreal	$PM_{2.5}$	6.99	0.29	9.61
		PM_{10}	69.95	1.91	24.02
		TPM	240.45	4.05	48.04
	Sherbrooke	$PM_{2.5}$	3.92	0.17	3.88
		PM_{10}	39.25	1.11	9.71
		TPM	134.92	2.35	19.42
	Trois Rivières	$PM_{2.5}$	4.74	0.30	7.34
		PM_{10}	47.42	2.01	18.36
		TPM	162.99	4.24	36.72
Quarry J	Montreal	$PM_{2.5}$	11.65	0.17	0.40
		PM_{10}	116.50	1.14	1.00
		TPM	400.47	2.40	1.99
	Sherbrooke	$PM_{2.5}$	7.00	0.10	0.16
		PM_{10}	69.99	0.66	0.40
		TPM	240.60	1.39	0.81
	Trois Rivières	$PM_{2.5}$	8.46	0.18	0.30
		PM_{10}	84.56	1.19	0.76
		TPM	290.66	2.52	1.52
Quarry M	Montreal	$PM_{2.5}$	7.48	0.37	7.98
		PM_{10}	74.82	2.44	19.95
		TPM	257.21	5.16	39.90
	Sherbrooke	$PM_{2.5}$	4.20	0.21	3.23
		PM_{10}	41.98	1.42	8.07
		TPM	144.32	2.99	16.13
	Trois Rivières	$PM_{2.5}$	5.07	0.39	6.10
		PM_{10}	50.72	2.56	15.25
		TPM	174.35	5.41	30.50

Table 6: Emissions calculated using daily/hourly climate averages

Site	Climate Data	Size Fraction	Emissions in Tonnes		
			Unpaved Roads	Material Handling	Storage Piles
Quarry B	Montreal	$PM_{2.5}$	12.06	0.35	8.54
		PM_{10}	120.59	2.28	21.36
		TPM	414.53	4.82	42.71
	Sherbrooke	$PM_{2.5}$	6.77	0.21	2.83
		PM_{10}	67.75	1.36	7.06
		TPM	232.88	2.88	14.13
	Trois Rivières	$PM_{2.5}$	7.88	0.26	6.01
		PM_{10}	78.76	1.74	15.02
		TPM	270.73	3.68	30.04
Quarry J	Montreal	$PM_{2.5}$	19.87	0.20	0.35
		PM_{10}	198.72	1.35	0.89
		TPM	683.11	2.86	1.77
	Sherbrooke	$PM_{2.5}$	12.17	0.12	0.12
		PM_{10}	121.72	0.81	0.29
		TPM	418.43	1.71	0.59
	Trois Rivières	$PM_{2.5}$	13.85	0.16	0.25
		PM_{10}	138.51	1.03	0.62
		TPM	476.15	2.18	1.25
Quarry M	Montreal	$PM_{2.5}$	12.90	0.44	7.09
		PM_{10}	129.00	2.91	17.74
		TPM	443.42	6.15	35.47
	Sherbrooke	$PM_{2.5}$	7.25	0.26	2.35
		PM_{10}	72.47	1.74	5.87
		TPM	249.11	3.68	11.73
	Trois Rivières	$PM_{2.5}$	8.42	0.34	4.99
		PM_{10}	84.25	2.22	12.47
		TPM	289.60	4.69	24.95

Table 7: Percentage differences between daily/hourly and 30-year average calculations

Site	Climate Data	Size Fraction	Percentage Difference (Daily/Hourly – 30-Year Averages) Unpaved Roads	Material Handling	Storage Piles
Quarry B	Montreal	$PM_{2.5}$	42.00%	16.01%	-12.48%
		PM_{10}	42.00%	16.01%	-12.48%
		TPM	42.00%	16.01%	-12.48%
	Sherbrooke	$PM_{2.5}$	42.07%	18.64%	-37.50%
		PM_{10}	42.07%	18.64%	-37.50%
		TPM	42.07%	18.64%	-37.50%
	Trois Rivières	$PM_{2.5}$	39.80%	-15.31%	-22.25%
		PM_{10}	39.80%	-15.31%	-22.25%
		TPM	39.80%	-15.31%	-22.25%
Quarry J	Montreal	$PM_{2.5}$	41.37%	16.01%	-12.48%
		PM_{10}	41.37%	16.01%	-12.48%
		TPM	41.37%	16.01%	-12.48%
	Sherbrooke	$PM_{2.5}$	42.50%	18.64%	-37.50%
		PM_{10}	42.50%	18.64%	-37.50%
		TPM	42.50%	18.64%	-37.50%
	Trois Rivières	$PM_{2.5}$	38.95%	-15.31%	-22.25%
		PM_{10}	38.95%	-15.31%	-22.25%
		TPM	38.95%	-15.31%	-22.25%
Quarry M	Montreal	$PM_{2.5}$	42.00%	16.01%	-12.48%
		PM_{10}	42.00%	16.01%	-12.48%
		TPM	42.00%	16.01%	-12.48%
	Sherbrooke	$PM_{2.5}$	42.07%	18.64%	-37.50%
		PM_{10}	42.07%	18.64%	-37.50%
		TPM	42.07%	18.64%	-37.50%
	Trois Rivières	$PM_{2.5}$	39.80%	-15.31%	-22.25%
		PM_{10}	39.80%	-15.31%	-22.25%
		TPM	39.80%	-15.31%	-22.25%

7.3.1 Unpaved Roads

As expected, emissions from unpaved roads had the largest mass at all three quarries, regardless of the resolution of the climate data. Table 7 shows that emissions calculated using daily/hourly data were between 39% and 43% higher than those calculated using 30-year averages which is attributable to differences in the abatement factor for

precipitation as well as to the increased breadth of values when averaging intervals are reduced. In the case of daily/hourly calculations, the threshold-based abatement factors from the NPRI's Industrial Unpaved Roads Calculator (see Table 2) were applied to each daily emission factor presented in Chapter 6. However, in the calculation using the 30-year averages, abatement was assumed to be 100% when more than 0.254mm of precipitation fell as is done in the NPRI's Simplified Industrial Unpaved Roads Calculator and is advocated by WRAP (2006). According to WRAP (2006), using the value of 0.254mm is a reasonable simplification, but it recommends using finer temporal and spatial resolution to account for changes in the moisture content of road surface material. Interestingly, this suggestion would require a new estimation method as currently only the silt content of the road is considered in the equation.

Calculating unpaved road emissions with daily/hourly data adds a degree of realism and precision that cannot be achieved using 30-year climate averages, even if the WRAP (2006) suggests that the accuracy of such an approach has not been extensively studied. One way of improving the 30-year averages calculation would be harmonizing the precipitation abatement factors with the thresholds used in the daily/hourly method; this means an assumption of 100% abatement only after 25mm of precipitation and thus counting the number of days with precipitation greater than 25mm rather than the current threshold 0.254mm. Although this ignores other precipitation values that would surely abate emissions, it would add a degree of conservativism which is a necessity when the calculation is purposely simplified.

As the primary source of fugitive dust emissions from quarries, unpaved road emissions are the most important to quantify accurately. Fortunately for quarry operators in Canada, the NPRI's Industrial Unpaved Road Dust Calculator automates the calculation using daily precipitation and snow depth data, and this tool is freely available for use. Using the calculator requires only moderately more effort to parameterize than the simplified version, but the issue is whether quarry operators would voluntarily report higher emissions to the NPRI as long as the simplified calculation method remains an acceptable practice.

7.3.2 Material Handling

The results of the comparison between calculations using daily/hourly data for material handling were less conclusive than those seen for the unpaved roads. When calculating emissions at all three quarries using hourly wind speeds from Montreal and Sherbrooke, the fugitive dust emissions were 16.01% and 18.64% higher, respectively, than the same calculations using 30-year averages, as shown in Table 7. A conflicting result was seen when hourly data from Trois Rivières was used, as the emissions were 15.31% lower at all quarries. The percentage differences depend only on the weather data used, and not on the magnitude of material handling operations at each quarry operations, as was expected. Using hourly wind speeds highlights peakedness in the variations more effectively than monthly averages.

These results can be explained by the average wind speeds in 2007 as calculated from the hourly data. In Trois Rivières, wind speeds during 2007 were slightly lower than the 30-

year average and thus daily/hourly emissions were lower, whereas data for Montreal and Sherbrooke had wind speeds above the 30-year average which resulted in higher daily/hourly emissions. Because average wind speeds from 2007 calculated using hourly data predicted whether the daily/hourly calculation would be higher or lower than the 30-year average calculation, a further investigation was conducted to determine whether using this simpler metric in the existing AP-42 methodology replaces the need to calculate emissions on an hourly basis as daily/hourly method commands.

To answer this new question, emissions for materials handling were recalculated for all sites using the 30-year climate averages methodology, but with the 2007 annual average wind speeds from each weather station substituted into each calculation. When comparing the new results to those from the daily/hourly analysis, the emissions calculated with 2007 average wind speeds were all lower; once again the percentage differences depended only on the weather data used and not on the specific material handling operations at each quarry. See the results in Table 8:

Table 8: Percentage differences between materials handling emissions calculated using 2007 climate averages and 2007 daily/hourly data

			Percentage Difference (2007 Daily/Hourly – 2007 Averages)
Site	**Climate Data**	**Size Fraction**	**Material Handling**
All Sites	Montreal	$PM_{2.5}$	7.04%
		PM_{10}	7.04%
		TPM	7.04%
	Sherbrooke	$PM_{2.5}$	10.67%
		PM_{10}	10.67%
		TPM	10.67%
	Trois Rivières	$PM_{2.5}$	1.41%
		PM_{10}	1.41%
		TPM	1.41%

Although the percentage differences of 7.04% and 10.67% seen with the Montreal and Sherbrooke data may be substantial on a mass basis at quarries with large material handling operations, the highest mass differences were observed at Quarry M using Montreal data and were 0.4 T of TPM, 0.19 T of PM_{10}, and 0.03 T of $PM_{2.5}$. These small differences are insignificant on the scale of the whole inventory. Although calculating emissions from material handling operations using daily/hourly data is more precise, the extra effort required to set up the calculations that use hourly wind speeds – in absence of an NPRI calculator – would seem to be unnecessary at these quarries where material handling is responsible for only a small fraction of the fugitive dust emissions.

While using hourly wind speeds for the material handling should be considered optional for these quarries, the percentage differences between the calculations using 2007 average wind speeds and the 30-year averages merit discussion. The results of the comparison are in Table 9:

Table 9: Percentage differences between material handling emissions calculated using 2007 climate averages and 30-year climate averages

			Percentage Difference (2007 Averages – 30 Year Averages)
Site	**Climate Data**	**Size Fraction**	**Unpaved Roads**
All Sites	Montreal	$PM_{2.5}$	10.10%
		PM_{10}	10.10%
		TPM	10.10%
	Sherbrooke	$PM_{2.5}$	9.96%
		PM_{10}	9.96%
		TPM	9.96%
	Trois Rivières	$PM_{2.5}$	-16.93%
		PM_{10}	-16.93%
		TPM	-16.93%

These percentage differences are more drastic than the small mass differences they represent, but logic dictates that when preparing an emissions inventory for any particular year, data from that particular year should be used. The percentage differences represent the error that can be easily avoided by simply calculating a temporally-relevant yearly average wind speed rather than using the 30-year average, and in addition provide justification for Environment Canada to improve their climate database. This principle follows from Vallack and Shillito's (1998) statement that monthly means are better measures for calculating emissions than annual means; and could be expanded to suggest that monitoring wind speed values for each second would be better yet than hourly averages.

7.3.3 Storage Piles

Fugitive dust emissions from storage piles using daily/hourly data were consistently lower than the totals calculated using 30-year averages for which there are two possible explanations. One explanation is that the two methodologies calculated the percentage of time with wind speeds greater than 19.3km/h in slightly different ways. This value was consistently lower when calculated using the daily/hourly methodology which therefore lowers the calculated emissions. A second reason is that the number of days with precipitation greater than 0.254mm in daily/hourly calculations considered daily precipitation values from 2007 only. If 2007 had had a higher number of days than the 30-year average, this difference would lower the calculated emissions values. However, both Montreal and Trois Rivières were drier than average in 2007, casting doubt on the importance of precipitation in the calculations.

Table 10: Relationship between the 2007 average and 30-year averages for percentage of time with wind speed greater than 19.3km/h and number of days with precipitation greater than 0.254mm and the resulting percentage differences in emissions

	2007-averaged % of time with wind speed greater than 19.3km/h		# of days in 2007 with precipitation greater than 0.254km/h		
Weather Data	**> 30-yr Average**	**< 30-yr Average**	**> 30-yr Average**	**< 30-yr Average**	**Percentage Difference in Emissions (Daily/Hourly – 30-yr Averages)**
Montreal		■		■	-12.48%
Sherbrooke		■	■		-37.50%
Trois Rivières		■		■	-22.50%

From Table 10 it can be seen that Sherbrooke was the only station that was wetter than average during 2007, and accordingly, the calculations using Sherbrooke data had the greatest percentage difference in emissions. Montreal and Trois Rivières were drier than average in 2007, yet still had much lower emissions because of the difference in the percentage-of-time calculation. This suggests that emissions are more sensitive to the determination of percentage of time with wind speed greater than 19.3km/h which is expected based on the formula.

7.4 Uncertainty Assessment

Monte Carlo simulation was used to assess the uncertainty introduced when using typical material properties from AP-42. As discussed in Chapter 6, the emissions calculations using climate averages for each quarry and weather station combination were recalculated with probability density functions (PDFs) to represent the range and likelihood of material properties. A summary of PDFs can be found in Table 11; detailed descriptions are found in Appendix C.

Table 11: Summary of Probability Density Function Parameters by Material

Material	Property	Shape	Parameters
Unpaved Roads	Silt Content	Lognormal	$\mu = 9.3$; $\sigma = 7.7$
Limestone	Silt Content	Lognormal	$\mu = 4.46$; $\sigma = 5.37$
	Moisture Content	Uniform	Min = 0.2; Max = 5.0
Coal	Silt Content	Lognormal	$\mu = 3.88$; $\sigma = 3.62$
	Moisture Content	Lognormal	$\mu = 5.48$; $\sigma = 1.36$
Coke	Silt Content	Uniform	Min = 4.4; Max = 5.4
	Moisture Content	Uniform	Min = 6.4; Max = 9.2
Lime Kiln Dust	Moisture Content	Uniform	Min = 14.2; Max = 37.4

7.4.1 Simulation Results

Simulations for each combination of quarry and weather station were conducted individually, and the $PM_{2.5}$, PM_{10}, and TPM emissions from unpaved roads, material handling, and storage piles were calculated separately. From each quarry and weather station combination, two values were collected for each fugitive emission source:

1. The simulated mean PM emissions and their 95% confidence limits (Table 12)
2. The probability that the PM emissions are higher than the value calculated using a weighted mean of typical material properties; namely the results found with 30-Year Climate Averages (Table 13)

Table 12 Simulated mean emissions with 95% confidence limits and, in brackets, the confidence limits expressed as a percentage difference from the simulated mean.

			Unpaved Roads			Material Handling			Storage Piles		
Site	Climate Data	Size Fraction	Lower 95% C.L.	Mean (T)	Upper 95% C.L.	Lower 95% C.L.	Mean (T)	Upper 95% C.L.	Lower 95% C.L.	Mean (T)	Upper 95% C.L.
Quarry B	Montreal	$PM_{2.5}$	1.46 (-77.85%)	6.59	19.31 (193.02%)	0.09 (-85.25%)	0.61	4.2 (588.52%)	0.99 (-89.92%)	9.82	41.07 (318.23%)
		PM_{10}	14.63 (-77.82%)	65.95	193.09 (192.78%)	0.6 (-85.22%)	4.06	27.72 (582.76%)	2.47 (-89.94%)	24.56	102.69 (318.12%)
		TPM	71.18 (-67.83%)	221.24	529.67 (139.41%)	1.26 (-85.31%)	8.58	58.6 (582.98%)	4.95 (-89.92%)	49.12	205.37 (318.10%)
	Sherbrooke	$PM_{2.5}$	0.82 (-77.90%)	3.71	10.01 (169.81%)	0.05 (-87.50%)	0.4	2.92 (630.00%)	0.4 (-89.61%)	3.85	14.48 (276.10%)
		PM_{10}	8.17 (-78.01%)	37.15	100.06 (169.34%)	0.35 (-86.69%)	2.63	19.26 (632.32%)	1.01 (-89.50%)	9.62	36.21 (276.40%)
		TPM	39.82 (-68.06%)	124.69	279.36 (124.04%)	0.73 (-86.85%)	5.55	40.73 (633.87%)	2.01 (-89.55%)	19.24	72.41 (276.35%)
	Trois Rivières	$PM_{2.5}$	0.93 (-79.38%)	4.51	11.87 (163.19%)	0.09 (-86.36%)	0.66	4.95 (650.00%)	0.74 (-89.34%)	6.94	27.38 (294.52%)
		PM_{10}	9.3 (-79.36%)	45.06	118.71 (163.45%)	0.62 (-85.71%)	4.34	32.71 (653.69%)	1.85 (-89.33%)	17.34	68.46 (294.81%)
		TPM	45.9 (-64.67%)	151.28	332.78 (119.98%)	1.31 (-85.71%)	9.17	69.15 (654.09%)	3.7 (-89.33%)	34.68	136.92 (294.81%)
Quarry J	Montreal	$PM_{2.5}$	2.84 (-74.87%)	11.3	34.49 (205.22%)	0.06 (-82.86%)	0.35	1.71 (388.57%)	0.3 (-25.00%)	0.4	0.67 (62.50%)
		PM_{10}	28.36 (-74.91%)	113.03	344.92 (205.16%)	0.41 (-82.40%)	2.33	11.27 (383.69%)	0.74 (-26.00%)	1	1.68 (65.00%)
		TPM	133.44 (-64.67%)	377.73	931.55 (146.62%)	0.86 (-82.56%)	4.93	23.83 (383.37%)	1.49 (-25.50%)	2	3.36 (62.96%)
	Sherbrooke	$PM_{2.5}$	1.6 (-75.46%)	6.52	18.4 (182.21%)	0.04 (-80.95%)	0.21	1.02 (385.71%)	0.12 (-25.00%)	0.16	0.26 (62.50%)
		PM_{10}	16.01 (-75.45%)	65.22	183.96 (182.06%)	0.23 (-83.21%)	1.37	6.74 (391.97%)	0.3 (-25.00%)	0.4	0.66 (65.00%)
		TPM	76.37 (-65.32%)	220.19	510.16 (131.69%)	0.49 (-83.10%)	2.9	14.25 (391.38%)	0.61 (-25.69%)	0.81	1.32 (62.96%)
	Trois Rivières	$PM_{2.5}$	2.02 (-75.46%)	8.23	22.86 (177.76%)	0.06 (-85.00%)	0.4	2.06 (415.00%)	0.23 (-25.81%)	0.31	0.51 (64.52%)
		PM_{10}	20.16 (-75.50%)	82.3	228.63 (177.80%)	0.43 (-83.52%)	2.61	13.62 (421.84%)	0.56 (-27.27%)	0.77	1.27 (64.94%)
		TPM	95.32 (-65.34%)	275.05	630.06 (129.07%)	0.9 (-83.70%)	5.52	28.79 (421.55%)	1.13 (-26.62%)	1.54	2.53 (64.29%)
Quarry M	Montreal	$PM_{2.5}$	1.48 (-79.75%)	7.31	21.32 (191.66%)	0.12 (-84.81%)	0.79	5.8 (634.18%)	0.85 (-89.25%)	7.91	33.33 (321.37%)
		PM_{10}	14.82 (-79.73%)	73.1	213.24 (191.71%)	0.76 (-85.47%)	5.23	38.32 (632.70%)	2.13 (-89.23%)	19.77	83.33 (321.50%)
		TPM	72.99 (-70.02%)	243.5	580.81 (138.53%)	1.61 (-85.43%)	11.05	80.94 (632.49%)	4.26 (-88.95%)	38.54	166.66 (332.43%)
	Sherbrooke	$PM_{2.5}$	0.92 (-77.28%)	4.05	11.78 (190.86%)	0.07 (-82.93%)	0.41	2.81 (585.37%)	0.3 (-90.96%)	3.32	12.97 (290.66%)
		PM_{10}	9.22 (-77.21%)	40.46	117.77 (191.08%)	0.44 (-83.82%)	2.72	18.57 (582.72%)	0.76 (-90.85%)	8.31	32.43 (290.25%)
		TPM	44.37 (-67.20%)	135.27	321.92 (137.98%)	0.93 (-83.83%)	5.75	39.26 (582.78%)	1.52 (-90.85%)	16.62	64.85 (290.19%)
	Trois Rivières	$PM_{2.5}$	1.18 (-75.72%)	4.86	14.1 (190.12%)	0.12 (-85.88%)	0.85	5.71 (571.76%)	0.58 (-90.75%)	6.27	25.15 (301.12%)
		PM_{10}	11.77 (-75.80%)	48.64	141 (189.88%)	0.79 (-85.99%)	5.64	37.71 (568.62%)	1.46 (-90.69%)	15.68	62.87 (300.96%)
		TPM	55.96 (-65.66%)	162.95	386.18 (136.99%)	1.67 (-86.00%)	11.93	79.72 (568.23%)	2.91 (-90.72%)	31.37	125.73 (300.80%)

When observing the results of the uncertainty assessment, the most striking – but expected - observations were the wide 95% confidence limits. Little literature exists for direct comparison, however Monni *et al.* (2004) report that the industry-wide emissions in Finland had confidence limits of -27% to +43% around the mean, and also suggested that individual sites likely had higher degrees of uncertainty.

Large confidence intervals around the results do not suggest that the calculations are incorrect, but rather that there is a distinct opportunity to improve the precision of emissions inventories. The validity of the assessment relies on several assumptions. It was assumed that silt and moisture contents from AP-42 tables are representative of the true range of values. Secondly, the shapes and parameters of the PDFs were assumed to be reasonable representations of the material's properties. It was also assumed that the predictive emissions factor equations and use of 30-year averages accurately quantify the emissions for the site. With all conditions true, there is 95% confidence that emissions fall at a point within the confidence limits; but with such uncertainty about the accuracy of the calculation, providing these emissions values to the public can be misleading.

Reporters may attempt to make an educated choice from the typical values provided in AP-42 and assume it to be true, however the small sample sizes and the lack of contextual information about sample locations make that impossible. The assessment shows that considering any single value from the table as "typical" for a quarry is making a daring assumption, and that without conducting any on-site sampling for confirmation, the calculation of emissions is almost certain to be inaccurate.

By placing the results of the 30-year averages method calculations onto the distribution of simulation results – see Figures 1 and 2 for examples – the probabilities that the use of weighted mean material properties would underestimate emissions were established. The results, in Table 13 below, show that using weighted average material properties instead of conducting on-site sampling is actually more likely to produce conservative overestimates of fugitive dust emissions.

Table 13: The probability that the PM emissions are higher than the value calculated using a weighted mean of typical material properties

			Probability of Exceedance		
Site	**Climate Data**	**Size Fraction**	**Unpaved Roads**	**Material Handling**	**Storage Piles**
Quarry B	Montreal	$PM_{2.5}$	33.79%	39.69%	31.92%
		PM_{10}	33.78%	39.71%	31.92%
		TPM	33.78%	39.69%	31.92%
	Sherbrooke	$PM_{2.5}$	34.65%	42.98%	32.38%
		PM_{10}	34.48%	43.39%	32.27%
		TPM	34.48%	43.34%	32.27%
	Trois Rivières	$PM_{2.5}$	35.62%	41.21%	30.63%
		PM_{10}	35.61%	40.60%	30.62%
		TPM	35.61%	40.24%	30.62%
Quarry J	Montreal	$PM_{2.5}$	33.19%	50.85%	36.90%
		PM_{10}	33.19%	50.13%	36.90%
		TPM	33.19%	50.45%	37.63%
	Sherbrooke	$PM_{2.5}$	33.73%	51.23%	37.39%
		PM_{10}	33.74%	51.26%	37.39%
		TPM	33.73%	51.44%	34.59%
	Trois Rivières	$PM_{2.5}$	36.21%	51.66%	32.69%
		PM_{10}	36.21%	51.60%	36.47%
		TPM	36.21%	51.55%	36.47%
Quarry M	Montreal	$PM_{2.5}$	35.49%	42.02%	30.97%
		PM_{10}	35.49%	42.04%	30.97%
		TPM	35.48%	42.03%	30.97%
	Sherbrooke	$PM_{2.5}$	36.46%	39.39%	31.52%
		PM_{10}	36.46%	38.67%	31.53%
		TPM	36.47%	38.81%	31.54%
	Trois Rivières	$PM_{2.5}$	33.80%	42.65%	33.75%
		PM_{10}	33.79%	42.68%	33.75%
		TPM	33.79%	42.68%	33.75%

7.4.2 Unpaved Roads

The most extreme example of uncertainty in terms of masses of PM emitted - as opposed to considering the confidence limits as a percentage of the mean – were emissions from unpaved roads at Quarry J calculated using Montreal data. TPM emissions were equally likely, in statistical terms, to fall anywhere between 133.44 T or 931.55 T; PM_{10} emissions to be 28.34 T or 344.92 T; and $PM_{2.5}$ emissions to be 2.84 T or 34.49 T (see Table 12). As the major source of fugitive dust emissions from the quarry, establishing a site-specific value for silt content of unpaved roads is of crucial importance for improving the accuracy of the inventory. Collecting samples of the unpaved road material and having them analyzed in a laboratory would not be too costly, particularly when the quality of the NPRI report could be improved dramatically.

7.4.3 Material Handling

On a percentage basis, emissions from material handling consistently had the most uncertainty of the three calculations. Quarry B emissions calculated using Trois Rivières data are the most striking examples as the lower 95% confidence limits for the three size fractions of PM were approximately 86% below, and 650% above the mean. The upper confidence limits are skewed right because the emissions factor approaches infinity as moisture content approaches zero – a limitation of the predictive emissions factor equation. Therefore, when low values were randomly selected from the distribution during the simulation, extremely high emissions values were calculated.

Although values 650% higher than the mean may appear to be outliers, they are statistically equally likely to be as correct as the mean value. This source of emissions is

less important than unpaved roads on a mass basis; however, establishing moisture contents of materials being handled would also greatly improve the precision and accuracy of the inventory. As desirable as this would be, it may prove difficult for materials that are stored outdoors – as coal and coke were at Quarry J – or freshly blasted limestone whose moisture would fluctuate based on the amount of precipitation, local geology, and drainage in the area.

7.4.4 Wind Erosion of Storage Piles

The uncertainty observed in the wind erosion of storage pile calculations ranged from relative high certainty in the case of Quarry J to relatively high uncertainty at Quarries B and M. At Quarry J, the lower 95% confidence limits for all PM sizes was around 25% below the mean for all three weather stations, showing that there is more certainty in the mean values at Quarry J than that Quarries B and M where the lower limits were approximately 90% below the mean. Upper confidence limits showed a similar relationship, as Quarry J's upper limit is about 64% greater than the mean, versus those seen at Quarries B and M in the order of 300%.

The similarities and difference in uncertainties between the sites can be explained by the materials stored in piles at the three quarries, and the variability in the probability density functions (PDFs) used to represent the materials. Quarry J had six small storage piles of coal and coke and no piles of limestone, whereas both Quarries B and M had much larger barriers made of waste limestone and no piles of coal or coke. This is significant, as the simulations of Quarry B and M needed only one PDF – the silt content of limestone – and the two results had nearly identical confidence limits expressed as percentages of the

mean. Conversely, the Quarry J simulation required PDFs for the silt contents of both coal and coke and had much more certain results. While the higher surface area of the waste rock barriers at Quarries B and M explain the higher emission tonnage at these two sites, this does not contribute to the magnitude of uncertainty.

Table 14: Statistics of the three PDFs used in the wind erosion of storage piles calculations

	Coke (silt)	Coal (silt)	Limestone (silt)
Distribution Shape	Uniform	Lognormal	Lognormal
Mean	4.90	3.88	4.45
Median	4.90	2.84	2.85
Standard Deviation	0.29	3.61	5.23
Variance	0.08	13.07	27.35
Skewness	0.00	3.49	4.28
Kurtosis	1.80	27.97	35.99
Coefficient of Variability	0.0589	0.9318	1.18
Minimum	4.40	0.00	0.00
Maximum	5.40	100.00	100.00

As Table 14 shows, the lognormal distributions for coal and limestone silt content have very similar properties, therefore they do not explain the greater certainty at quarry J; the answer lies with the uniform distribution used for coke. At Quarry J there are four piles of coke versus two piles of coal, and because more of surface area is attributed to coke, the uniform distribution is dominant. The resulting distribution of simulated emissions has properties that fall between two distributions, but the model output retains a lognormal shape as shown in Figure 1. Even with this shape, the output distribution incorporated the negligible variance of the uniform distribution, and this improves the overall certainty of the calculation. The effects of the uniform distribution for the silt content of coke are especially pronounced because the maximum and minimum values of

4.4% and 5.4% were very close together and bounded the results within a small range of values. Because of these factors, the output of Quarry J had a standard deviation of 0.286; see Figure 1.

At Quarries B and M, only limestone was stored in piles which resulted in an outputted distribution that inherited all the variability of the lognormal distribution; thus calculations with more uncertainty as shown in Figure 2.

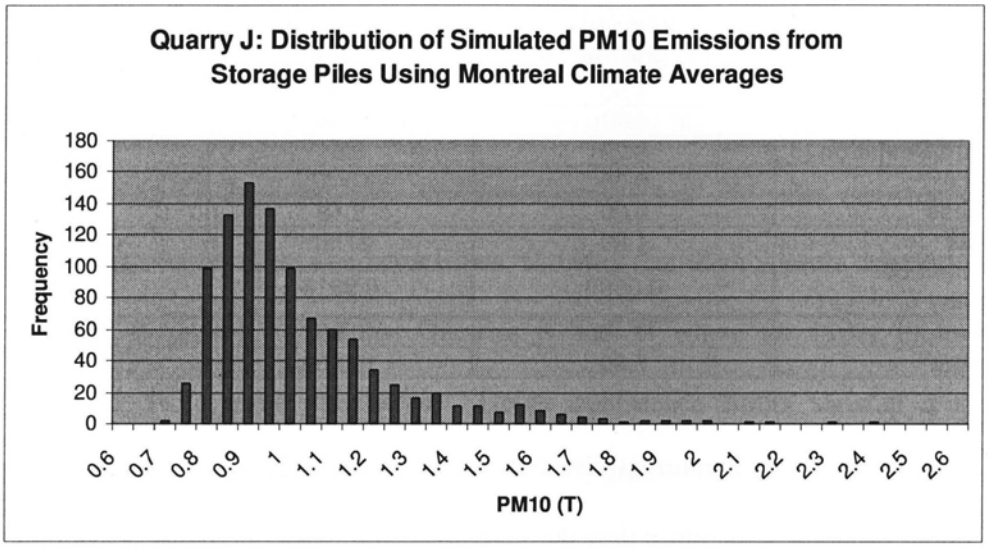

Figure 1: Distribution of simulated PM_{10} emissions from Quarry J

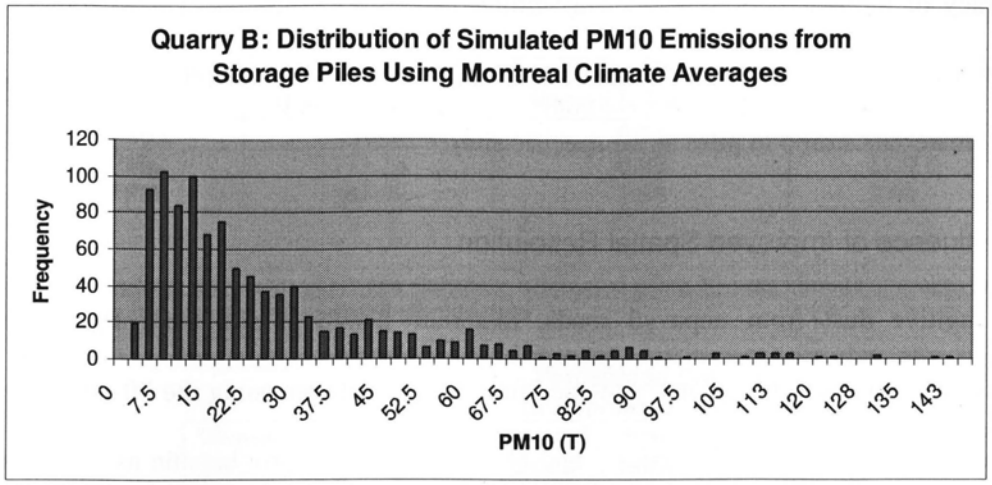

Figure 2: Distribution of simulated PM_{10} emissions from Quarry B

Although the results of the analysis show more certainty in emissions from storage piles at Quarry J than both Quarries B and M, this is mainly a product of subjective distribution choice in a situation with limited data availability. Had there been adequate data to make the assumption that the silt content of coke was also lognormally distributed, the uncertainty in the calculation for Quarry J would have been in the same order as Quarries B and M. It is interesting to note that having little data about the silt content of coke actually improved the certainty of the calculation. If reporters had collected and analyzed the properties of the coke themselves and knew that the distribution was truly uniform between the small range of 4.4% and 5.4%, this would represent a great degree of certainty, and a truly ideal situation when calculating emissions. Unfortunately, this range published by AP-42 is based on two samples and cannot be considered representative of all coke. While the uniform distribution technically adds certainty and precision to the calculations, it does not improve the accuracy of the site's fugitive dust emissions inventory. The only way to confirm

accuracy of the calculations - again assuming that the use predictive emissions factor equations produce accurate results for quarry application - is by sampling the silt content of the materials stored in piles at the specific site.

7.5 Influence of Improved Spatial Resolution

The fugitive dust from unpaved roads, materials handling, and storage piles were calculated using the interpolated site-specific climate averages using the 30-year climate averages methodology for Quarries J and M. Quarry B was problematic as it is located outside the area of spatial interpolation – therefore involving extrapolation – so neither the average wind speed nor percentage of time with wind speed greater than 19.3km/h could be predicted; therefore, it was excluded from this section of the study. Quarry M was also slightly problematic as predicting the percentage of time would have also involved extrapolation from the dataset, so the approach was to test three scenarios using High (Montreal), Medium (Trois Rivières) and Low (Sherbrooke) values for this parameter. All of the parameters were predicted for Quarry J. The interpolated climate parameters were predicted using the best available local climate data and are listed in Table D.1 in Appendix D accompanied by figures of the interpolated surfaces generated in ArcMap 9.

7.5.1 Interpolation with All Climate Parameters

The Quarry J emissions calculated with site-specific, interpolated values are shown below in Table 15. These results are compared with the emissions calculated with 30-year climate averages in Table 16.

Table 15: Emissions from Quarry J calculated with interpolated climate parameters

	Emissions in Tonnes		
	Unpaved Roads	Materials Handling	Storage Piles
$PM_{2.5}$	11.11	0.14	0.40
PM_{10}	111.13	0.93	1.00
TPM	382.02	1.98	2.00

Table 16: Percentage differences between emissions calculated using 30-year climate averages and emissions using site-specific interpolated climate data

		Percentage Difference (Interpolated – Average)		
Climate Data	Size Fraction	Unpaved Roads	Material Handling	Storage Piles
Montreal	$PM_{2.5}$	-4.83%	-21.64%	0.04%
	PM_{10}	-4.83%	-21.64%	0.04%
	TPM	-4.83%	-21.64%	0.04%
Sherbrooke	$PM_{2.5}$	37.02%	29.50%	59.58%
	PM_{10}	37.02%	29.50%	59.58%
	TPM	37.02%	29.50%	59.58%
Trois Rivières	$PM_{2.5}$	23.91%	-18.33%	23.59%
	PM_{10}	23.91%	-18.33%	23.59%
	TPM	23.91%	-18.33%	23.59%

The values in Table 16 show that the Quarry J emissions calculated with the interpolated values were closest to the calculation using averages from Montreal which is logical as this quarry is located closest to the Montreal station. The difference in emissions from storage piles is negligible, and those from unpaved roads are less than 5% which, in context of the uncertainty and error calculated in this study, represents a difference that can be considered negligible. The percentage difference of emissions from materials handling is more pronounced at -21.64%, but might be attributable to the small number of data points used for interpolating the average wind speed.

The closeness in values between emissions calculated at Quarry J using interpolated climate data and those calculated with the nearest station Montreal data might be

suggestive that there is little value in the interpolation. However, before drawing this conclusion it is important to consider that the storage pile calculations had nearly identical emissions values, yet both had markedly different values for number of days with precipitation greater than 0.254mm and percentage of time with wind speed greater than 19.3km/h. In this case, the two variables values balanced one another out and it was only by chance that the emissions values were almost identical.

Considering the larger differences between the emissions from all sources calculated with Trois Rivières data, and that this weather station is only 20km further away from the site than the Montreal station, this larger difference and relative closeness of the station suggests that interpolation may still be more accurate than the nearest station. However, without any field analysis, the accuracy cannot be judged.

Like other analyses in this study, the masses of emissions from storage piles and materials handling are insignificant compared to those from unpaved roads. The only climate parameter in the unpaved roads calculation – number of days with precipitation greater than 0.254mm – had the best availability of data and was likely the most accurate interpolated value. As shown in Table 16, using this value for the calculation would have lowered emissions by 4.83% versus nearest station Montreal data, which coincides with a saving of 0.54T of $PM_{2.5}$, 5.37T of PM_{10}, and 18.45T of TPM. The effort required to interpolate the climate data is not entirely merited from the perspective of a reporter as the increased accuracy is unlikely to force them over, or pull them under, the reporting thresholds. However, using interpolated data, in the absence of physically monitoring

on-site meteorology, might be more scientifically valid and thus improve the quality of NPRI emissions reports.

7.5.2 Interpolation with Missing Data

There were insufficient data to interpolate the percentage of time with wind speed greater than 19.3km/h at Quarry M, therefore the calculations proceeded with high (Montreal), medium (Trois Rivières) and low (Sherbrooke) scenarios for this variable; the other parameters were all able to be interpolated. The three cases are shown below in Table 17.

Table 17: Emissions from Quarry M calculated with interpolated climate parameters

		Emissions in Tonnes		
Percentage of Time with Wind Speed >19.3km/h	**Size Fraction**	**Unpaved Roads**	**Material Handling**	**Storage Piles**
High Scenario: Montreal (35.2%)	$PM_{2.5}$	5.44	0.28	7.01
	PM_{10}	54.41	1.83	17.53
	TPM	187.04	3.87	35.06
Medium Scenario: Trois Rivières (26.0%)	$PM_{2.5}$	5.44	0.28	5.18
	PM_{10}	54.41	1.83	12.95
	TPM	187.04	3.87	25.90
Low Scenario: Sherbrooke (16.6%)	$PM_{2.5}$	5.44	0.28	3.31
	PM_{10}	54.41	1.83	8.28
	TPM	187.04	3.87	16.55

As can be seen in Table 17, emissions from unpaved roads and material handling are independent of the high, medium and low scenarios so these values can be compared directly with the values calculated with 30-year climate averages, identical to the approach taken to Quarry J in the previous subsection. Results of this comparison are shown in Table 18 and discussed in the subsections following.

Table 18: Percentage differences between Quarry M emissions calculated using 30-year climate averages and emissions using site-specific interpolated climate data

		Percentage Difference (Interpolated – 30-Year Averages)	
Climate Data	**Size Fraction**	**Unpaved Roads**	**Material Handling**
Montreal	$PM_{2.5}$	-37.51%	-33.35%
	PM_{10}	-37.51%	-33.35%
	TPM	-37.51%	-33.35%
Sherbrooke	$PM_{2.5}$	22.84%	22.71%
	PM_{10}	22.84%	22.71%
	TPM	22.84%	22.71%
Trois Rivières	$PM_{2.5}$	6.78%	-29.73%
	PM_{10}	6.78%	-29.73%
	TPM	6.78%	-29.73%

Unpaved Roads

Table 18 shows that the Quarry M unpaved road emissions calculated with site-specific interpolated climate data was closest to the value calculated with Trois Rivières averages. This is counterintuitive considering that Sherbrooke is about 20km for the quarry whereas Trois Rivières is over 100km away. The comparison to Montreal is as expected, given that this station is furthest away from the site. For an explanation of the percentage differences and why the unexpected result occurred, the relationship between the interpolated values and 30-year averages is considered in Table 19.

Table 19: Relationships between interpolated climate data and the 30-year averages used for unpaved road emissions calculation

	Interpolated # of Months with Snow Depth > 15cm		Interpolated # of days with precipitation greater than 0.254mm		
Weather Data	**> 30-yr Average**	**< 30-yr Average**	**> 30-yr Average**	**< 30-yr Average**	**Percentage Difference in Emissions (Interpolated – 30-Year Averages)**
Montreal	■		■		-37.51%
Sherbrooke		■		■	22.84%
Trois Rivières		■	■		6.78%

When the interpolated climate variables are compared to the averages from the nearest station, Sherbrooke, the smaller number of months with snow depth greater than 15cm increases the calculated emissions, as does the smaller number of days with precipitation greater than 0.254mm. Compared to Trois Rivières, the interpolated number of months is also lower, but the increased precipitation has a decreasing effect on emissions. The two effects balance, and the overall percentage difference of emissions from the average value is smaller, despite being over 100km away from the quarry.

Material Handling

The results of the material handling calculation using interpolated data are as expected, with the values being closest to those from Sherbrooke, the nearest station. However the difference in Table 18 of 22.71% is quite large considering the relatively short distance between station and quarry, and that the percentage differences with the other stations that are over 100km away have similar magnitudes but in the negative direction. If the emissions using the site-specific, interpolated value are assumed to be accurate, calculating the emissions from Quarry M using the 30-year average wind speed from the nearest station has the potential to distort results from reality in the order of 20-30%. As was concluded in previous sections regarding materials handling, this source of emissions is insignificant on a mass basis relative to unpaved roads and storage piles, so extra effort on accurate quantification must be weighed against the benefits.

Storage Piles

As introduced earlier, there were insufficient data to predict a site-specific value for the percentage of time with wind speed greater than 19.3km/h, a key variable for the wind erosion from storage pile calculation (Equation 3). The results of the high, medium, and low scenarios are presented for comparison to the values calculated using the 30-year averages in Table 20. For simplicity, only TPM values are presented.

Table 20: Comparison between storage pile emissions at Quarry M calculated with interpolated and 30-year climate average data

Scenario	TPM Emissions (T)	Weather Station	TPM Emissions (T)
Low	16.55	Sherbrooke	16.13
Medium	25.90	Trois Rivières	30.50
High	35.06	Montreal	39.90

Readers are reminded the low, medium, and high scenarios use the percentage of time with wind speed greater than 19.3km/h for Sherbrooke, Trois Rivières, and Montreal, respectively, therefore the differences in calculated emissions are entirely attributable to the number of days with precipitation greater than 0.254mm parameter. Because the interpolated number of days is higher than Montreal or Trois Rivières' averages, and marginally lower than the Sherbrooke average, the results in Table 20 are as expected.

Although traditionally reporters are asked to make conservative estimates of emissions, calculating low, medium, and high emissions scenarios would give reporters an easy avenue for adding a measure of uncertainty to their emissions inventory. If the only sources of fugitive dust at Quarry M were storage piles, material handling and unpaved roads – and ignoring any other sources of uncertainty - the site total of TPM could be reported as a range between the sums of low and high scenarios 207.47 to 225.97 which

is a tight range, but acknowledges that imperfections in the calculation have been considered.

8 Conclusions and Future Directions

Over the course of this research, several concerns about the methodology and guidance provided by the National Pollutant Release Inventory (NPRI) for calculating fugitive dust emissions from pits and quarries have been analyzed and addressed. The primary goals were 1) To investigate the potential for error when calculations using 30-year climate averages and develop a methodology to incorporate temporal fluctuations in meteorology; 2) To measure the uncertainty introduced by the use of typical material properties; and 3) To use spatial interpolation to improve the spatial relevance of climate data used by quarries in areas without an Environment Canada weather station.

Doing so involved making simple changes to the AP-42 predictive emissions factor method with the aim of improving calculations without inconveniencing stakeholders by drastically changing the established procedures. The analysis was conducted using 2007 operational data from three quarries in southern Quebec following a "What If" approach; namely what if the quarries had been located in a town where both 30-year climate averages and daily/hourly data were available.

Fugitive dust sources considered were unpaved roads, material handling and wind erosion from storage piles. Other relevant sources at quarries such as overburden removal, grading, drilling, and blasting were excluded because the AP-42 predictive emissions factor equations do not involve either site-specific climatological or material parameters. Current literature suggests that climate and material parameters are among the many influences on fugitive dust deflation, transportation and deposition, therefore the accuracy

of these oversimplified equations merit investigation. This limitation is also relevant to the three fugitive dust sources considered in this study, unpaved roads being a crucial example, as road moisture content is not included in the equation despite being acknowledged by WRAP (2006) as an important factor influencing emissions.

As discussed earlier, the AP-42 predictive emissions factor equations were developed based on a small number of studies of fugitive dust at coal mines in the United States, yet have been regularly applied to other materials and transplanted into a Canadian context. Therefore, further research is needed to validate the equations in the field in a Canadian setting, in a variety of climate types, and with several types of materials.

When temporal resolution was increased by calculating emissions using a combination of daily and hourly climate data instead of the 30-year averages advocated by the NPRI, the differences in emissions calculated using the established procedure were not consistently higher or lower, but were source- and weather station-dependant. Compared to the 30-year climate average method, the use of daily/hourly climate data resulted in emissions from unpaved roads between 38.95% and 42.50% higher; emissions from materials handling that were between 15.31% lower and 18.64% higher, and emissions from wind erosion of storage piles were all lower by 12.48% to 37.50%.

The low masses of emissions from materials handling and storage piles at these quarries suggest that there is little justification for altering the existing quantification procedures for these two operations because the error is unlikely to be noticed on the scale of site-

wide emissions. However this low mass may be misleading as wind erosion also occurs at quarry faces and on other flat surfaces so two major erodible surfaces are missing. Despite this consideration, the high percentage differences between the storage pile emissions calculated with the two methodologies suggest that the daily/hourly methodology would benefit quarries with more materials handled or more storage piles where higher masses of emissions are expected.

Unpaved roads are another issue as differences between the two methodologies were in the order of hundreds of tonnes in some scenarios, meaning that reporters should be using daily data for the calculations in order to provide the most accurate emissions information to stakeholders. Fortunately, the NPRI's Industrial Unpaved Road Dust Calculator facilitates easy implementation of this practice and it should be made standard procedure, particularly where daily data is available. However, the pits and quarries guidance should still remain flexible to allow reporters to use more accurate field quantification methods or to use any methods that improve on AP-42 (Swart *et al.*, 2007).

Sites that handle more materials – particularly dry, dusty materials – might benefit by using hourly wind speeds for the calculations to account for fluctuations in wind, which can achieved using the procedure established in this research. Storage pile calculations highlighted the importance of using data from the specific reporting year to calculate emissions instead of using 30-year averages. This is one of simplest and most necessary recommended changes to the NPRI reporting procedures; if daily/hourly data is not used, an average for the specific reporting year should be.

As was discovered during this research, not all weather stations report all of the 30-year averages needed for calculations, fewer report daily and hourly data, and none reported averages for the specific reporting year. The most import variable to quantify correctly is the percentage of time with wind speed greater the 19.3km/h, yet the existing NPRI guidance instructs reporters to follow a methodology that will assign this parameter a value of zero. Therefore, Environment Canada, in order to facilitate the most accurate reporting possible, must improve the availability of spatially- and temporally-relevant of climate data and revise its guidance documents accordingly.

Considering that the government is unlikely to install weather stations in every location across the country, nor provide all existing stations with the means of publishing the full array of daily and hourly data, one way to improve the quality of climate data without mandating that quarries install monitoring equipment would be to establish a web-based tool for interpolating site-specific data. Using site-specific climate data would begin to account for local topography at quarries, but Environment Canada should consider revising its procedures to improve accuracy by better accounting for topographical features. Although interpolation does not guarantee accuracy, it does help establish a degree of consistency and comparability between reports that cannot be attained when climate data is selected subjectively. Having a centralized methodology for collecting climate parameters would prevent reporters from using values that optimistically lower their reported emissions values and would effectively eliminate one source of contention during audits. Although site-specific interpolated values were shown to have the

potential to be more accurate than the nearest station method, the available climate dataset was rather small and a field-study would be needed to validate the results.

The overarching message of the uncertainty assessment is that assuming the typical silt and moisture contents from AP-42 as true, without any confirmatory sampling, is essentially a guess and thus adds considerable uncertainty to the reported emissions. The widths of the confidence intervals were large, in worst cases ranging between 87.50% below the mean to 650% above it. Because the uncertainty is skewed towards the high values, the chance of underreporting emissions was greater than 30% for each operation at the three quarries. The inverse probabilities indicate that there is a 50% to 70% chance of being unnecessarily conservative and over-reporting emissions when using typical values.

From the perspective of community right-to-know, it is concerning that the existing methodology of selecting "typical" material properties without any field verification leads to such high uncertainty in reported emissions, but from the perspective of quarries, using the typical values result in overly conservative calculations that may tarnish their environmental image. This is a rare case where both sides share a common interest in improving the accuracy of the reports which can be easily achieved by mandating the sampling material properties. All materials exhibit natural variability, and will have different silt and moisture contents depending on location, time of year, and weather conditions. Therefore samples should be collected at several points during the year to establish the possible ranges of properties and ultimately to build a probability density

function. If not already a routine procedure, analyzing dust-generating materials for impurities would be beneficial as the concentrations of heavy metals bound to the harmful $PM_{2.5}$ fraction can be unexpectedly high. Kolominskas and Sullivan (2004) suggest that all companies should have laboratory data related to major emissions sources, but that this unlikely to be undertaken without coercion through regulation or public pressure.

Including measures of uncertainty in NPRI reports would allow reporters to show, with statistical confidence, the expected value of emissions during the reporting year and to disclaim the calculations by providing realistic high and low values. Ideally, with the measurement of site-specific material properties, systematic uncertainty would be reduced to a level where no over- or under-estimate of emissions occurs (Tulles, 2008). It would also be worthwhile for the NPRI to allow the inclusion of uncertainty estimates in reports, building on recommended best practices from IPCC (2000). Attaching confidence limits to emissions estimates identifies opportunities to improve the inventory, and highlights the most likely emissions values while also acknowledging the plausible - but less expected - low and high emissions values (Poulter, 1998; IPCC, 2000).

The concepts explored and conclusions drawn over the course of this study are centred on the fugitive dust emissions from Canadian pits and quarries, but this one substance from this sector is only the tip of the iceberg. For example, all surface mines in Canada likely use the same emissions factors to calculate PM emissions for unpaved roads, material

handling operations, and storage piles, meaning that the issues stretch wider than just limestone pits and quarries. Consider too that many other sectors which report to the NPRI have unpaved roads in their facilities, and therefore would be using the same AP-42 emissions factor methods to calculate the fugitive dust from this important source.

Other sectors also emit hundreds of other substances from a variety of industrial processes, and many reporters use equally suspect emission factors to quantify these releases. Ultimately, the use of site-specific climatic and material data, coupled with a thorough analysis of uncertainty would result in more useful, more transparent emissions report for pits and quarries – and hundreds of other industries – that fits closer with Environment Canada's goal of giving the public the right-to-know.

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Appendix A – Quarry Operational Data

Table A.1: Fleet parameters for Quarry B

Vehicle	Mean Weight (T)	Annual VKT (km)
Heavy Hauler	75	56 250
Heavy Hauler	52.5	56 250
Light Hauler	18.5	12 000
Light Hauler	18.5	80 000
Loader	36.5	5 000
Loader	24	5 000
Transport Truck	13.5	3 422.5
Supervisor Vehicle	1	15 000

Table A.2: Fleet parameters for Quarry J

Vehicle	Mean Weight (T)	Annual VKT (km)
Heavy Hauler	55	124 995
Loader	45	58 500
Loader	20	58 500
Loader	20	58 500
Service Truck	2	4 628
Supervisor Vehicle	1	2314
Water Tanker	10	58 500

Table A.3: Fleet parameters for Quarry M

Vehicle	Mean Weight (T)	Annual VKT (km)
Heavy Hauler	65.4	126 782
Loader	56.2	750
Loader	47.4	750
Loader	47.8	300
Loader	25.7	15 000
Maintenance Truck	2.25	76 166
Supervisor Vehicle	2.25	45 360
Drill	18.75	112.5
18-Wheeler	19.4	1800

Appendix B – Material Properties

Table B.1 Silt Content of unpaved roads

Industry	Road Use	Sample Size	Silt Content (%)			Weight
			Minimum	Mean	Maximum	
Copper Smelting	Plant Road	3	16	16[B]	19	0.011
Iron and Steel Production	Plant Road	135	0.2	6	19	0.496
Sand and Gravel Processing	Plant Road	3	4.1	4.3[B]	6	0.011
	Material Storage Area	1	N/A	7.1	N/A	0.004
Stone Quarrying and Processing	Plant Road	10	2.4	10	16	0.037
	Haul Road to/from Pit	20	5	8.3	15	0.074
Taconite Mining and Processing	Service Road	8	2.4	4.3	7.1	0.029
	Haul Road to/from Pit	12	3.9	5.8	9.7	0.044
Western Surface Coal Mining	Haul Road to/from	21	2.8	8.4	18	0.077
	Plant Road	2	4.9	5.1[A]	5.3	0.007
	Scraper Route	10	7.2	17	2.5	0.037
	Haul Road to/from Pit (freshly graded)	5	18	24	29	0.018
Construction Sites	Scraper Route	20	0.6	8.5	23	0.074
Lumber Saw Mills	Log Yards	2	4.8	8.4	12	0.007
Municipal Solid Waste Landfills	Disposal Routes	20	2.2	6.4	21	0.074
Summary Statistics	**No. of Samples**	**272**	A - Data point not included in calculations as the minimum and maximum are the true values of the 2 samples B - Value is not the mean. With sample size of 3, a true sample value was back-calculated from the published mean.			
	Weighted Mean (μ_w)	**9.3**				
	Weighted Std. Dev. (σ_w)	**7.7**				

(United States Environmental Protection Agency[2], 1995)

Table B.2 Silt content of limestone

Industry	Material	Sample Size	Silt Content (%) Minimum	Mean	Maximum	Weight
Iron and Steel Production	Limestone	3	0.4	0.3[A]	2.3	0.23
Stone Quarrying and Processing	Crushed Limestone	2	1.3	N/A	1.9	0.15
	Various Limestone Products	8	0.8	3.9	14	0.62
Summary Statistics	**No. of Samples**	**13**	A- Value is not the mean. With sample size of 3, a true sample value was back-calculated from the published mean. N/A – Data not available			
	Weighted Mean (μ_w)	**4.46**				
	Weighted Std. Dev. (σ_w)	**5.37**				

(United States Environmental Protection Agency[2], 1995)

Table B.3 Silt content of coal

Industry	Material	Sample Size	Silt Content (%) Minimum	Mean	Maximum	Weight
Iron and Steel Production	Coal	12	2	4.6	7.7	0.14
Western Surface Coal Mining	Coal	15	3.4	6.2	16	0.17
Coal-Fired Power Plant	Coal (as received)	60	0.6	2.2	4.8	0.69
Summary Statistics	**No. of Samples**	**87**				
	Weighted Mean (μ_w)	**3.88**				
	Weighted Std. Dev. (σ_w)	**3.62**				

(United States Environmental Protection Agency[2], 1995)

Table B.4 Silt content of coke

Industry	Material	Sample Size	Silt Content (%) Minimum	Mean	Maximum	Weight
Iron and Steel Production	Coke Breeze	2	4.4	4.9	5.4	1.00
Summary Statistics	**No. of Samples**	**2**				
	Weighted Mean (μ_w)	**4.9**				
	Weighted Std. Dev. (σ_w)	**0.71**				

(United States Environmental Protection Agency[2], 1995)

Table B.5 Moisture content of limestone

Industry	Material	Sample Size	Moisture Content (%) Minimum	Mean	Maximum	Weight
Iron and Steel Production	Limestone	2	N/A	0.2	N/A	0.17
Stone Quarrying and Processing	Crushed Limestone	2	0.3	0.7[A]	1.1	0.17
	Various Limestone Products	8	0.46	2.1	5	0.67
Summary Statistics	**No. of Samples**	**12**				
	Weighted Mean (μ_w)	**2.12**				
	Weighted Std. Dev. (σ_w)	**1.95**				

A - Data point not included in calculations as the minimum and maximum are the true values of the 2 samples analyzed for moisture content.

N/A – Data not available

(United States Environmental Protection Agency[2], 1995)

Table B.6 Moisture content of coal

Industry	Material	Sample Size	Moisture Content (%) Minimum	Mean	Maximum	Weight
Iron and Steel Production	Coal	11	2	4.8	11	0.14
Western Surface Coal Mining	Coal	7	2.8	6.9	20	0.09
Coal-Fired Power Plant	Coal (as received)	59	2.7	4.5	7.4	0.77
Summary Statistics	**No. of Samples**	**77**				
	Weighted Mean (μ_w)	**5.48**				
	Weighted Std. Dev. (σ_w)	**1.36**				

(United States Environmental Protection Agency[2], 1995)

Table B.7 Moisture content of coke

Industry	Material	Sample Size	Silt Content (%) Minimum	Mean	Maximum	Weight
Iron and Steel Production	Coke Breeze	2	6.4	7.8[A]	9.2	1.00
Summary Statistics	**No. of Samples**	**2**				
	Weighted Mean (μ_w)	**7.8**				
	Weighted Std. Dev. (σ_w)	**1.98**				

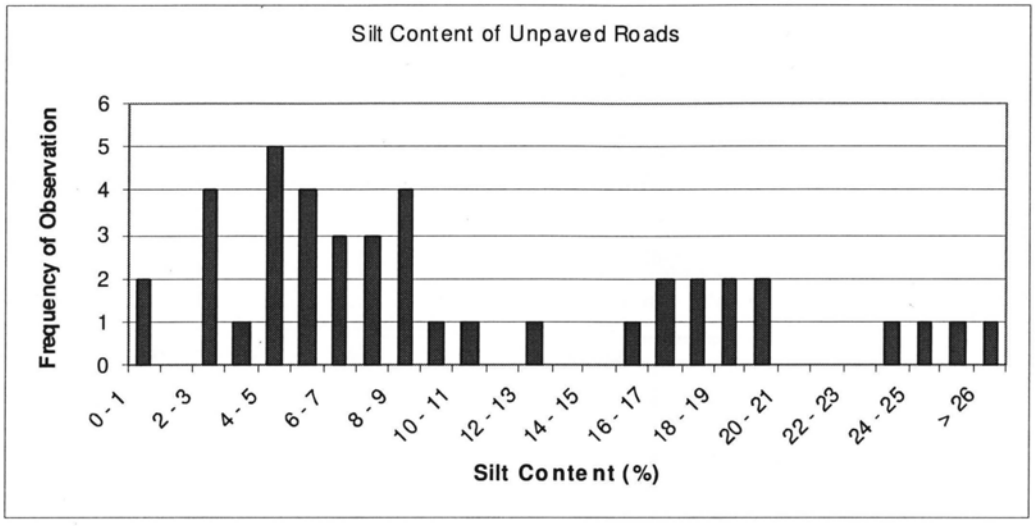
A - Data point not included in calculations as the minimum and maximum are the true values of the 2 samples analyzed for moisture content.

(United States Environmental Protection Agency[2], 1995)

Appendix C - Probability Density Functions

Silt Content of Unpaved Roads

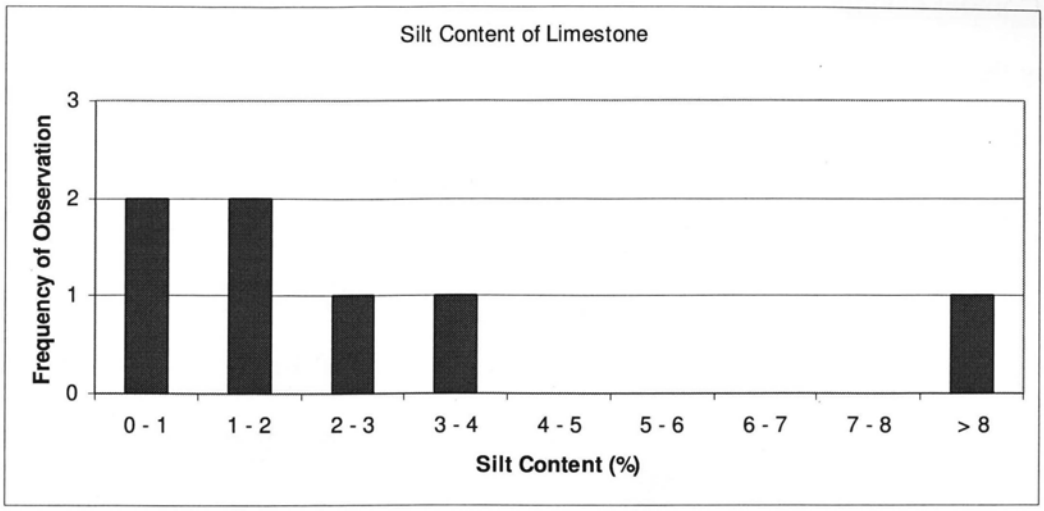
Using the minimum, mean, and maximum values from Table A.4 as unique data points, the following histogram was produced for the silt content of unpaved roads:



The histogram shows that the data is skewed to the right and that the distribution has an approximately lognormal shape. Therefore, a lognormal distribution using the weighted mean ($\mu_w = 9.3$) and weighted standard deviation ($\sigma_w = 7.7$) from Table A.4 was used to represent the silt content of unpaved roads in all simulations. The distribution was truncated by an upper bound of 100.00%, as this is the theoretical maximum; although the random selection of silt contents greater than 50% is exceptionally rare given the shape of the distribution.

Silt Content of Limestone

Using the minimum, mean, and maximum values from Table A.5 as unique data points, the following histogram, Figure 6.2, was produced for the silt content of limestone:



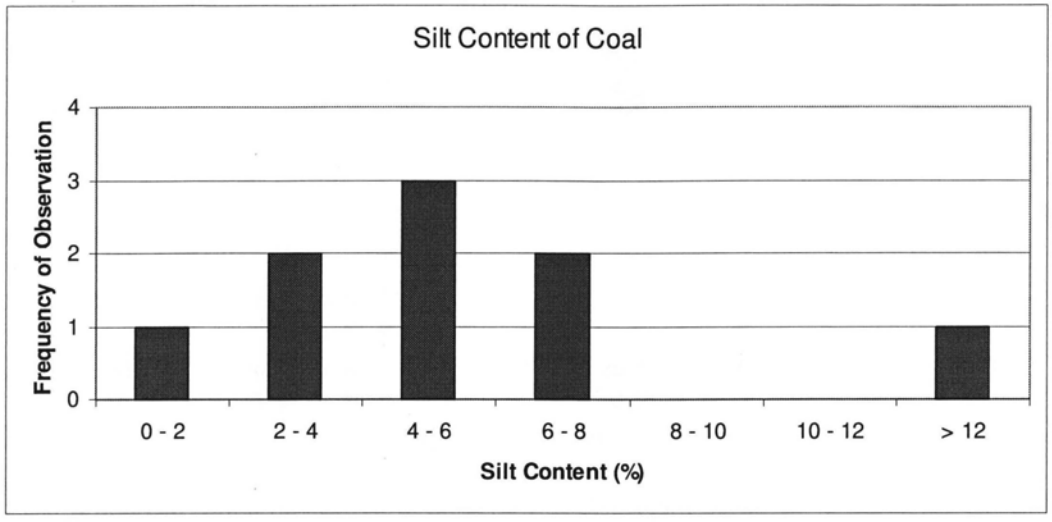
Due to the small number of data points, no specific distribution shape is clear. However, the frequency of observations decreases as silt content increases, suggesting that the PDF should emphasize lower values. For this reason, a choice was made to use a lognormal distribution using the weighted mean ($\mu_w = 4.46$) and weighted standard deviation ($\sigma_w = 5.37$) from Table A.5. The distribution was truncated at 100% to prevent the selection of impossible values. A triangular distribution was also considered, however it would have disproportionately increased the frequency of values between 4% and 8% that were not observed.

Moisture Content of Limestone

Using the minimum, mean, and maximum values from Table A.8 as unique data points, a histogram was produced for moisture content of limestone. In this case, however, there was no strong emphasis on any part of the distribution, therefore a truncated uniform distribution was used to give equal probabilities to all values between the minimum (0.2%) and maximum (5.0%) observations in Table A.8.

Silt Content of Coal

Using the minimum, mean, and maximum values from Table A.6 as unique data points, the following histogram was produced for the silt content of coal:



Similar to the silt content of unpaved roads in Figure 6.1, the data appears skewed to the right, giving the distribution an approximately lognormal shape. Therefore, a lognormal distribution was parameterized with the weighted mean (μ_w = 3.88) and weighted standard deviation (σ_w = 3.62) from Table A.6. The PDF was truncated at 100%, the theoretical maximum silt content. An argument could be made that the data is best represented by a normal distribution, however this would add undue emphasis on silt contents higher than 8% which were only observed once. An additional complication of using a normal distribution for this data is that truncations needed to prevent the random selection of negative values would have distorted the mean and standard deviation of the

distribution to unrepresentative values. Applying these truncations to a normal distribution would have, in the end, given the PDF an approximately lognormal shape.

Moisture Content of Coal

Using the minimum, mean, and maximum values from Table A.9 as unique data points, a histogram was produced for the moisture content of coal. Like the silt content of coal, the distribution was skewed to the right. A lognormal distribution was selected for this variable as well, using the weighted mean ($\mu_w = 5.48$) and weighted standard deviation ($\sigma_w = 1.36$) from Table A.9. A triangular distribution was also tested, however using it would have emphasized the values between 12% and 20% which is less representative of the available data.

Silt Content of Coke

Because there were only two values for the silt content of coke, a uniform distribution was used to give equal weight to all values between the observations of 4.4% and 5.4%.

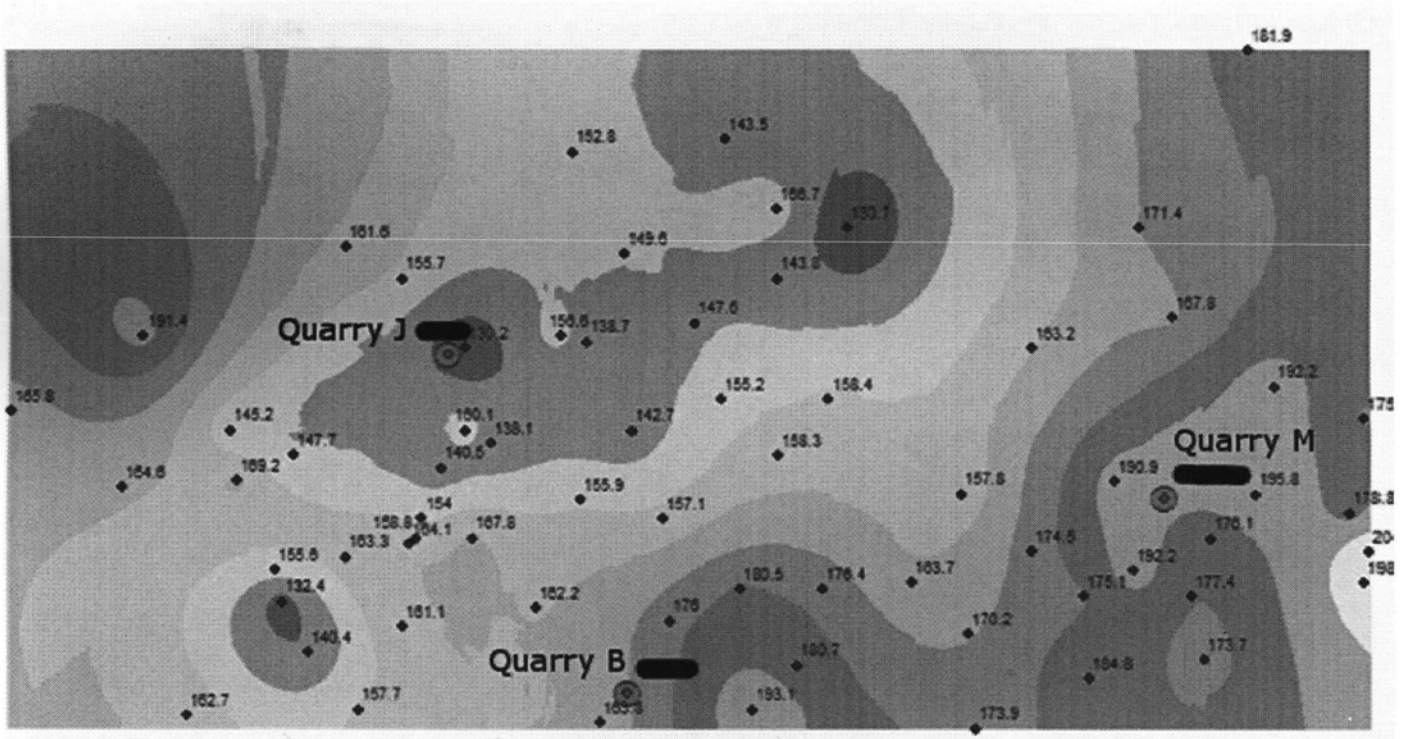
Moisture Content of Coke

Similarly, only two values for the moisture content of coke were available, so a uniform distribution was used to give equal weight to all values between the observations of 6.4% and 9.2%.

Appendix D – Site-Specific Interpolated Climate Data

Table A.11: Site-Specific Interpolated Climate Averages

Site	Variable	Interpolated Average
Quarry J	Wind speed	12.23 m/s
	Number of days with precipitation greater than 0.2mm	139 days
	Percentage of time with wind speed greater than 19.3km/h	31.37%
	Number of months with snow depth greater than 15cm	3 months
Quarry M	Wind speed	11.60 m/s
	Number of days with precipitation greater than 0.2mm	187 days
	Percentage of time with wind speed greater than 19.3km/h	Multiple
	Number of months with snow depth greater than 15cm	3 months



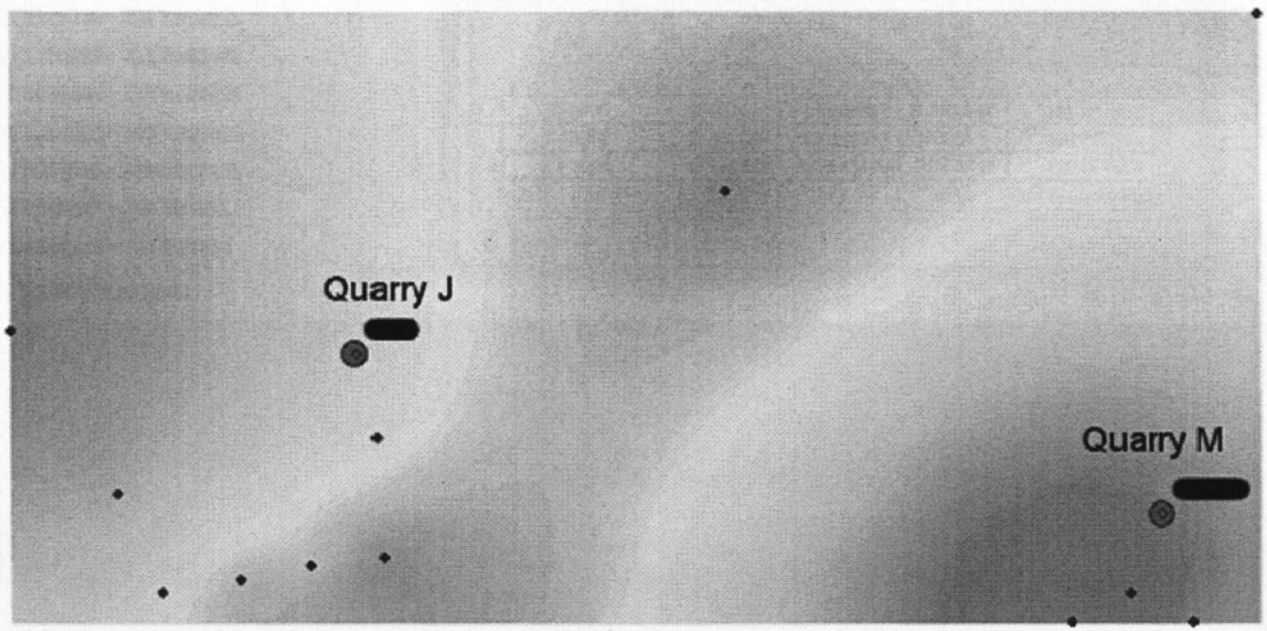
nnual Precipitation

- 134.4386749 - 141.694931
- 141.6949311 - 148.9511871
- 148.9511872 - 156.2074432
- 156.2074433 - 163.4636993
- 163.4636994 - 170.7199554
- 170.7199555 - 177.9762115
- 177.9762116 - 185.2324677
- 185.2324678 - 192.4887238
- 192.4887239 - 199.7449799

Attributes of Extract_Quarrie1

FID	Shape *	Y	X	SITE	RASTERVALU
0	Point	45.110914	-73.012158	Quarry B	170.014908
1	Point	45.998889	-73.479217	Quarry J	138.527237
2	Point	45.621814	-71.604961	Quarry M	187.491699

Record: 1 Show: All Selected ecords



Average Annual Windspeed

Value

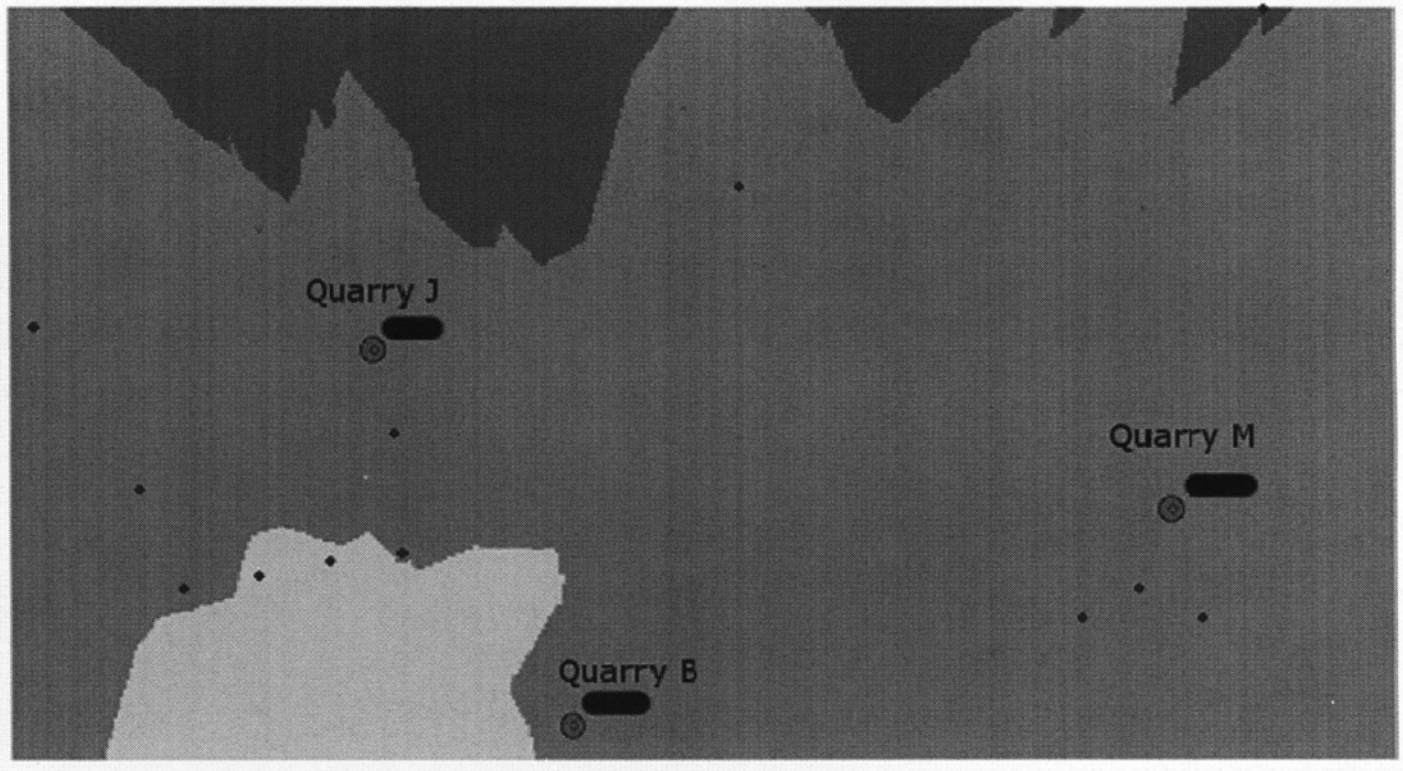
High : 131576

Low : 11.0295

Attributes of Extract_Quarrie5

FID	Shape *	Y	X	SITE	RASTERVALU
0	Point	45.110914	-73.012158	Quarry B	-9999
1	Point	45.998889	-73.479217	Quarry J	12.295608
2	Point	45.621814	-71.604961	Quarry M	11.46476

Record: 1 Show: All Selected Records



Number of Months With Snowfall over 15 cm

Value

High : 4

Low : 2

Attributes of Extract_SnowMonths

FID	Shape *	Y	X	SITE	RASTERVALU
0	Point	45.110914	-73.012158	Quarry B	3
1	Point	45.998889	-73.479217	Quarry J	3
2	Point	45.621814	-71.604961	Quarry M	3

Record: 3 Show: All Selected Records