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Assessment of Inhalable Particulate Matter Associated with a Refinery in Curaçao

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Abstract

Inhalation and atmospheric pollution studies have focused on particulate matter due to correlations and associations with various morbidities and mortalities. This research analyzed ambient concentrations of inhalable particulate matter (PM_{10}) on the island of Curaçao in order to evaluate through comparative literature analysis and recommended public health guidelines the potential health risks. Available hourly, daily and monthly PM_{10} measurements were accessed from June 2010 through December 2014 from a local air monitoring station in Willemstad. Mean annual concentrations of PM_{10} (31 - 122 μg/m^3) in Curaçao are among the highest reported globally, demonstrating an increasing trend over time and exceed current public health guidelines recommended by local and international agencies. While the epidemiological evidence is inadequate to infer a causal association between health effects and long-term exposures of the measured PM_{10} concentrations, the results indicate that emissions controls are not adequate for compliance with international exposure standards.

Keywords

Inhalable Particulate Matter, PM_{10}, Curaçao, Refineries

1. Introduction

Air pollution may be considered an environmental health risk having been associated with a number of acute (e.g., respiratory and cardiovascular events, hospital and emergency room admissions) and chronic (e.g., chronic bronchitis, lung cancer, mortality) effects [1]. In many cases it is difficult to determine direct causality with one particular constituent of air pollution (PM, sulfur dioxide, carbon monoxide, VOCs, etc.) due to its complexity and multiple sources, the inability to assign area exposure values to individuals, as well as the unavoidable confounding inherent to large scale environmental epidemiology studies. While more than 98% of air pollution in urban settings are gases or vapor-phase compounds, more recent inhalation and atmospheric pollution studies have focused on particulate matter due to some correlations with mortality and adverse respiratory health effects [2] [3]. It has been suggested that up to 8% of premature mortalities globally are related to both indoor and outdoor concentrations of particulate matter, though these findings are difficult to interpret given the uncertainties present in the respective study designs [4] [5] [6] [7]. A number of studies have suggested associations between long-term exposure to PM and various health effects including accelerated cardiovascular and respiratory mortality, compromised lung function and relative increase of lung cancer risk [2] [8] [9] [10] [11]. Inhalable particulate matter (PM_{10}), specifically, has been associated with increases in daily mortality and hospital admissions for respiratory distress (pneumonia, asthma and decreased lung function in children) [9] [12]. Though the multi-factorial nature of these outcomes and the inability to disentangle confounding factors make it impossible to know if PM is the causative agent behind morbidities and mortalities the findings warrant investigating potential risks from exposure to this type of air pollution.

Many factors contribute to the chemical composition of PM, including combustion sources, climate, season, and type of urban and/or industrial pollution [3]. Particulate matter can be emitted from both natural and anthropogenic sources with both primary (directly emitted) and secondary (atmospherically derived) components [13]. Primary sources include wildfires, sea spray, organic matter and the combustion of both fossil fuels and biofuels. Secondary sources of PM include wood smoke, gaseous vegetative emissions and vehicular emissions and other condensates from atmospheric chemistry. The major components of PM consist of inert compounds, semivolatile organic compounds, metals, carbonaceous material mainly from combustion and vehicle exhaust, biological material, and minerals. The density, concentration, and composition of particulate matter can vary widely among geographic locations.

Refinery operations have been associated with atmospheric emissions of a wide variety of criteria air pollutants, including particulate matter. The type and quality of the crude oil, refinery process and refined products all influence the variability, composition and amount of emissions from one refinery to another. One of the largest and oldest refineries in the Wider Caribbean Region is Isla Refineriá, which opened in 1918 and is located within the densely populated capital of Willemstad, Curaçao on the shores of Schottegat Bay. Although, the refinery was considered obsolete in the mid-1980s, it is still in use today processing ~335,000 barrels per day yet it has not been able or required to comply with environmental standards and permit requirements [14].

The objectives of this study were to analyze hourly, daily, and monthly PM_{10}

measurements over a four year period (2010-2014) to determine if any temporal trends exist and to compare levels with public health guidelines in order to assess potential public health risks in Curaçao.

2. Materials and Methods

2.1. Site Selection

Curaçao is an island in the southern Caribbean, ~40 miles off the Venezuelan coast. It is currently considered a constituent country of the Kingdom of the Netherlands since its dissolution in 2010 from the Netherland Antilles. Curaçao is located in the Southern Caribbean Dry Zone approximately 12˚ north of the equator, which is characterized by a semi-arid to arid climate, with a distinguishable dry (March-September) and rainy season (October-February), and sustained easterlies. Curacao is typically warm and sunny with an average temperature of 27˚C and an average annual rainfall of approximately 570 mm. The island is approximately 59 kilometers in length, 4 - 11 kilometers wide and a total land mass area of ~443 km^2. The population of ~152,000 consists of greater than 50 nationalities with Dutch and Papiamento as the official languages. The majority of the population (>130,000) resides in Willemstad which is home to the Isla Refineriá.

2.2. Local Monitoring Station Descriptions

Since mid-2010, two air monitoring stations, Beth Chaim and Kas Chikitu, located in Willemstad, Curaçao have been collecting validated and continuous measurements of air quality parameters (SO_2, PM_{10}, TSP, H_2S). The Beth Chaim station is located at the western edge, downwind of the Schottegat industrial area of the refinery and only measures SO_2 and TSP. Kas Chikitu is located approximately 2 - 3 km downwind in the Marchena/Wishi residential area and is primarily used to monitor the residential load of SO_2, hydrogen sulfide (H_2S) and PM_{10}. Available hourly and daily measurements of PM_{10} were downloaded from June 1, 2010 through December 31, 2014 for analysis from the Kas Chikitu station. Twenty-four hour PM_{10} daily means ($n = 1603$) measured at the Kas Chikitu station were analyzed. Monitoring stations operate in accordance with the ISO/IEC 17025 accreditation (certificate number L 426) of GGD Amsterdam using tapered element oscillating balance (TEOM 50C) methodology to measure PM_{10}.

2.3. Data Analysis

All PM_{10} data is expressed in μg/m^3. All data analysis was performed using Statistica Version 6.1 (Stat Soft, Inc., Tulsa, OK). If data did not meet the assumptions of normality, nonparametric hypothesis tests were performed. If a potential explanatory variable was categorical (e.g. year), the nonparametric Kruskal-Wallis ANOVA was run using $\alpha = 0.05$. If the Kruskal-Wallis ANOVA was found to be significant multiple comparisons revealed significant differences between categorical factors. Concentrations of PM_{10} were compared daily and annually and correlations were also evaluated against environmental factors (e.g. temperature, humidity and wind speed).

3. Results and Discussion

3.1. Ambient Concentrations of Inhalable Particulate Matter

Daily PM_{10} concentrations were downloaded from the Kas Chikitu station (n = 1603) from June 1, 2010 through December 31, 2014. The 24-hour daily mean concentrations ranged from 0.37 - 341 µg/m^3 (**Figure 1**). Mean annual PM_{10} concentrations at the Kas Chikitu station ranged from 31 µg/m^3 in 2010 to 122 µg/m^3 in 2014 (**Figure 2**). There were statistically significant temporal trends

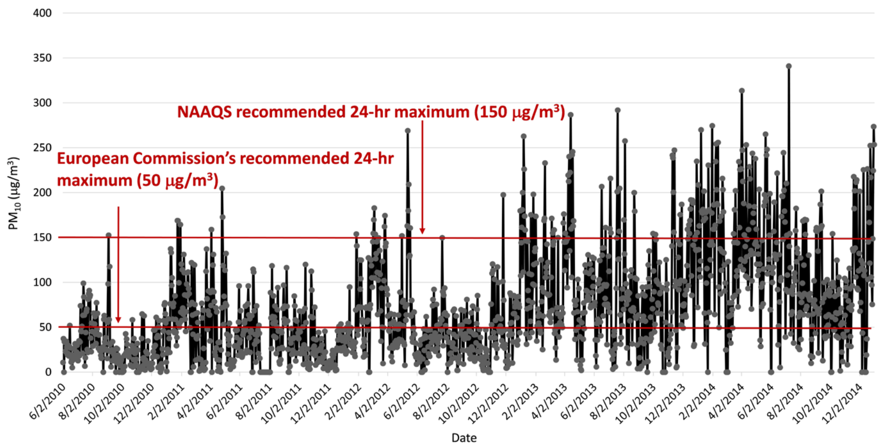

Figure 1. Daily 24-hour mean PM_{10} concentrations (µg/m^3) collected at the Kas Chikitu air monitoring station in Curaçao from June 1, 2010 through December 31, 2014. Current 24 hour guidelines recommended by NAAQS (150 µg/m^3) and the European Commission (50 µg/m^3) are depicted with dashed lines, respectively.

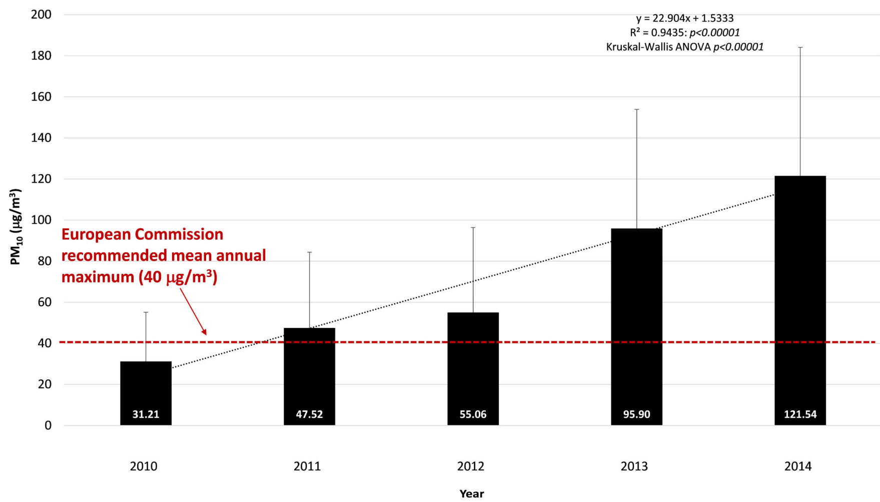

Figure 2. Annual mean concentrations of PM_{10} (µg/m^3) measured at the Kas Chikitu air monitoring station in Curaçao for the years 2010 through 2014. The current European Commission's annual (40 µg/m^3) recommendation is depicted with a dashed line.

observed with a strong increasing trend (R^2 = 0.94) over time. In general, the 2014 mean annual PM_{10} concentrations were significantly ($p > 0.00001$) higher than the previous four years.

3.2. Global Comparisons of Ambient PM_{10} Concentrations

The annual average PM_{10} concentrations have increased 74% since 2010. Excluding 2010, since measurements are only for a seven month period, the annual average increased 61% since 2011. In contrast, PM_{10} concentrations in the US have shown a 34% decrease in the 24-hour average concentrations since 1999 and a 31% decrease in annual average ambient concentrations since 1990 (http://www.epa.gov/airtrends). The 2014 annual mean concentrations for PM_{10} in Curaçao (121.5 μg/m^3) is among some of the highest concentrations reported globally, measuring approximately 13 times higher than those reported in Iceland (9 μg/m^3), yet were two times lower than levels recorded in Pakistan (282 μg/m^3).

3.3. PM_{10} Compliance with Public Health Guidelines

The maximum annual mean concentrations for PM_{10} that are currently recommended by Curaçao and the European Commission are 75 and 40 μg/m^3, respectively. The annual mean concentrations for the years 2011 through 2014 exceeded the current PM_{10} guidelines recommended by the European Commission (40 μg/m^3). Additionally, mean annual PM_{10} concentrations for 2013 (95.9 μg/m^3) and 2014 (121.5 μg/m^3) both exceeded the island's guidelines for PM_{10} (75 μg/m^3). Mean 24-hour maximum concentrations of PM_{10} have also been recommended by NAAQS (150 μg/m^3), the European Commission (50 μg/m^3) and Curaçao (150 μg/m^3). The number of days that have exceeded the 24-hour daily maximum concentrations of PM_{10} has demonstrated strong increasing trends in this study (**Figure 3**). The majority of 2010 (82%), 2011 (64%), and 2012 (60%) were compliant with recommended guidelines for measured PM_{10} concentrations (**Figure 4**). Conversely, a total of 77% of 2013 and 85% of 2014 exceeded all recommended 24-hour guidelines for PM_{10} concentrations. Curaçao allows 5% of the calendar days to exceed 150 μg/m^3; however, 10%, 22%, and 35% of 2012, 2013, 2014, respectively, exceeded this value [15].

3.4. Potential Risks of PM_{10} Inhalation

There are a number of epidemiologic studies reporting the associations of PM_{10} with various health outcomes, including mortality, morbidity and increased emergency room visits and hospitalization, though these results are difficult to interpret due to the inability of these studies to adequately control for confounding and the potential for exposure misclassification (**Figures 5-7**) [11] [16]-[43]. This study primarily focused on literature published after 2010 since there are several meta-analyses and reviews covering literature published prior to 2010.

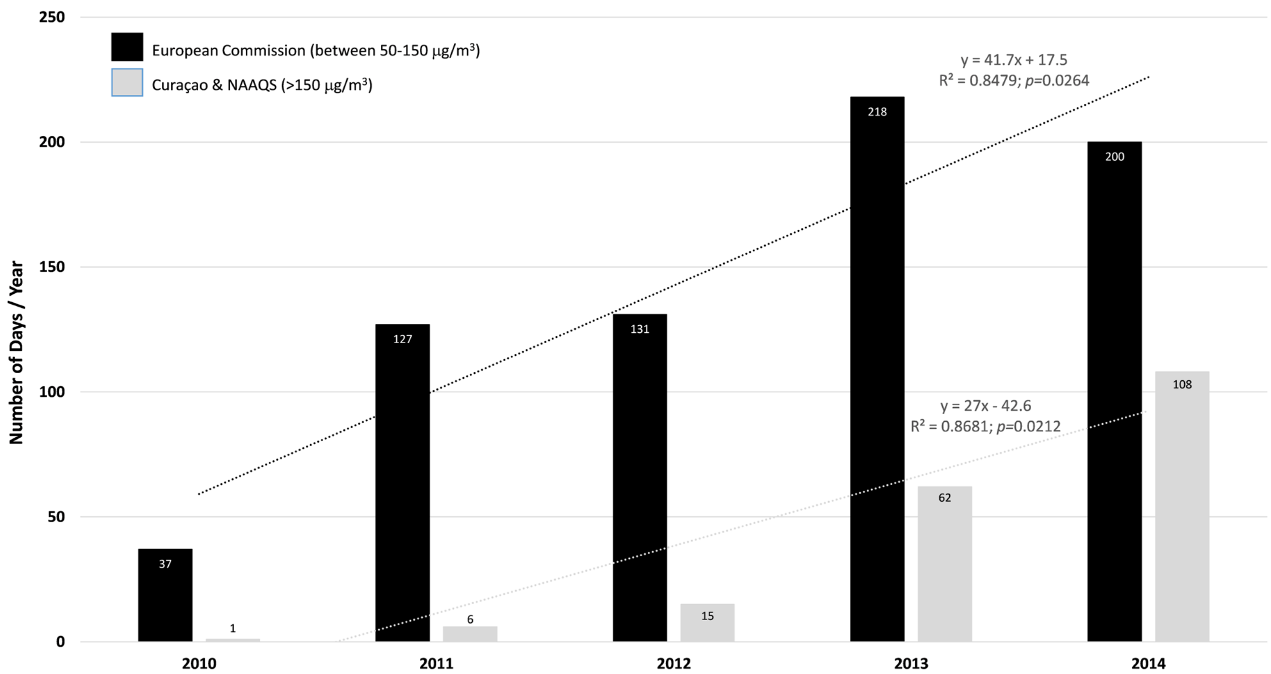

Figure 3. Number of days per year PM_{10} concentrations exceeded recommended guidelines. Strong increasing trends were observed for the number of days exceeding the European Commission's recommended guidelines (R^2 = 0.85) as well as recommendations by Curaçao and the NAAQS (R^2 = 0.87). The current 24-hour maximum concentrations recommended by the European Commission, NAAQS and Curaçao are 50 µg/m³, 150 µg/m³ and 150 µg/m³, respectively.

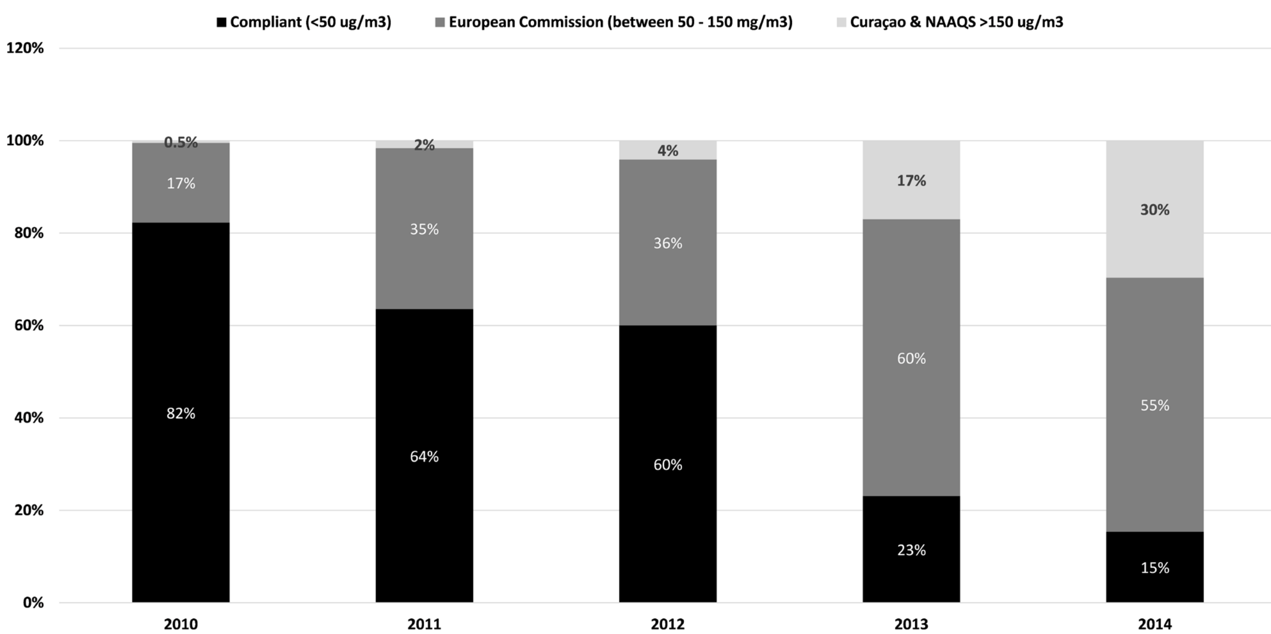

Figure 4. Percentages of each year that were either in compliance or exceeded current maximum 24-hour guidelines for PM_{10} concentrations. Strong decreasing trend was observed in annual compliance.

The relative risk reported for various mortality demonstrate positive associations with PM_{10}, however the data is somewhat inconsistent (**Figure 5**). For instance, two studies reported positive associations for respiratory mortality,

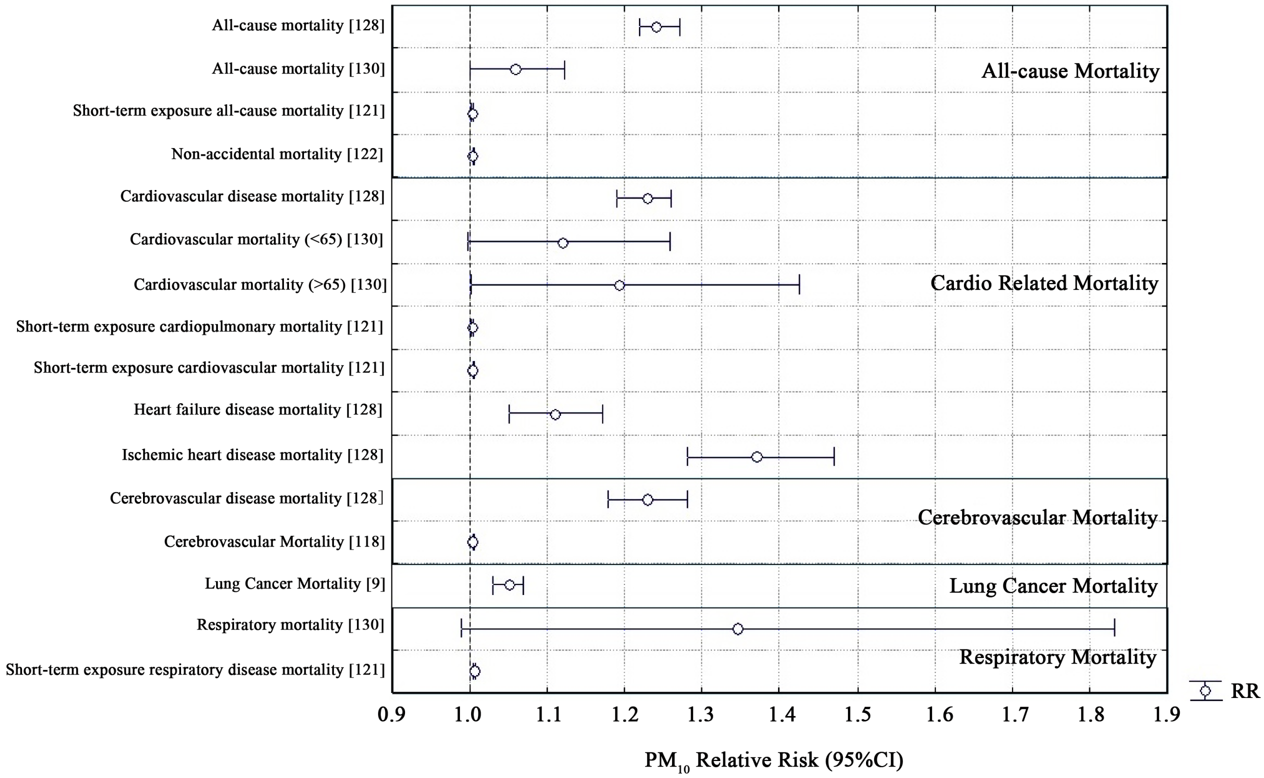

Figure 5. Relative risk estimates (95% CI) for PM_{10} associated mortality from published literature.

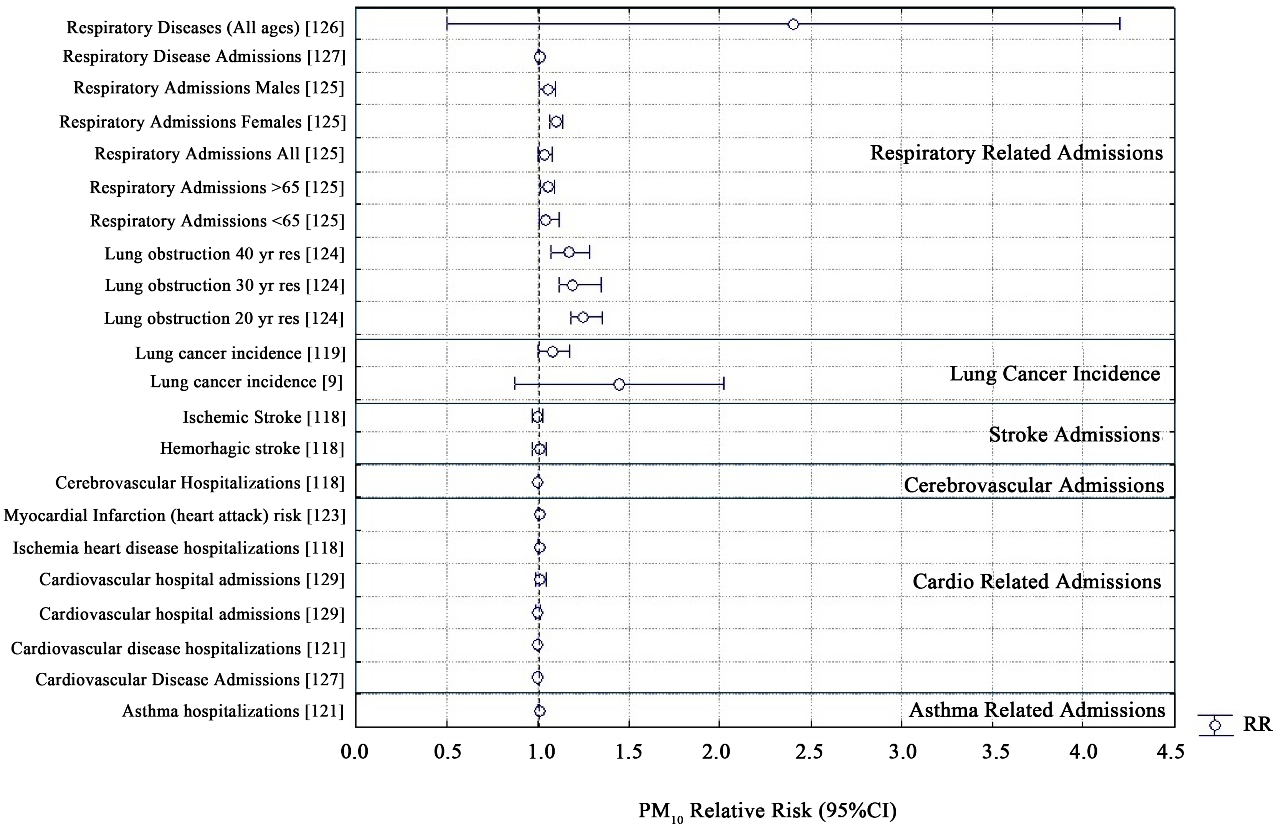

Figure 6. Relative risk estimates (95% CI) for PM_{10} associated morbidity and hospital admissions from published literature.

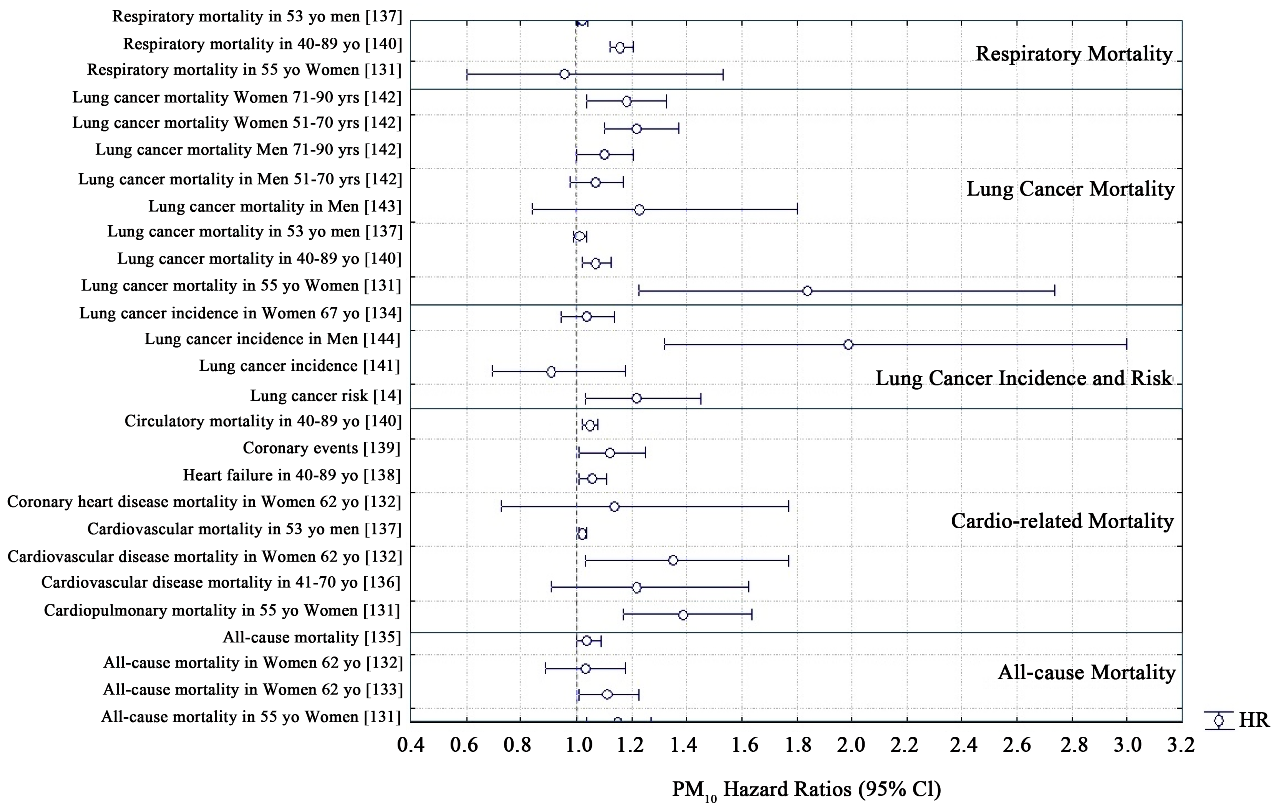

Figure 7. Hazard ratios (95% CI) for PM_{10} associated mortality and morbidity from published literature.

where only one was statistically significant [19] [28]. Liang *et al.* [28] reported positive associations, with respiratory mortality (RR: 1.347, 95% CI: 0.990 - 1.833) during the winter months in Taiwan with a mean PM_{10} concentration of 66.7 μg/m^3. However this association was not significant since the RR included unity and had a wide confidence interval range suggesting greater uncertainty. In contrast, a meta-analysis consisting of 26 studies in China with annual PM_{10} concentrations ranging from 44 - 156 μg/m^3, reported positive associations between short-term PM_{10} exposure and respiratory mortality (RR: 1.0057, 95% CI: 1.004 - 1.0075), though the de minimis magnitude of effect makes this finding difficult to interpret [19].

Similarly, relative risks for cardio related mortality also demonstrated both significant and non-significant results with PM_{10} [16] [19] [26] [28]. For each 10 μg/m^3 of PM_{10}, some significant positive associations were reported for cardiovascular disease mortality (RR: 1.23, 95% CI: 1.19 - 1.26), ischemic heart disease mortality (RR: 1.37, 95% CI: 1.28 - 1.47) and heart failure mortality (RR: 1.11, 95% CI: 1.05 - 1.17) within a retrospective cohort, containing over 39,000 subjects from northern China [26], but these findings are unique. Liang *et al.* [28] used a time-series regression model to analyze mortality among central Taiwan residents and reported non-significant positive associations during the winter months (mean PM_{10} of 66.7 μg/m^3) between PM_{10} and cardiovascular mortality for residents less than 65 years of age (RR: 1.12, 95% CI: 0.998 - 1.258) and borderline, yet significant for residents greater than 65 years old (RR: 1.194; 95% CI: 1.0025 - 1.425). A meta-analysis in China also reported associations with short-term exposures to PM_{10} (annual means ranging from 44 - 156 μg/m^3) and cardiopulmonary (RR: 1.0034; 95% CI: 1.0023 - 1.0046) and cardiovascular mortality (RR: 1.0049; 95% CI: 1.0034 - 1.0063) [19]. Positive associations were also reported for all-cause and non-accidental mortality [11] [16] [19] [20] [25] [28]. All were significant with the exception of the study by Liang *et al.* [28] which found a positive yet non-significant association between PM_{10} and all-cause mortality in Taiwan (RR: 1.059; 95% CI: 0.999 - 1.122). In addition, positive associations between lung cancer mortality and long-term PM_{10} exposures (RR: 1.05; 95% CI: 1.03 - 1.07) were found in a meta-analysis of 19 studies conducted globally [11]. However, none of these measures of association achieved a magnitude of effect that clearly demonstrates a direct impact of PM on mortality in exclusion of other factors.

The relative risks reported for various morbidity and hospitalizations is much less convincing of positive associations with PM_{10} since many of the studies report near or include unity (**Figure 6**) [11] [16] [17] [19] [21] [22] [23] [24] [25] [27]. Significant positive associations were found in a number of studies between respiratory diseases, respiratory related hospital admissions and lung obstruction and PM_{10} (annual PM_{10} ranged 31 - 270 μg/m^3). However, a study in the highly polluted industrial city of Lanzhou, China (PM_{10} daily mean: 197 μg/m^3) reported positive, non-significant associations between PM_{10} and respiratory diseases (RR: 2.4, 95% CI: 0.5 - 4.2) and significant positive associations with pneumonia (RR: 5.3, 95% CI: 1.3 - 9.5) and upper respiratory tract infections in people less than 65 years of age (RR: 13.7, 95% CI: 2.5 - 26.2) [24]. However, the confidence intervals are relatively wide suggesting increased uncertainty. Relative risks reported for incidences of lung cancer among two meta-analyses were also positive yet were not statistically significant since both included unity [11]. The meta-analysis consisting of 60 studies from 1966-2014 by Wang *et al.* [16] reported evidence of inconsistent, nonsignificant associations between short-term changes in PM_{10} and hemorrhagic stroke (RR: 1.009; 95% CI: 0.976 - 1.043), ischemic stroke (RR: 1.0; 95% CI: 0.976 - 1.024) and cerebrovascular disease (RR: 1.003; 95% CI: 0.999 - 1.008). A study in Scotland reported positive, nonsignificant associations between PM_{10} (20 - 22 μg/m^3 mean annual PM_{10}) and cardiovascular hospital admissions [27]. In contrast, a study in China (44 - 156 μg/m^3 mean annual PM_{10}) and Iran (111.3 μg/m^3 mean annual PM_{10}) both reported significant positive associations with cardiovascular related hospital admissions [19] [25].

Hazard ratios were also reported in a number of studies for various mortality and risks associated with PM_{10} [29]-[43] (**Figure 7**). In 2008, Puett *et al.* [31] reported significant positive associations between PM_{10} and all-cause mortality (HR: 1.11; 95% CI: 1.01 - 1.23) and cardiovascular disease mortality (HR: 1.35; 95% CI: 1.03 - 1.77) in the Nurses' Health Study consisting of 66,250 women with a mean age of 62 years. In 2009, Puett *et al.* [30] reported nonsignificant results between $PM_{10-2.5}$ and all-cause mortality (HR: 1.03; 95% CI: 0.89-1.18) and cardiovascular disease mortality (HR: 1.14; 95% CI: 0.73 - 1.77) also for the Nurses' Health Study. It is important to note in the latter study $PM_{2.5}$ was subtracted from PM_{10} concentrations suggesting the associations found with PM_{10} in the earlier study were potentially influenced by $PM_{2.5}$. Nonsignificant results (HR: 1.22, 95% CI: 0.91 - 1.63) were found between long-term PM_{10} (13.5 - 48.1 μg/m^3 annual PM_{10}) exposure and cardiovascular disease mortality in twenty cohorts across 13 countries in Europe (ESCAPE Project) [34]. In contrast, several studies found significant positive associations with cardio-related events. A prospective cohort consisting of 4800 women (mean age 55 years old) in Germany found significant positive associations between long-term PM_{10} (34.8 - 52.5 μg/m^3 annual mean PM_{10}) exposure and cardiopulmonary mortality (HR: 1.39, 95% CI: 1.17 - 1.64) [29]. In addition, a study of 11 cohorts in the ESCAPE project reported a positive association (HR: 1.12, 95% CI: 1.01 - 1.25) with long-term PM_{10} (14 - 48 μg/m^3 annual mean) exposure and coronary events [37].

A large cohort study in England consisting of over 800,000 patients, aged 40 - 89 years, reported associations (HR: 1.16, 95% CI: 1.12 - 1.21) between PM_{10} (19.7 μg/m^3 annual mean) and respiratory mortality [38]. Associations (HR: 1.023, 95% CI: 1.005 - 1.042) were also reported for a cohort of 71,000 middle aged Chinese men exposed to much higher concentrations of PM_{10} (104 μg/m^3 annual mean) than those measured in the English cohort study [35]. In contrast, the prospective cohort study in Germany reported nonsignificant associations (HR: 0.96, 95% CI: 0.6 - 1.53) with respiratory mortality in middle aged women [29].

A number of cohort studies in the USA, Germany, England, Norway and China have also reported inconsistent associations (HRs) between PM_{10} and lung cancer mortality [29] [35] [38] [41] [42]. Associations were found between PM_{10} and lung cancer mortality in Norwegian women between the ages of 51 and 70 (HR: 1.22; 95% CI: 1.1 - 1.37) and between 71 and 90 (HR: 1.18; 95% CI: 1.04 - 1.33) [41]. A German cohort study also reported associations (HR: 1.84; 95% CI: 1.23 - 2.74) in women with a mean age of 55, however the confidence interval range is large [29]. In contrast, the USA, Norwegian and Chinese cohorts reported nonsignificant positive associations of PM_{10} and lung cancer mortality in men [35] [41] [42].

In summary, the epidemiological studies presented demonstrate some associations between health effects and PM_{10} although results are mixed, often presented with large amounts of uncertainty, and effect sizes on the level of minutiae. Cardiovascular and respiratory effects and mortality were observed in locations with annual mean concentrations ranging from 7.7 - 270 μg/m^3. Potential inconsistencies between studies and results could be due to different PM_{10} constituents between geographical regions as well as various study designs and methodology, though there is an inherent inability of the cited studies to adequately control confounding and reliably assign area exposure estimates to individuals. The USEPA concluded that the evidence provided in the literature and the biological plausibility was suggestive of a causal relationship between short-term exposures to $PM_{10-2.5}$ and cardiovascular effects, respiratory effects, mortality, yet there was inadequate evidence to suggest causative relationships with long-term exposures [18].

3.5. Environmental Factors and PM_{10} Concentrations

Daily temperature, humidity, precipitation and wind speeds were analyzed for trends and correlations with PM_{10} concentrations (Figure 8). The mean temperature (±standard deviation) from 2010 through 2014 was 27.9˚C (±1.2) and although Kruskal-Wallis multiple comparisons revealed annual differences ($p < 0.000001$) there were no observable trends ($R^2 = 0.0007$). The mean humidity (77.7% ± 4.1%; $R^2 = 0.73$) and precipitation (1.70 ± 6.9 mm; $R^2 = 0.75$) from 2010 through 2014 both demonstrated moderately strong decreasing trends. Mean annual wind speeds (19.6 ± 4.7 km/h) demonstrated a strong increasing trend over time ($R^2 = 0.91$). This is consistent with increasing global trends in wind speed [44]. There was a weak to moderately strong correlation between PM_{10} concentrations and wind speed (Figure 9). There were no correlations found between PM_{10} and temperature (r = 0.04) and only a weak, negative correlation with humidity (r = −0.22).

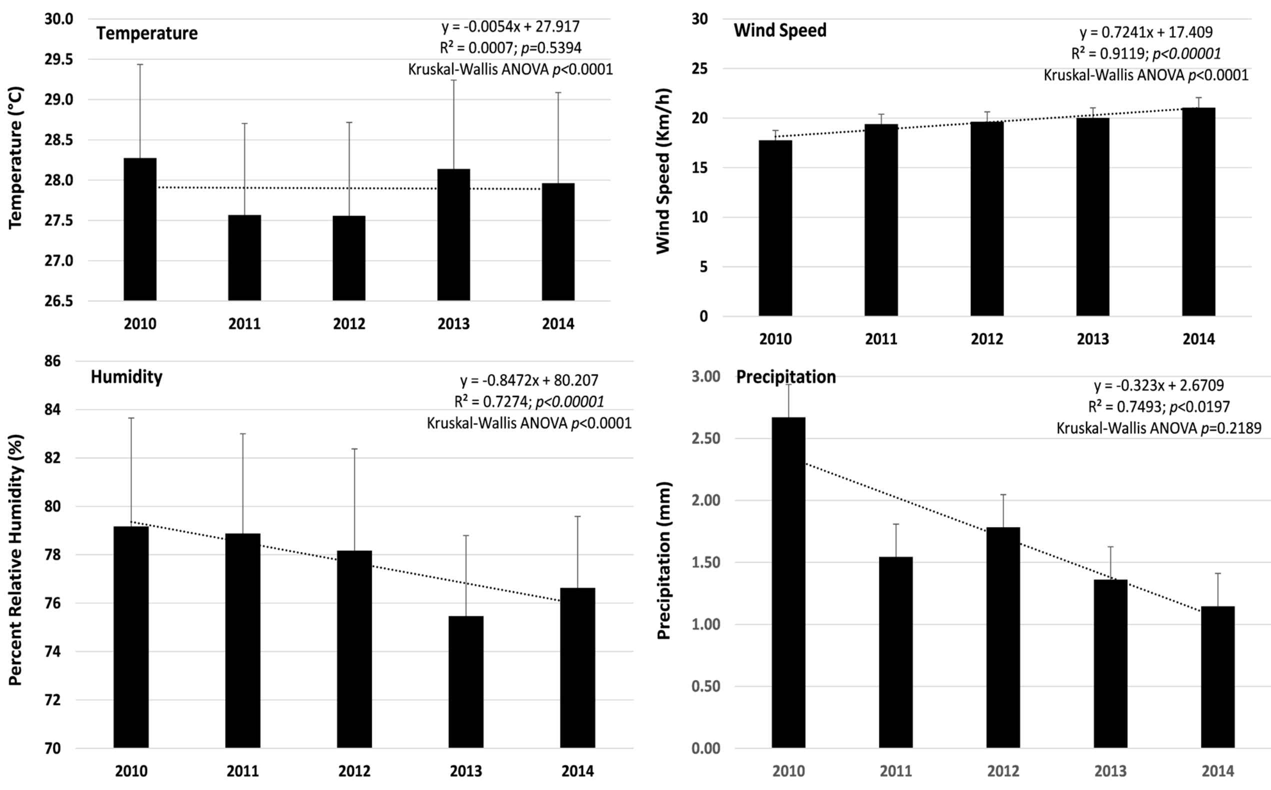

Figure 8. Annual environmental parameters measured in Curaçao, 2010-2014. Trends are denoted with dash lines.

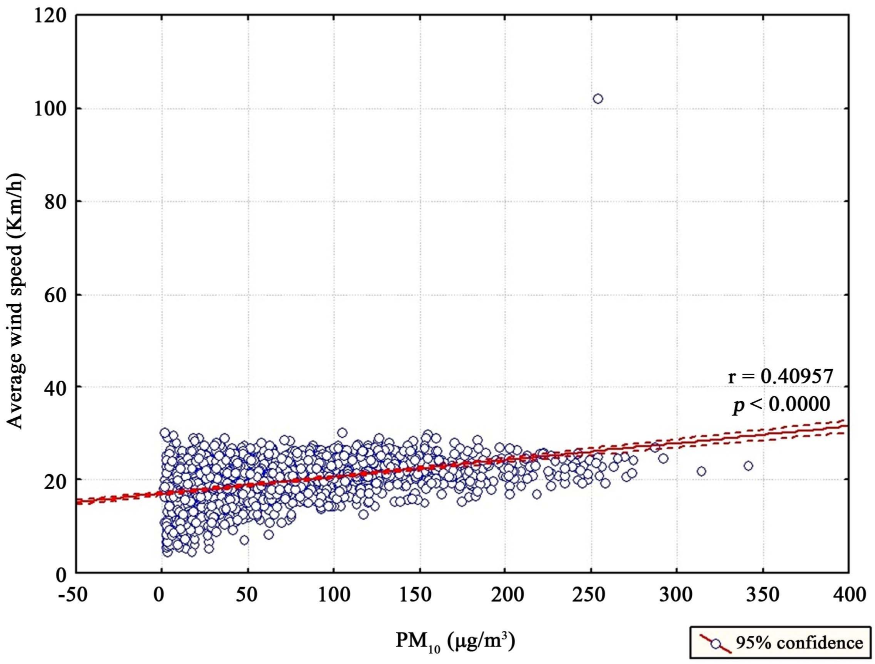

Figure 9. Regression analysis between daily PM_{10} (µg/m^3) and wind speed measurements in Curaçao, 2010-2014. Regression bands represent 95% confidence limits.

4. Conclusions

The objectives of this investigation were to analyze levels of PM_{10} in ambient air surrounding Willemstad, Curaçao in order to determine if any temporal trends exist in the measured concentrations, verify if measured levels exceed current public health guidelines and to identify potential health risks. In conclusion, concentrations of PM_{10} in Curaçao are among the highest reported globally and demonstrate an increasing trend over time. Levels of PM_{10} exceeded the annual and 24-hour guidelines recommended by Curaçao, the European Commission, World Health Organization and the USEPA. Furthermore, both the 24-hour and annual mean concentrations of PM_{10} measured in Curaçao were within the ranges speculated by the USEPA to impact cardiovascular and respiratory outcomes and mortality as a result of short-term exposures. In addition, increasing wind speeds were correlated with increasing PM_{10} concentrations over time potentially impacting residents within the plume trajectory. However, as the epidemiological evidence is inadequate to infer causality between health effects and chronic, long-term exposures to PM_{10}, the strongest conclusion that can be drawn from this analysis is that local emissions controls are inadequate to meet international PM_{10} public health standards.

Future research needs in Curaçao include expanding the air monitoring efforts to include areas upwind of the refinery as well as additional petrochemical emissions, including but not limited to sulfur dioxide, particulate matter (PM_{10} and $PM_{2.5}$), benzene, as well as both the vapor and particulate phases of ambient PAHs. Additional environmental studies are encouraged to evaluate the extent of contamination in a variety of biota and matrices (*i.e.* water, sediment, and fish).

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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