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Human health risk characterization of petroleum coke calcining facility emissions

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ABSTRACT

Calcining processes including handling and storage of raw petroleum coke may result in Particulate Matter (PM) and gaseous emissions. Concerns have been raised over the potential association between particulate and aerosol pollution and adverse respiratory health effects including decrements in lung function. This risk characterization evaluated the exposure concentrations of ambient air pollutants including PM_{10} and gaseous pollutants from a petroleum coke calciner facility. The ambient air pollutant levels were collected through monitors installed at multiple locations in the vicinity of the facility. The measured and modeled particulate levels in ambient air from the calciner facility were compared to standards protective of public health. The results indicated that exposure levels were, on occasions at sites farther from the facility, higher than the public health limit of 150 $\mu g/m^3$ 24-h average for PM_{10}. However, the carbon fraction demonstrated that the contribution from the calciner facility was de minimis. Exposure levels of the modeled SO_2, CO, NO_x and PM_{10} concentrations were also below public health air quality standards. These results demonstrate that emissions from calcining processes involving petroleum coke, at facilities that are well controlled, are below regulatory standards and are not expected to produce a public health risk.

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1. Introduction

Calcined coke is a high quality carbon material produced by calcining green petroleum coke. Calcining is the process of heating green petroleum coke in a kiln to remove excess moisture, extract all remaining hydrocarbons and modify the crystalline structure of the coke, resulting in a denser more electrically conductive product. The temperatures in the kiln during the calcining process can range from 1200 °C to 1450 °C. The final product, calcined coke, is primarily used to make carbon anodes for the aluminum industry and recarburizing agent for other industries such as the steel industry.

Green coke is basically an inert substance and the health hazards assigned to green coke are mostly associated with Particulate Matter (PM) exposures, which can be produced in the occupational setting during the process of calcination (McKee et al., 2014). If not appropriately controlled, this process could lead to the excess production of particulate emissions from either handling or storing of raw coke, or from the stack emissions during the production of calcined coke. Though industries that store, transfer or process petroleum products are broadly described as “petroleum industries”, the coke calcining industry is unique in that it can and is a standalone process distinct from other petroleum industries in producing a different profile of air emissions compared to what could be termed “petrochemical” industries, such as oil refineries. Unlike calcined coke production, petroleum oil refineries produce a variety of products and each product has a varied level of hazard potential (Clark et al., 2013). Calcining facilities process green coke to produce the final product, calcined coke, which substantially limits the constituents and quantity of emissions. The high temperatures required for the process of calcination and the further processing of the emissions gases in a combustion chamber, sometimes referred to as a pyro scrubber, provides sufficient heat and retention that destroys most of the emissions from this process for volatile organic compounds and combustible particles (CONCAWE, 1993).

Though calcined coke has shown low hazard potential in human populations due to low volatile content, there remains some public health concern regarding the emissions from these facilities. The purpose of this research is to evaluate the emissions of a calcining

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facility engaged in the handling and storage of green coke, as well as in the calcining process itself, and characterize the presence of any public health risks associated with these emissions. Particular emphasis is placed on the particulate matter generated, the resultant ambient air quality, and the risks associated with these levels of particulate matter.

2. Methods and materials

The calcining facility evaluated in this study has the potential to produce approximately 444,000 short tons of calcined coke product per year. The calcining facility is located on the west side of a shipping canal within a port in eastern Argentina. The long and narrow property containing the facility abuts the west side of the shipping canal and has a south-southwest to north-northeast orientation. From a turning basin at its south-southwestern terminus, the shipping canal extends north-northeast to the city of Rio de la Plata.

Meteorological data were taken into account in evaluating emissions. The general direction of the air flowing over the weather station located in calcining facility is towards urban areas, and previously passes over the River Plate (influenced by its interaction with the water) and then, over a narrow surface on the shore, between the river and the weather station. The air towards the monitors flows previously over urban areas. Proximate to the area in which the calciner resides is an oil refinery that supplies the green coke feed, industrial ports, and a large, populated community. There are multiple sources of pollution in the area including the refinery, sand and gravel storage and transport, and other industrial activity. Substantial sources of non-industrial pollution exists in the local area including unpaved roads, non-municipal garbage burning, fuel combustion from automobiles and heating, as well as a large number of wood burning stoves in homes for cooking.

The emissions constituents evaluated included PM_{10} and the gaseous emissions of CO, SO_2, and NO_x. Volatile Organic Compounds (VOC's) are a major byproduct of the petroleum refinery industry and concentrations of approximately 15% VOC's are found in green coke as a result of the coking process. However, these VOC's and other volatiles present in the green coke are efficiently combusted and destroyed because of the high temperatures during the calcining process and their presence is not detectable in calcining emissions; as a result, VOC's were not included in this analysis (Cetin et al., 2003; Lin et al., 2004; Hassan, 2005).

Both measured and modeled emissions from the kiln stacks, as well as actual ambient air monitoring were conducted for various pollutants. Ambient air quality PM_{10} monitoring data was collected continuously by three tapered element oscillating microbalance (TEOM 1400) monitors, and one multi-angle absorption photometer (MAAP) monitor at various locations surrounding the facility. Air data collection for year 2009 was conducted by the Center for Environmental Investigations (CIMA by its Spanish acronym). For the period of 2010 through the second quarter 2013, air data was collected by the facility under the direct supervision of The Provincial Agency for Sustainable Development (which acts as the regulatory agency and is known as OPDS by its Spanish acronym). For the third quarter 2013 through second quarter 2014, air data was collected by the Geochronology and Isotopic Institute (INGEIS by its Spanish acronym). Both CIMA and INGEIS are scientific research centers and are considered independent third party experts in this activity.

As part of the data collection activities, CIMA utilized three TEOM 1400 monitors, two HI-Vol PM_{10} monitors, and a MAAP monitor to measure the ambient air quality at various locations surrounding the calcining facility. The TEOM 1400s and MAAP monitors continued to be used for PM_{10} monitoring by the calcining facility at various locations from 2010 through the first half of 2013. INGEIS continued to use the same monitors at various locations around the facility for the second half of 2013 to the present. The locations and distance of these monitors have varied over time with the nearest location just at the entrance of the facility and the furthest at 1190 m from the calcining facility. TEOM monitors measure continuous ambient air quality values, while Hi-Vol samplers are based on 24-h samples that are typically taken on 3 or 6 day intervals. The estimated contribution from other sources of particulate emissions in the area was determined by wind trajectory analysis to estimate the emissions that contributed to PM_{10} concentrations.

A wind trajectory analysis was performed using 5-min meteorological data and coincident TEOM data to estimate the potential for the calcining plant to contribute to the reported Hi-Vol concentration. This analysis was performed using wind direction data from the facility meteorological tower that were coincident in time with the 24-h measured data collected at the monitors. The comparison was performed for 5-min average data since the TEOM PM_{10} data are available over 5-min intervals. If the wind was blowing in a direction such that a parcel of air would traverse the facility site and be transported to the specific monitor site, then the potential for percent contribution of particulate from the facility was identified.

The MAAP monitor measures the black carbon concentration of the PM_{10} size particles. The entirety of particulate emissions from the calciner facility is black carbon since green petroleum coke and calcined coke are black carbon. Black carbon is an indicator of the amount of carbon related material such as calcined coke and raw petcoke that is present in the particulate (PM_{10}) sample collected at the TEOM and can distinguish between emissions potentially from the calcining facility and non-calcining emissions. However, some black carbon measured at monitoring sites could also be attributed to diesel combustion or debris burning. The black carbon concentrations being measured by the MAAP monitor would, for the most part, represent emissions from the calciner facility since it is the largest source of black carbon in the area.

The facility also conducted periodic stack testing to determine PM, NO_x, CO and SO_2 emission rates from the two kilns from 2011 through the third quarter of 2014. Particle size distribution was conducted on the measured total particulate matter to define the portion of PM_{10} in the stack gases. The kiln stack samples were collected using the EPA Conditional Test Method (CTM) 022/030. Per the CTM, a continuous stream of stack gas is pulled into the analyzer for a set period of time to determine the concentrations. The rates of emissions were calculated from the resulting concentrations and the volumetric flow.

The rates of emissions of the various pollutants were then input into an air dispersion model along with 5 years of meteorological data. The USEPA AERMOD model was used for these assessments. AERMOD is a steady state Gaussian dispersion model developed by the USEPA and American Meteorological Society (AMS) and represents the current state of the art modeling (EPA, 2004). The modeling was performed in general accordance of the Argentinean modeling requirements of Stage III of Resolution Nr. 242/97 and using techniques that would be acceptable to EPA for the air quality assessment of a project located within the United States. The EPA's AERMOD model (Version 07026) was used with hourly meteorological data to predict PM emission impacts at ground-level receptor locations for 24-h averaging periods. AERMOD is a Gaussian plume model that calculates impacts at each receptor for each hour in the meteorological data set (typically a full year of data) and it provides maximum ground-level concentrations for point sources such as the two Kiln stacks. It also predicts concentrations in the cavity zone and uses Building Profile Input Program (BPIP) data to simulate the influence of proximate structures to estimate the

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