IMPACT OF GASEOUS AND PARTICULATE MATTER EMISSION FOR FLUID CATALYTIC CRACKING UNIT

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ABSTRACT

Fluid catalytic cracking unit is a major part of petroleum refineries as it treats heavy fractions from various process units to produce light ends (valuable products). FCC unit feedstock consists of heavy hydrocarbon with high sulphur contents and the catalyst used is zeolite impregnated with rare earth metals i.e. Lanthanum and Cerium. Catalytic cracking reaction takes place at an elevated temperature in fluidized bed reactors generating sulphur-contaminated coke on the catalyst with large quantity of attrited catalyst fines. In the regenerator, coke is completely burnt producing SO_2, PM emissions are mainly due to high attrition of cold makeup catalyst charge and operating conditions, vapour velocity particle velocity, particle collision and particle degradation. This study is dedicated to the quantitative analysis of the impact of harmful emissions resulting from FCC units on the environment.

INTRODUCTION

Fluid catalytic cracking (FCC) of heavy ends into high value liquid fuels is commonly carried out in the oil refining industry. In this process the heavy feedstock containing sulphur as a major contaminant is cracked to light products. Sulphur is redistributed in the liquid and gaseous products and coke on the catalyst. In the regenerator coke with sulphur contamination is completely burnt and flue gas containing SO_2 is discharged with catalyst fines produced, mainly due to high attrition of cold makeup catalyst charge and operating conditions i.e. vapour velocity, particle velocity, particle collision and particle degradation (Abdul Wahab et al., 2002).

In the present work, a comprehensive emission inventories from FCC unit in an oil refinery have been prepared. These inventories are calculated based on complete combustion of sulphur and coke impregnated on the catalyst in the regenerator. Mainly for SO_2 and Particulate matter (PM) emission rates are calculated accurately using material balances for a yearlong period considering seasonal variations in the operation of the process unit, Yateem et al., (2010). PM emission inventory is used in dispersion model to assess its impact on the immediate surroundings of the refinery.

The most advanced dispersion model Aermod (Caputo *et al.*, 2003; Isakov *et al.*, 2007; Kesarkar *et al.*, 2007) has been selected for prediction ground level concentration of PM based on comprehensive year long emission inventory of FCC unit.

Aermod is a dispersion model that uses Gaussian distribution for the stable conditions and non-Gaussian probabilities density function for the unstable conditions. Aermet (Aermod pre-processor) provides planetary boundary layer parameters over a high altitude to yield accurate predicted concentration values for a given meteorological conditions. It can accommodate large meteorological data (multiple years). Aermap (Aermod pre-processor) generates regular receptors over a given terrain for the evaluation of pollutants ground level concentrations.

The meteorological data for year 2008 are obtained and are used in pre-processor Aermet to generate planetary boundary layers parameters. These generated data are used in Aermod for actual emission rates to predict ground level concentrations of PM and study the influence of prevailing meteorological conditions at this particular site.

MODEL APPLICATION

1. *Input Data*

Aermod dispersion model implementation requires the following items of data:

1. Source information: including pollutant emission rate (g/s), location coordinates in Universal Transverse Mercator (UTM) (m), base elevation from the sea level (m), stack height (m), exit stack inner diameter (m), exit stack gas velocity (m/s), and exit stack gas temperature (^oK).

2. Meteorological information for the region of interest: includes anemometer height (m), wind speed (m/s), wind

direction (flow vector from which the wind is blowing) (in degrees clockwise from the north), ambient air temperature (^oC), stability class at the hour of measurement (dimensionless) and hourly mixing height (m).

3. Receptor information: This can be specified or generated by the program to predict the pollutants' concentrations at the selected receptors.

The entire required source input data are obtained from FCC unit in the refinery. A stack of 80 m height, an inner diameter of 2.3 m, with an average exit gas velocity of 20 m/s and exit gas temperature of 550 ^oK are fed into the model. Monthly emission variation is considered with total SO_2 emission rate of 6089.2 g/s and total PM emission rate of 302 g/s as presented in detail (Yateem *et al.* 2010).

2. *Area of Study*

The area of study in this work covers portion of Ahmadi governorate in the state of Kuwait. Fahaheel area is adjacent to the petroleum refinery has one of the Kuwait EPA air quality monitoring station located at a polyclinic. Both areas Fahaheel and Ahmadi are surrounded by arid desert in the west side and bordered by the Persian Gulf from the east.

Two different types of receptor coordinates are used as input to the Aermod model to predict the ground level concentration of SO_2 and PM, these are:

1. Discrete Cartesian receptors specified at the sensitive areas viz., a school, a shopping area and EPA monitoring stations in Fahaheel. A hospital and petroleum services companies' offices are selected in Ahmadi.
2. Uniform Cartesian Grid receptors covering the entire area of study, where the FCC stack (emissions source) is located almost in the centre of the mesh grid.

The receptors selected are based on the actual sites in a UTM location coordinate of the area of interest map. Table 1 shows the selected discrete receptors information.

The uniform grid receptors of a total 1764 (42 x 42) were divided into (Δx = 300 m x Δy = 250 m) to cover about 12 x 10 km area of study. The optimum selection of the mesh size is based on the computational accuracy and time.

Table 1 the selected discrete receptors information Coordinates are related to the centre of wind rose

ID Number	Discrete receptor identity	X-coordinate	Y-coordinate
1	Fahaheel Polyclinic	219854.25	3219765.79
2	Petroleum Services Offices in Ahmadi	216666.87	3220105.63
3	School in Fahaheel	220300.00	3219820.85
4	Ahmadi Hospital	213458.86	3221523.64
5	Shopping area in Fahaheel	219274.32	3219554.21

RESULTS AND DISCUSSION

A yearlong comprehensive metrological data are processed by Aermet to generate boundary layer parameters and to pass all meteorological observations to Aermod.

Figure 1 shows wind direction and magnitude for a period of year 2008. It is observed that most of the time; the prevailing wind direction is from North West. There is strong influence from the neighbouring Persian Gulf as the refinery is located at the coast, resulting into strong sea breeze blowing from East direction. Wind class frequency distribution for the entire year confirming 2 % calm conditions, while 39.8 % is between 3.6 - 5.7 m/s. the highest wind class 8.8-11.1 m/s is less than 1%.

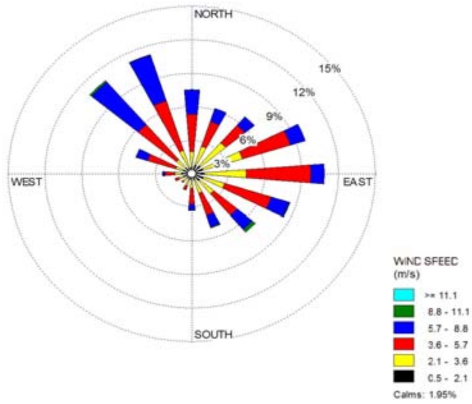

Fig. 1 wind rose for a period of year 2008

A model run is performed for actual monthly emission variation with total SO_2 emission rate of 6089.2 g/s and PM emission rate of 302 g/s. Monthly emission factors for SO_2 is tabulated in Table 2 and Monthly emission factors for PM is tabulated in Table 3. A discrete receptor is selected at Kuwait Environmental Public Authority monitoring station located at polyclinic in Fahaheel area. Concentrations of SO_2, NO_x, H_2S, O_3, CO, CO_2, methane, non-methane hydrocarbon, Benzene, Toluene, Xylenes, ethylbenzene, total suspended particulates and meteorological parameters are continuously recorded on hourly basis.

Table 2 SO_2 monthly emission factors

January	February	March	April	May	June
0.077	0.083	0.096	0.1	0.077	0.088
July	**August**	**September**	**October**	**November**	**December**
0.067	0.067	0.088	0.077	0.1	0.75

Table 3 PM monthly emission factors

January	February	March	April	May	June
0.093	0.097	0.091	0.079	0.079	0.083
July	**August**	**September**	**October**	**November**	**December**
0.064	0.063	0.085	0.079	0.079	0.1

Hourly predicted ground level concentrations at specified discrete receptor showed large scatter due to variation in meteorological conditions and the recorded values influenced by the contribution of various emission sources has made the comparison impracticable. Therefore, daily average measured concentrations of SO_2 were compared with the daily-predicted concentrations to validate the model output.

Figure 2 shows the plot between the measured top 20 daily average values versus the daily predicted top 20 values at the discrete receptor, Kuwait-EPA monitoring station.

The slope is equal to 0.72, reflecting high measured values compared to predicted values, depicting the contribution of other emission sources. The correlation coefficient is equal to 0.91 reflecting an acceptable validation of the model output with measured average daily SO_2 concentrations.

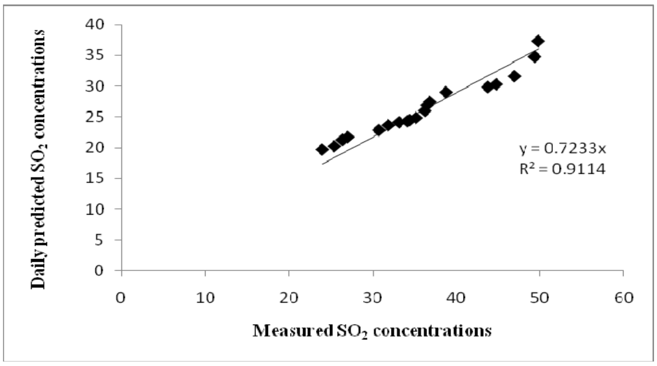

Fig. 2 Daily predicted SO_2 concentrations vs. measured SO_2 concentrations

The predicted hourly average ground level concentrations of SO_2 are compared with Kuwait-EPA Ambient Air Quality Standards (AAQS) at all of the selected receptors. The maximum allowable level for the hourly average concentration of SO_2, specified by Kuwait-EPA, is 444 μg/m^3. Fig. 3 shows the isopleths of the predicted hourly average ground level concentration of SO_2 calculated at the selected uniform grid receptors.

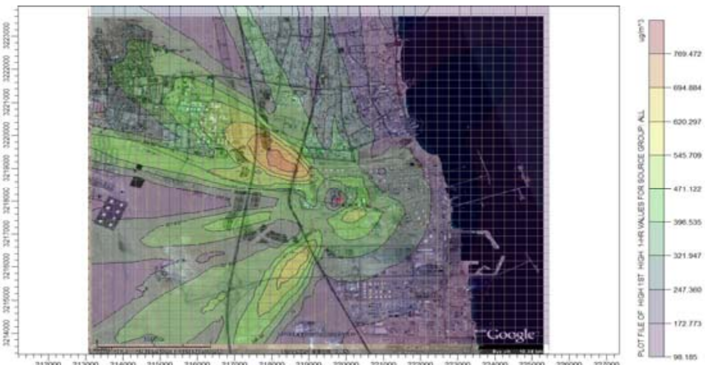

Fig. 3 Isopleths plot of the predicted hourly average ground level concentration of SO_2

The isopleths indicate the predicted spatial variations of the ground level concentrations of SO_2. The maximum predicted hourly average ground level concentration of SO_2 in the vicinity of the refinery exceeded by as much as 300 μg/m^3. The highest predicted concentration is equal to 769 μg/m^3, observed on the 8th of March 2008 at 8:00 hour and about 1.713 km in the NW direction from the FCC stack, and not far from the Fahaheel and Ahmadi areas at the receptor coordinates of X = 218557.94, Y = 3219169. This high value of the predicted SO_2 concentration is expected due to the elevated SO_2 emission rate, which resulted from the high sulphur content in the FCC feedstock and other operational conditions and the prevailing meteorological conditions (temperature, humidity, wind speed, wind direction, stability class and planetary boundary layer).

A thorough inspection on fig. 3 indicates that predicted concentrations of SO_2 exceed the allowable hourly limit at 5.3 % of the study area from North West and South West direction from the stack.

Similarly, the predicted daily average ground level concentration of SO_2 is compared with Kuwait EPA ambient air quality standards at all receptors. The allowable level for the daily average concentration of SO_2 is 157 μg/m^3. Fig. 4 shows the isopleths of the predicted daily average ground level concentration of SO_2 computed at the selected uniform grid receptors.

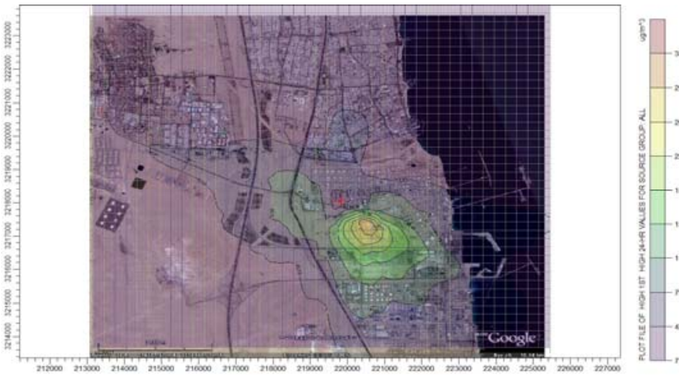

Fig. 4 Isopleths plot of the predicted daily average ground level concentration of SO_2

The isopleths indicate the daily predicted spatial variations of the ground level concentrations of SO_2 in the area of study. The highest daily predicted concentration is equal to 335μg/m^3, observed on the 9th of November 2008 and about 0.75 km in the SE direction from the stack, at a receptor coordinates of X = 220357.94, Y = 3217419 affecting the neighbouring Shuaiba industrial area, Kuwait main industrial complex. This high value of the daily predicted SO_2 concentration is exceeded the allowable level by 157 μg/m^3 and obviously influenced by the prevailing meteorological conditions, especially the predominant North West wind and other meteorological factors.

Discrete receptor 2, is located at Petroleum services offices, has shown the highest SO_2 hourly concentration equal to 544μg/m^3 on 27th February at 8:00 hours. The hourly concentration level rise beyond acceptable peak is occurred four times at this location throughout the study period. The highest daily concentration at the same receptor is equal to 39μg/m^3 on 8th March.

Discrete receptor 3 shows the highest SO_2 hourly concentration equal to 279μg/m^3 on 2nd March at 4:00 hours. This concentration is below the Kuwait EPA hourly standards. The daily highest concentration is equal to 57μg/m^3 on 2nd March. Discrete receptor 4, is located at Ahmadi hospital, has shown the highest SO_2 hourly ground level concentration equal to 288μg/m^3 on 27th February at 8:00 hours. This value is also below the specified hourly limit set by Kuwait EPA. The daily predicted concentration is equal to 23μg/m^3 on 30th April. Discrete receptor 5, is located at shopping area, has shown the highest SO_2 hourly ground level concentration is equal to 336μg/m^3 on 23rd October at 8:00 hours. The daily predicted concentration is equal to 45μg/m^3 on 22nd April. Both hourly and daily predicted values are below Kuwait EPA hourly and daily ambient air quality standards.

1. *Model Sensitivity*

To observe the computational model sensitivity, another run is performed using two finer meshes consisting of 21 x 21 uniform receptor points, the first covering hourly highest ground level concentration area, the second covering daily highest predicted ground level concentration area. The output accuracy has improved for both pollutants due to application of interpolation using small values of $\Delta x = 150$ m, $\Delta y = 110$ m for the first mesh and $\Delta x = 100$ m, $\Delta y = 100$m for the second mesh. There is 0.65% increase in the hourly highest ground level concentration and 2.8% increase in the daily highest ground level concentration, which are insignificant.

2. *Parametric Study*

FCC stack sensitivity analysis is performed on 3 scenarios (stack height, SO_2 emission rate and stack diameter). In scenario 1, analysis for stack heights 50 m, 80 m, 120 m, 160 m and 200 m is conducted while keeping the emission rate, exit flue gas velocity, exit temperature and stack diameter constant. The influence of stack height is shown in fig. 5. It is obvious from the figure that the highest predicted hourly and daily ground level concentrations of SO_2 are reduced substantially as stack height is increased. The reduction in the highest computed hourly ground level concentration of SO_2 is almost 50% when stack height is doubled. The decrease in evaluated hourly SO_2 concentration as a function of stack height is given as an exponential expression $C(\mu g/m^3) = 1600.7e^{-9.071x10^{-3}h}$ and r^2 is 0.999, where h is the stack height (m). The hourly gradient dC/dh $= 14.52e^{-9.071x10^{-3}h}$ becomes insignificant at higher stack elevations. The highest daily predicted ground level concentration as a function of stack height is given as $C(\mu g/m^3) = 1409.8e^{-1.732x10^{-2}h}$ and r^2 is 0.984. The daily highest predicted concentration gradient is dC/dh$= 24.42e^{-1.732x10^{-2}h}$. The locations of hourly highest predicted concentrations of SO_2 from the stack, as a function of stack height is shown in figure 7 and related as $D(km) = 0.597e^{1.16x10^{-2}h}$ and r^2 is 0.9.

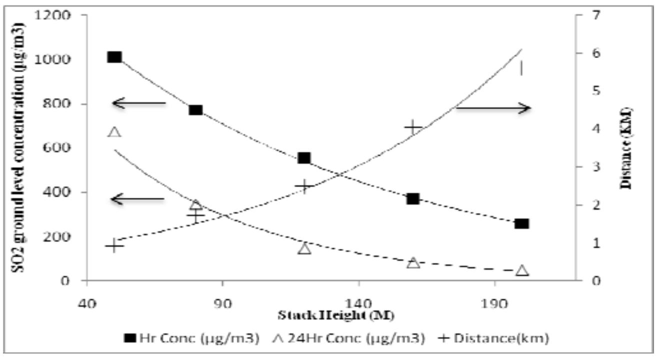

Fig. 5 Stack height vs. hourly and daily predicted ground level concentrations of SO_2

In scenario 2, SO_2 emission rate effect from FCC stack is tested at stack height of 80 m for different total monthly emission rates of 3000 g/s, 4000 g/s, 5000 g/s, 6000 g/s, 7000 g/s and 8000 g/s, taking into consideration the monthly emission variations (by using emission factors, table 2) and fixing other stack parameters i.e. exit temperature, exit flue gas velocity and stack diameter.

It is noticed from fig. 8 that the highest predicted hourly and daily ground level concentrations of SO_2 is substantially decreased as SO_2 emission rate is reduced. At 50% reduction in the emission rate, the highest hourly and daily ground level concentrations decreased by 50%.

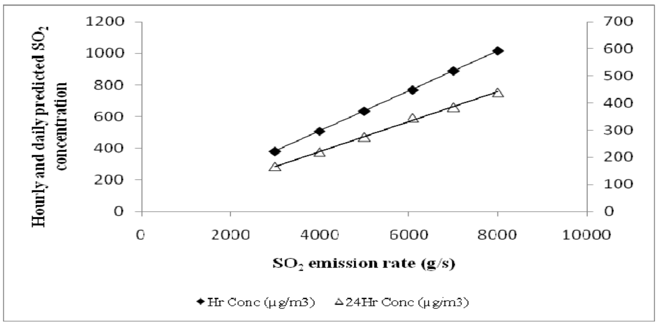

Fig. 6 SO_2 emission rate vs. hourly and daily predicted SO_2 ground level concentrations

In scenario 3, FCC stack diameter effect is examined at stack height of 80 m for different diameters of 1.5 m, 2.3 m, 3 m and 4 m. The exit flue gas velocity is also changed as directly related to the square of the diameter for a fixed exit flue gas flow rate. It is observed that the dispersion and rise of the plume are not affected by diameter variation and the predicted ground level concentration of SO_2 remained almost unaltered. The hourly and daily predicted concentrations of SO_2 are almost identical for all the cases.

Kulkarni et al., (2009) have reported that Lanthanum and Lanthanides are used as markers for particulate matters pollution as $PM_{2.5}$ in petroleum refineries, mainly from FCC units. US EPA daily $PM_{2.5}$ standard is 35µg/m^3. In the present work, the application of Aermod to predict ground level concentration of PM is considered as $PM_{2.5}$ for rare earth elements i.e. Lanthanum and Cerium. $PM_{2.5}$ is inhalable and has adverse impact on public health causing cardiovascular diseases. Kuwait EPA has no standard for $PM_{2.5}$ and has only specified daily and yearly standard for PM_{10}. Figure 5 shows the isopleths of the predicted hourly average ground level concentration of PM calculated at the selected uniform grid receptors.

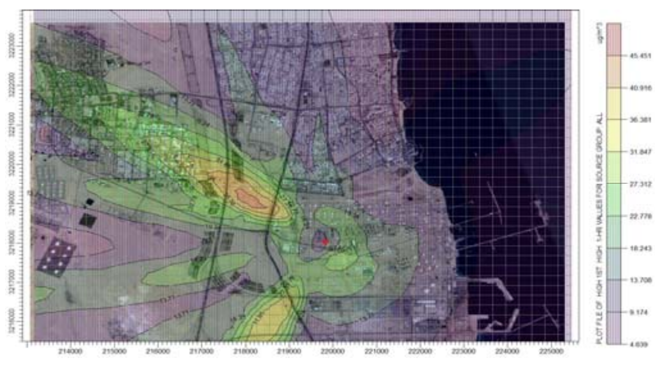

Fig. 7 Isopleths plot of the predicted hourly average ground level concentration of PM

The isopleths indicate the hourly predicted spatial variations of the ground level concentrations of PM. The maximum hourly predicted average ground level concentration of PM is equal to 45μg/m^3, observed on the 27th of February 2008 at 8:00 hour and about 1.56 km in the NW direction from the FCC stack, and at receptor coordinates of X = 218557.94, Y = 3218919.

Similarly, the predicted daily average ground level concentration of PM is compared with US EPA ambient air quality standards for $PM_{2.5}$ at all receptors. Figure 6 shows the isopleths of the predicted daily average ground level concentration of PM computed at the selected uniform grid receptors.

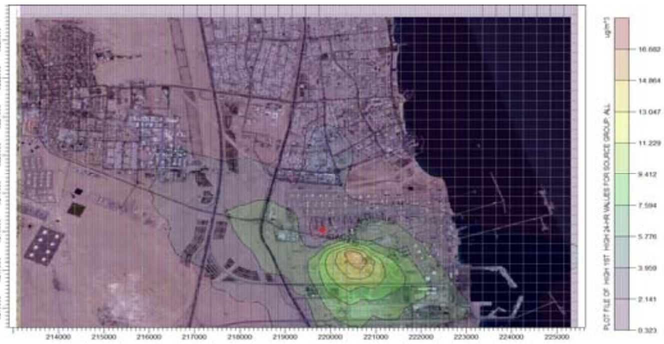

Fig. 8 Isopleths plot of the predicted daily average ground level concentration of PM

The isopleths indicate the daily average predicted spatial variations of the ground level concentrations of PM in the area of study. The highest daily predicted concentration is equal to 16μg/m^3, observed on the 29th of December 2008 and about 0.75 km in the SE direction from the stack, at a receptor coordinates of X = 220657.94, Y = 3217419 due to the influence of the prevailing meteorological conditions, especially the predominant North West wind and other meteorological factors.

To observe the computational model sensitivity, another scenario run is performed adding two finer meshes consisting of 21 x 21 uniform receptor points, the first one covering hourly highest ground level concentration area, the other one covering daily highest predicted ground level concentration area. The output accuracy has improved for both pollutants due to application of interpolation using small values of Δx = 150 m, Δy = 110 m for the first mesh and Δx = 100 m, Δy = 100m for the second mesh. There is 0.65% increase in the hourly highest ground level concentration and 2.8% increase in the daily highest ground level concentration, which are insignificant.

CONCLUSIONS

FCC unit in a refinery is a major contributor to SO_2 and PM emissions. These gases have adverse impact on the immediate neighbourhood of refineries. In this study a complete emission inventory for a year long period have been prepared for SO_2 and PM. A model run performed for actual monthly emission variation with total SO_2 emission rate of 6089.2 g/s and PM emission rate of 302 g/s, taking into consideration monthly emission factors for both SO_2 and PM.

The daily predicted ground level concentrations of SO_2 are compared with Kuwait EPA monitoring station daily measured SO_2 concentrations at the same discrete receptor and showed acceptable validation of the model output.

The highest hourly predicted concentration of SO_2 is equal to 769 μg/m^3. It is observed on the 8th of March 2008 at 8:00 hour, due to elevated SO_2 emission rate in this month and the prevailing meteorological conditions, especially sea breeze effect in the early morning hours. The highest daily predicted concentration is equal to 335μg/m^3. It is observed on the 9th of November 2008, and obviously influenced by the predominant North West wind and high SO_2 emission rate in the month of November.

The maximum hourly predicted average ground level concentration of PM is equal to 45μg/m^3. It is observed on the 27th of February 2008 at 8:00 hour. The highest daily predicted concentration is equal to 16μg/m^3, observed on the 29th of December 2008.

The stack sensitivity is explored by changing stack height, total emission rate and stack diameter independently. It is observed that the higher stack facilitated good dispersion, thus lowering the ground level average concentration of the pollutant up to 50% when the stack height doubled.

It is notice that the highest predicted hourly and daily ground level concentrations of SO_2 are substantially decreased as SO_2 emission rate is reduced. At 50% reduction in the emission rate, the highest hourly and daily ground level concentrations decreased by almost 48%.

The influence of stack diameter inherently changed the exit flue gas velocity due to invariable flue gas flow-rate. The plume rise and dispersion are related to the exit flue gas velocity, which decreased with the increase of stack diameter because of proportionality to the square of diameter. For a fixed load there is no noticeable change in the average hourly and daily predicted ground level concentrations of SO_2. The study results presented in this paper provide, for the first time, a comprehensive quantitative analysis of the impact of a typical FCC unit on its surrounding environment.

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