



Preliminary Re-evaluation of an Air Monitoring Network Based on Pollutant and Meteorological Measurements in the Area of Influence of an Oil Refinery in Brazil

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ABSTRACT

Landulpho Alves Refinery (RLAM) is located in the north coastal zone of Baía de Todos os Santos bay, in the district of Mataripe, part of the municipality of São Francisco do Conde, approximately 56 km far from the urban area of Salvador, Brazil. The entire site, including an asphalt plant, comprises 3.5 million square meters in the city of Madre de Deus, which is located 3.5 km from the Refinery. RLAM was the first refinery of PETROBRAS system in Brazil. The plant has operated since 1950 and has been presently undergoing several modernization changes aimed at becoming one of the largest oil units of an increasingly competitive market. As part of this policy, environmental preservation has always been one of the major concerns of the company. The daily operations of RLAM include processing up to 48,500 m³/day of crude oil and natural gas. Pollutants, such as NO_x, CO, PM, VOC and SO₂, are emitted to the atmosphere by refining or processing operations. A mathematical simulation was performed in order to support the installation of an automatic air-quality measurements system within the refinery area of influence. The simulation was based on emission and meteorological parameters and aimed at defining strategic metering points for pollutant concentrations within the targeted area. The modeling outputs revealed the points where maximum concentrations of pollutants should be



observed and, therefore, where the monitoring grid should comprehend: Candeias, Madre de Deus, São Francisco do Conde and Fazenda Ouro Negro. This paper re-evaluates these points by means of a new mathematical simulation, based on actual data measured by the stations, and compares calculated figures to the results of automatic grid measurements. This first analysis included only SO₂ and NO_x emissions. The results indicate an expressive difference between measured and calculated values, considering normal operational conditions of the refinery and the same meteorological conditions. Based on these results, and considering the preliminary character of the analysis, this paper recommends further studies, including climatic seasonality parameters, in order to re-evaluate the location of metering stations to support a future positioning of the automatic stations of the grid, if required.

INTRODUCTION

Landulpho Alves Mataripe Refinery (RLAM), located in Bahia state, was the first refinery of Brazil and it is part of the Business Units of PETROBRÁS Holding, a mixed economy company with 51% from the government capital shares. The refinery has operated since 1950 and is presently the second largest unit out of the thirteen refineries of the country, being also among the ten largest companies of Brazil in terms of incomes (US\$ 2.3 billions in 2002). RLAM processes up to 48,700 m³/day, installed in a 3.5 million square meters site, which includes an asphalt plant. The plant produces over 43 types of products, including gasoline, diesel oil, natural gas, LPG, naphtha and lube oils. As part of the company Management Policy, pollution prevention, minimizing environmental risks and the social accountability before nearby communities are aspects that play an important role and rule actions throughout the entire productive chain. In order to operate in compliance with the law, the plant must have an Operation License granted by the environmental state agency, CRA (“*Centro de Recursos Ambientais*”), which required, among a number of conditions, studies addressed to the implementation and operation of an air monitoring network. In order to comply with both legal requirements and the company environmental policy, RLAM performed important studies addressing climatic aspects and the behavior of pollutants within the site area of influence. An inventory of pollution sources was performed for the 56 units that compose RLAM, focusing, among other elements, the emissions of NO_x, CO, PM, VOC and SO₂, which are typical emissions of oil processing companies. The automatic air monitoring network has started operating in May 2002, aimed at assessing the air quality if the urban areas influenced by the plant, in compliance with the standards defined by Resolution CONAMA 003/90 (National Environmental Council). The main steps of the network design comprised: inventory of air emissions, application of the mathematic model U.S. EPA ISCST; characterization of micrometeorological conditions of the region and information on local topographic features. The study identified four areas where the network stations should be located. This paper reviews some aspects of the study by running a new mathematic simulation using the model ISC3^{1, 2}, using air emission and meteorological inputs from the previous study, as well as current concentration and meteorological data measured by the automatic air quality network. The results of the first simulation are then compared to both the current simulation and data measured



by the network. Only SO₂ and NO_x emissions were addressed in this analysis, based on normal operational conditions of the plant and similar meteorological conditions.

LOCATION

Landulpho Alves Refinery (RLAM), is located in the north coastal zone of Baía de Todos os Santos bay, at latitude 13° S and longitude 38° W. The entire site, including an asphalt plant, comprises 3.5 million square meters in the city of Madre de Deus, which is located 3.5 km from the Refinery. The refinery is installed in the district of Mataripe, part of the municipality of São Francisco do Conde, approximately 56 km far from the urban area of Salvador, Bahia state, (Figure 2). Other important receptors located nearby include the urban perimeter of Candeias and the following villages: Passe, Monte Recôncavo, Caípe, Socorro, Paramirim, Santo Estevão, Cinco Rios, Pitanga and Ponto Ferrolho, as well as the entire urban perimeter of Madre de Deus island. The topography of the area of influence is considerably smooth, with heights ranging between 5 and 180 m. The dispersion model will be applied to this region, as depicted in Figure 1.

Figure 1 – Study area and meteorological domain.

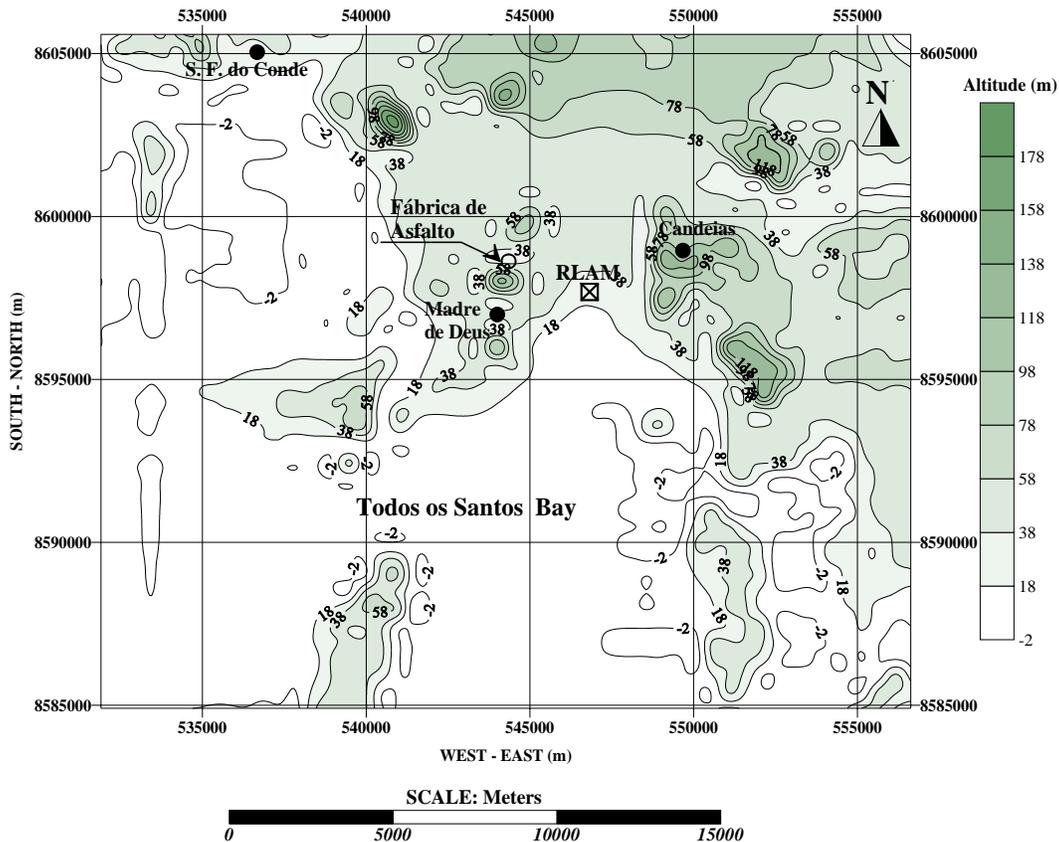




Figure 2 – Study area in Bahia state, Brazil.



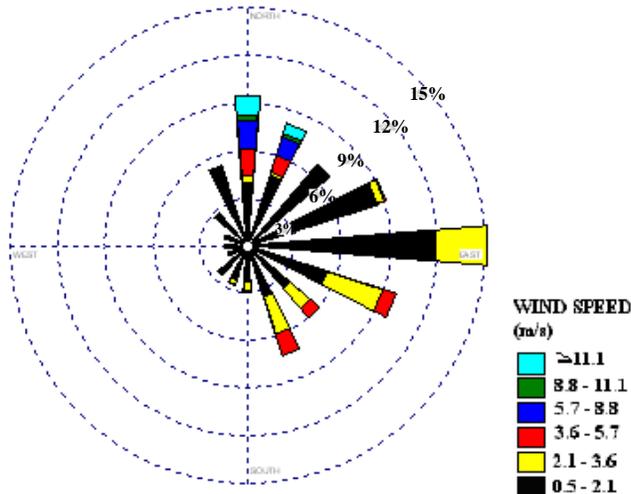
Meteorological Factors Affecting Air Quality in the Mataripe Region

The local climate of Baía de Todos os Santos area is quite peculiar, dominated by maritime circulation and ruled by the effects of both maritime and terrestrial breezes. The small scale air circulation, associated to daily variations on several meteorological parameters, namely atmospheric stability and wind direction and speed, defines specific pollutant dispersion conditions over the Refinery area. This phenomenon displays two well defined characteristics: a daytime pattern, when the wind is blowing from sea to continent over the entire influence area of RLAM, sometimes reaching the municipalities of São Francisco do Conde, Candeias and Santiago do Iguape; and a nighttime pattern, when the air flow is reversed and the winds blow from continent to ocean. The climate of the northeast region of the Brazil changes according to seasons and to the movement of both cold fronts and high-pressure systems (subtropical maritime anticyclone) toward Bahia state. These changes are directly mirrored by pollutant dispersion capacity of the atmosphere. The behavior of the coastal climate of the bay has been studied based on a three-year series of meteorological data (1998 – 2000)³ generated by all four automatic stations (PP4, COPES, Entrepasto and Estação de Água) installed in the influence area of the Refinery. This analysis aims at identifying the trends of the local circulation in terms of pollutant dispersion. The regional mean temperature is 25° C, with a minimum mean of 22.7° C and a maximum mean of 28.° C. The prevailing winds blow from E (16%), at mean velocities ranging between 2.25 and 3.17 m/s, as observed in Figure 3, which depicts the wind roses for 1998 and 1999. Further details about the local climate of the region of Baía Todos os Santos can be found in the considered study⁴. Temperature, wind and other important data were used in the dispersion model ISC3, as well as the data generated by the automatic air quality network concerning the period between May and December 2002.



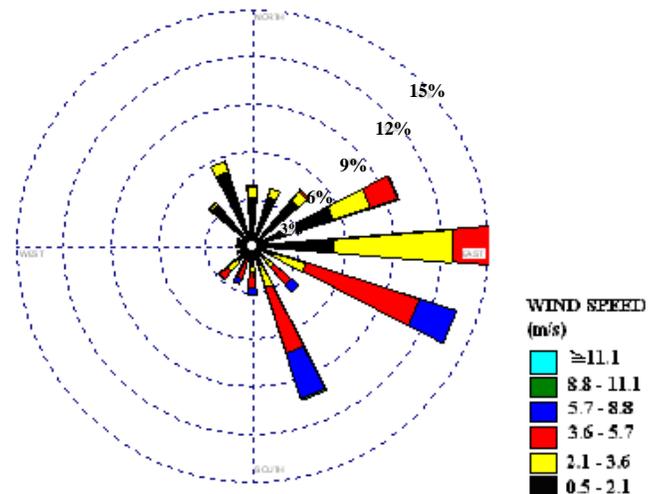
Figure 3. Annual wind rose for 1998 and 1999. RLAM stations.

METEOROLOGICAL STATION - RLAM - 1998



CALMS WINDS FREQUENCY: 6.97%
AVERAGE WIND SPEED: 2.25 m/s

METEOROLOGICAL STATION - RLAM - 1999



CALMS WINDS FREQUENCY: 4.61%
AVERAGE WIND SPEED: 3.17 m/s

Considerations About Processing Meteorological Data

In the first dispersion study addressed to designing RLAM monitoring network the meteorological data file required for the simulation was built as follows: the hourly mixing height was measured with an acoustic sounder equipment in the municipality of Camaçari, which is located 70 km far from the site; the wind direction and speed and the air temperature were measured at RLAM automatic station and the atmospheric stability class was defined based on the standard deviation of the horizontal direction of the winds as measured by RLAM station.

The preparation of the meteorological data files considered follows method: wind direction inputs were considered 180° added whenever the wind direction was equal or less than 180° ; and 180° were subtracted whenever the direction was equal or higher than 181° . However, the preparation of these data files indicated a number of problems, as follows:

- The transformation of wind direction as described above considers the wind as scalar quantity, only. However, winds are vectorial quantities and, therefore, any changes on the winds should take in account both the wind speed and direction and not only one of them. This error was corrected in this study, which considers the resulting wind flow vector in terms of its intensity and direction.



- The mixing layer heights obtained with acoustic sounder equipment in Camaçari were adjusted using other method based on temperature profile measured by radiosonde data from Salvador Airport to RLAM location. The refinery is located on the shoreline, under strong influence of maritime breezes that are often observed at Baía de Todos os Santos bay. Therefore, the mixing layer height over the bay area is influenced by the coastal mixing layer and not by the continental layer measured in Camaçari. 70 km far from the coastal zone of Mataripe.
- The atmospheric stability class definition was performed based on data from RLAM meteorological station. This parameter has a straight relation to the mixing height fluctuations over the influence area of RLAM. The fluctuations on the mixing height depend on the changes on the atmospheric stability in the same place where the mixing height is being measured⁵. Therefore, the atmospheric stability defined for the vicinities of RLAM can not influence the hourly oscillation of a mixing layer located 70 km (in Camaçari) far from the refinery (in Mataripe region), since the atmospheric stability and mixing height are phenomena related to local meteorology aspects, whose sensible and convection heat flows prevail in the air turbulence within the coastal marine boundary layer surface.

Based on these considerations, this study reviewed and the same hourly meteorological data used in the previous study and measured by RLAM automatic stations in 1998 and 1999 and re-processed these data using the software PCRAMMET⁶, taking in account the local micro-meteorological aspects required to build hourly ASCII files comprising the atmospheric stability, urban and rural mixing layers and wind flow vectors, including associated flow direction and mean speed. After properly built, these files were used as inputs in the program ISC3. Subsequently, hourly meteorological data from 1998 and 1999 were condensed in a single file by means of a RAMMET View application, *The Five Year Preprocessed Met File Utility*. Likewise, the calculation of the mixing layer heights was performed likewise, but using *The Mixing Height Data Utility* and based on hourly meteorological data measured by RLAM ground station.

Emission data

Pollutant emission rates were an important set of data of previous dispersion study. The present study was performed as part of the monitoring network project, which describes the assumed methodology and presents all surveyed data⁷. The full inventory was based on a survey comprehending 56 significant sources within the site. The inventory comprised four types of sources: point source, fugitive, area sources and mobile. The point sources included stacks, furnaces and boilers, whose emissions result from firing fuel oil and natural gas, totalizing 42 sources, out of which 38 are presently operating in the site. The inventory included fuel consumption and composition data, flow rate and temperature of flue gases, operation time and stack diameters and heights. The total consumption figures of fuel at RLAM in 1998, as indicated by the inventory results, are presented in Table 1.



Table 1. Fuel consumption at RLAM in 1998.

Sources	Fuel	Fuel oil (t/year)	Natural gas (t/year)
RLAM		185,967.0	1,156,420.0
Asphalt plant (FASF)		11,717.0	-
Total		197,684.0	1,156,420.0

ANALYSIS OF RESULTS

This item comprises three parts: presentation of the results of the initial concept of RLAM monitoring network; presentation of the results obtained by this study, using ISC3 model for the meteorological data from 1998 and 1999; and the comparison between both studies and the data measured by the automatic air quality network in 2002.

Results for the Network Project

The results of the first simulation applied to the network project are displayed in Table 2, where it can be observed that the maximum 240h mean concentration of SO₂ was 689.93 µg/m³, which is 90% higher than the corresponding standard for this pollutant, 365 µg/m³. In terms of NO_x, the maximum 1-h mean concentration was 1,124.84 µg/m³, which is 251,5% higher than the hourly standard for NO₂, 320 µg/m³ for moderately stable atmospheric conditions⁸. Therefore, this study, modeled downwind ground-level concentrations it was reported in terms of total NO_x instead NO₂ (i.e. the calculation assumes that all NO_x released to the air is converted to NO₂). This is a conservative assumption, considered for authors, in this case resulted in violations of the NO₂ – air quality standards.



Table 2. Maximum concentrations obtained for the targeted pollutants within RLAM area of influence and corresponding meteorological parameters. First simulation⁸.

Parameters		Maximum Concentration ($\mu\text{g}/\text{m}^3$)	Air Quality Standard ($\mu\text{g}/\text{m}^3$)	Stability Class	Wind Velocity (m/s)	Receptor distance from source (km)	Receptor grid point
SO ₂	24h	689,9	SO ₂ - Air Quality Standard – 365,0	4	4,4	6,3	FASF and vicinities
	Annual	224,6	SO ₂ - Air Quality Standard – 80,0	4	3,0	6,3	FASF
NO _x	1 hour	1.124,8	NO ₂ - Air Quality Standard – 320,0	6	0,7	8,9	North from Monte Recôncavo
	Annual	34,2	NO ₂ - Air Quality Standard – 100,0	4	3,0	6,3	FASF

¹ FASF – asphalt plant.

Results for the Current Simulation

The results of the current simulation, performed by ISC3 View, are displayed in Table 3, where it can be observed that the maximum 24-h mean concentration obtained for SO₂ was 402.3 $\mu\text{g}/\text{m}^3$, which is 10.2% higher than the corresponding standard, 365 $\mu\text{g}/\text{m}^3$, and 41.7% lower than the concentration obtained in the first simulation, 689.93 $\mu\text{g}/\text{m}^3$. In terms of NO_x, the maximum 1-h mean concentration calculated by the model was 591.0 $\mu\text{g}/\text{m}^3$, which is 84.7% higher than the corresponding NO₂-air quality standard 1-h, 320.0 $\mu\text{g}/\text{m}^3$, and 47.49% lower than the maximum concentration observed for this pollutant in the first study, 1124.4 $\mu\text{g}/\text{m}^3$. The results for SO₂ were obtained for a mean wind velocity of 1.1 m/s and an atmospheric stability class 6. The hourly mean concentrations for NO_x were calculated using a mean wind velocity of 1.5 m/s and atmospheric stability class 6, while the annual means were calculated using 2.7 m/s and neutral stability class 4. The mean wind velocities used in this simulation are typical of the targeted region (weak winds) and lower than the values used in the first simulation. It can be observed that the maximum concentrations were found for practically the same areas in both cases, except for the maximum hourly concentration of NO_x that, in the first study, was observed at Monte Recôncavo and in this study was found in the vicinities of Candeias. This city is located northeast the refinery, while Monte Recôncavo is located to the north. The annual mean concentrations for both SO₂ and NO_x did not exceed the corresponding standards in the present simulation. In the first simulation, the annual concentration of SO₂ was surpassed.



Table 3. Maximum concentrations for SO₂ and NO_x within RLAM area of influence and corresponding meteorological parameters. Current simulation.

Parameters		Maximum Concentration (µg/m ³)	Air Quality Standard Resolution. (µg/m ³)	Stability Class	Wind velocity (m/s)	Receptor distance from source (km)	Receptor grid point
SO ₂	24h	402,3	SO ₂ - Air Quality Standard – 365,0	6	1,1	6,8	FASF and vicinities
	Annual	45,6	SO ₂ - Air Quality Standard – 80,0	4	2,7	6,7	FASF and vicinities
NO _x	1 hour	591,0	NO ₂ -Air Quality Standard –320,0	6	1,5	5,1	NE RLAM and close to Candeias.
	Annual	45,6	NO ₂ - Air Quality Standard –100,0	4	3,0	6,3	FASF and vicinities

¹ FASF – asphalt plant.

Comparison Between Calculated and Measured Values

The air quality network comprises four automatic stations located in the following places: Madre de Deus Island (5.8 km away from RLAM); São Francisco do Conde (14.4 km); Malembá (4.1 km) and Fazenda Ouro Negro (10.5 km). The network monitors SO₂, NO_x, H₂S, NO, NO₂, VOC, methane and non-methane hydrocarbons, O₃, CO, Particulate Matter and meteorological parameters, but, as already mentioned, only SO₂ and NO_x were addressed in this study. The monitoring network has started operating in May 2002, generating data every 15 minutes at all four stations. This analysis used air quality and meteorological data for the period between May and December 2002 for the considered pollutants. Out of this set of data, the highest maximum mean concentrations were extracted for all four stations. The maximum concentrations were then calculated by selecting the days and times of the maximum mean occurrences and processing the corresponding data within the respective mean intervals, i.e., 1 hour mean for NO_x and 24 hour mean for SO₂. In order to generate the data used for comparison purposes, the model ISC3 used as inputs the 2002 meteorological data file from RLAM automatic station for the same dates when the maximum concentrations were recorded in each station and the corresponding emission parameters for SO₂ and NO_x from the source inventory. The simulation was then performed for the dates when the maximum concentrations of the considered pollutants were measured in all four stations. The results of these steps are compared to the measured values and presented in Table 4.



Table 4. Comparison between the values calculated by the model and the values measured by the automatic stations for SO₂ and NO_x.

Automatic station	Concentration (µg/m ³) SO ₂ (24h)		Concentration (µg/m ³) NO _x (1h)		Air quality standard (µg/m ³)	
	Measured	Calculated	Measured	Calculated	SO ₂ (24h)	NO _x (1h)
Ouro Negro	17.8	12.6	41.3	30.6	365.0	320.0
São Francisco do Conde	46.2	50.0	55.7	61.0		
Madre de Deus	30.4	41.5	107.9	172.0		
Malembá (Candeias)	124.7	104.8	353.3	437.4		
R ² Statistic	0.94		0.93			

The Table displays the comparison between the concentrations calculated by the model ISC3 and measured at the automatic stations, for SO₂ and NO_x. The results indicate a positive coefficient of determination (R²): 0,94 for the SO₂, characterizing an excellent adjustment between measured and calculated values and 0,93 for the NO_x, also indicating a good adjustment but with data a little more scattered than the SO₂. This difference was caused by the nature of the measurement and the calculation used for each pollutant: 24-h mean for the SO₂ and 1-h mean for the NO_x. Figures 4 and 5, next, depict the positive adjustment between measured values and those calculated by ISC3 model. It was observed that, for both cases, the highest concentrations are observed at Malembá station, the closest downwind station to the plant. The lowest concentration was observed at Ouro Negro station, which is located 10.5 km away from the refinery. Although the NO_x hourly standard (320 µg/m³) was exceeded in 10.4% (353.3 µg/m³), this concentration is still far from the maximum value calculated by the first dispersion study (1,124.4 µg/m³).



Figure 4. Comparison between measured and calculated SO₂ concentrations.

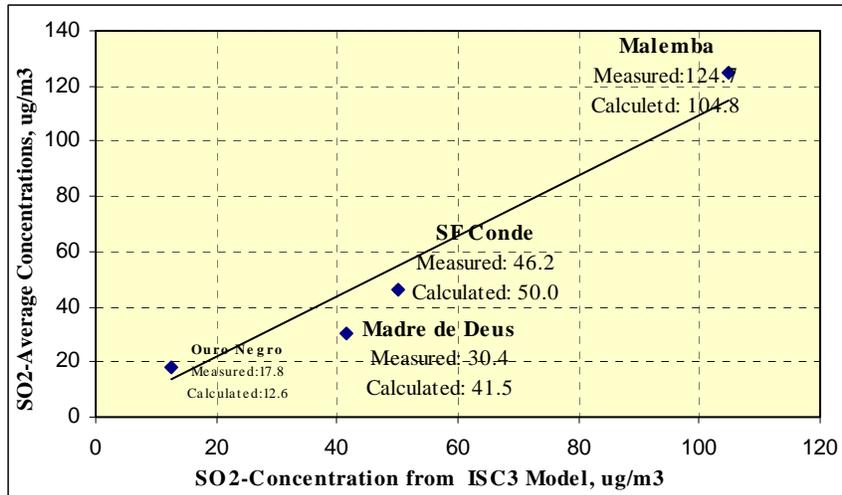
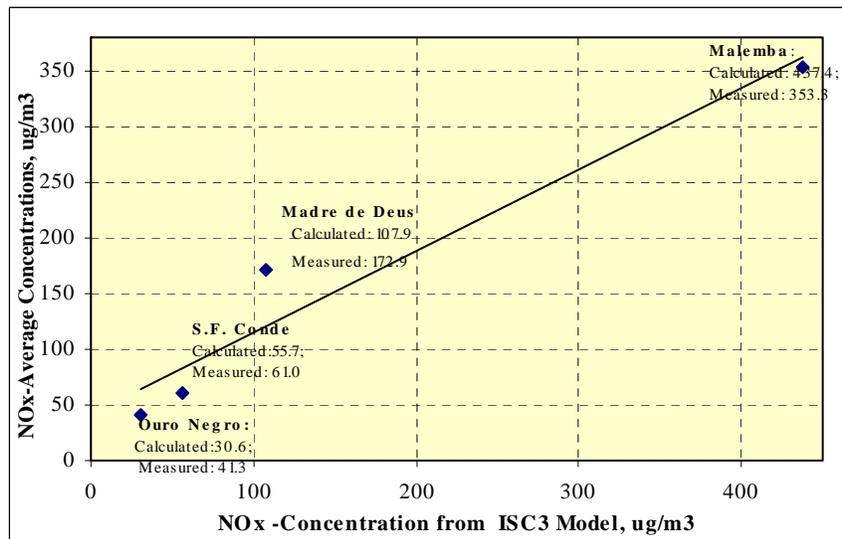


Figure 5. Comparison between measured and calculated NO_x concentrations.



CONCLUSION

This study aimed at reviewing an atmospheric dispersion study performed to design an automatic air quality monitoring network to be installed in the influence area of Landulpho Alves Refinery, RLAM, of PETROBRÁS, namely in the municipalities of Mataripe, Candeias, Madre de Deus, São Francisco do Conde and Fazenda Ouro Negro. The network was designed considering a



survey on meteorological conditions, the inventory of the refinery sources and the topographic characterization of the region. The results of the network project indicated that the air quality of the region was highly compromised as a result of high concentrations of a number of pollutants in the air, including nitrogen oxides, NO_x , and sulfur dioxide, SO_2 . Based on the results of the study, four areas were selected for the installation of the automatic stations addressed to monitor the pollution caused by the refinery emissions. The network design considered monitoring SO_2 , NO_x , H_2S , NO , NO_2 , VOC, methane and non-methane hydrocarbons, O_3 , CO , Particulate Matter and meteorological parameters. Nevertheless, only NO_x and SO_2 were addressed by this study, since pollutants presented the highest concentrations when compared to the corresponding air quality standards. The current study considered the following aspects:

- Re-assessing of hourly meteorological data for 1998 and 1999 measured by the meteorological station installed within the refinery site. This analysis was performed by using the application PCRAMMET, which generated ASCII files that were used as inputs into the mathematic model ISCView3;
- The new approach considered the same emission data from the previous study and the same topographic file of the region within the simulation grid.
- The dispersion was simulated by using the mathematical model ISC3. The first study used the program U.S.EPA ISCST.

The concentrations of SO_2 and NO_x calculated in this review were significantly lower than those observed in the network design. Subsequently, the concentrations of the targeted pollutants were calculated at each air quality station, using as inputs the emission data from the source inventory and both the meteorological conditions and temporal concentrations of SO_2 and NO_x measured by the monitoring network between May and December of 2002. The maximum concentrations of both pollutants were selected from that measurement period, maintaining the same meteorological conditions and considering the respective mean values defined by the federal air quality standard. The model was then applied to each meteorological condition that caused an increase in the concentration at each station. The values calculated by the model ISC3 were then compared to the concentration measured by each station, resulting in a R^2 statistics positive and higher than 0.94 and 0.93.

The study concluded that the maximum 1-h mean concentration of $1,124.84 \mu\text{g}/\text{m}^3$ for NO_x and the maximum 24-h mean concentration of $689.93 \mu\text{g}/\text{m}^3$ for SO_2 should not be reached in the influence area of RLAM, as demonstrated by the simulation performed in this study and by the maximum concentrations of these pollutants recorded by the automatic station between May and December of 2002. Although the total NO_x hourly in comparison with NO_2 -standard ($320 \mu\text{g}/\text{m}^3$) was surpassed in 10.4% ($353.3 \mu\text{g}/\text{m}^3$), this value is significantly lower than the maximum concentration of $1,124.84 \mu\text{g}/\text{m}^3$ calculated by the dispersion study performed for designing the air quality network

Concentrations as high as that should only be reached as a result of an accidental release of those



pollutants. Still, high long-term concentrations would not be observed at the ground level as a result of the good atmospheric dispersion conditions observed along Baía de Todos os Santos area and its vicinities.

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Key Words

Emissions rate, Air pollution, Inventory, Monitoring Network, Mixing height, Shoreline, Stability, Micrometeorological, Wind direction.