

Assessment of the contributions of the highest NO₂ concentrations by industrial sources using AERMOD dispersion model

M.F. Abari¹, S. M. Zarandi^{2*}, N. Mansouri¹

¹ Department of Environment and Energy, Science and Research Branch, Islamic Azad University, Tehran, Iran

² Department of Environmental Health Engineering, Health Faculty, Shahid Beheshti University of Medical Sciences, Tehran, Iran

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The Environmental Protection Agency (EPA) recently announced a 1-hour NO₂ standard based on the multiyear average of the 98th percentile of the annual distribution of daily maximum 1-hour values, not exceeding 188 µg/m³. The AERMOD-the EPA's preferred dispersion model for near-field applications was recently modified to fully support the form of the 1-hour NO₂ National Ambient Air Quality Standard (NAAQS), which contains three different NO-to-NO₂ conversion methods for estimating the ambient concentrations of NO₂. The prevalence of nitrogen oxide (NO_x) emissions from the Esfahan Oil Refinery Complex, petrochemical and power plant is in the form of nitric oxide (NO) rather than NO₂. NO gases in the emission plume mix with the atmosphere and react with ozone and other oxidants to oxidize a portion of the NO to NO₂. In this study, the contribution of industrial groups in randomly selected receptors in all the directions within three, six and 30 kilometres from the centre point were determined in relation to the screening approach of Tier I, assuming full conversion of NO to NO₂. The results showed that the cumulative first highest max daily contribution 1-hour value averaged over 1 year (2012) equalled 26.7 and 25.8%, calculated from total NO₂ ground level concentration of 590.7 and 571.973 µg/m³ in the West and South receptors(3km). Contributions of 15.5 and 10.6% were devoted to the receptors located within 6km and 30 km respectively. While 36 and 50% SE receptor first-highest max concentration were portioned to the petrochemical and refinery, the total of cumulative apportionment in the SW receptor was released from the power plant. The cumulative first-highest max daily contribution of 1-hour value during 2012 reached to 2218.329 (µg/m³); that highest one 88.6% (1966.4 µg/m³) was related to the refinery. As well as, the rank of 50 highest 1-hour NO₂ that indicated the exceeding the values of EPA's Standards only distributed in 34 receptors in south and 16 receptors in west over year throughout the domain. The max 1-hour concentration of NO₂ in Esfahan city receptor was 73.7 (µg/m³) that was below the standard level.

Keywords: EPA, Cumulative, industrial groups, receptor, 1-hour NO₂

INTRODUCTION

Rapid increase in industrialization, urbanization, communication and transport systems have been well recognized as one of the major issues impact on environmental pollution which concern the quality of life in urban areas across the globe. Investigations has made it clear that noise pollution [1], as well as groundwater high nitrate concentration from agricultural activates [2] has reached to a hazardous level over the years.

Among pollution issues, poor air quality attracts a high level of interest within the scientific community and engages public opinion because of the known relationship between exposure to many air pollutants and increased adverse short- and long-term effects on human health [3-9]. In fact, poor air quality is generally result of increasing levels of gaseous pollutants, which are mainly considered toxic for humans and other living organisms due to their extensive natural or anthropogenic activities [10]. The NO_x gases are

an important type of these gaseous pollutants and usually emitted by fuel combustion sources in the form of nitric oxide (NO), and in smaller quantities as NO₂ gas [11]. The NO gases in the emission plume mixes with the atmosphere and reacts with ozone and other oxidants to oxidize a portion of the NO to NO₂. There are numerous other atmospheric reactions of NO_x species; these include further oxidation of NO₂ to nitrate radical (NO₃) and nitric acid (HNO₃), as well as photo-dissociation of NO₂ back to NO through the absorption of UV radiation during the daytime [12]. However, during the early stages of the dispersion of a NO_x emission plume (i.e., at distances ranging from approximately 0.1 to 10 km over time intervals of 10-300 min), the principal NO_x reaction is NO oxidation by ozone to form NO₂ [13]. In reference to these issues, EPA has promulgated a 1-hour average National Ambient Air Quality Standard (NAAQS) for NO₂, and major sources of NO_x emissions must estimate their NO₂ air quality impacts as part of EPA's air quality permitting programs. The AERMOD (The American Meteorology Society-Environmental Protection Agency Regulatory Model) dispersion model has been

* To whom all correspondence should be sent:
E-mail smotesaddi@sbmu.ac.ir

developed by EPA for these air quality impact analyses, and AERMOD contains three different NO to NO₂ conversion methods for estimating the ambient concentrations of NO₂ [14]. EPA's "Guideline on Air Quality Models" (GAQM), 40 CFR Part 51 Appendix W, describes a three-tiered screening approach to calculating NO₂ concentrations based on dispersion model predictions of NO_x concentrations (NO_x is modeled as if it is a conserved or non-reactive tracer). The three tiers, arranged in order from simplest to most refined, are:

Tier 1 – Assume full conversion of NO to NO₂, so that the NO_x predicted by AERMOD is 100 % NO₂

Tier 2 – Ambient Ratio Method (ARM), where model predicted NO_x concentrations are multiplied by a NO₂/NO_x ambient ratio, derived from ambient monitoring data

Tier 3 – More detailed methods that account for the plume dispersion and chemistry may be considered on a case-by-case basis, including the Ozone Limiting Method (OLM) and the Plume Volume Molar Ratio Method (PVMRM) [15].

The Total Conversion Method assumes 100 percent conversion of NO to NO₂. This is the simplest and most conservative method of evaluating NO₂ impacts from NO_x sources. Due to the conservative nature of the method, no justification is needed for its use and it is often applied as the screening method for the assessment of NO₂ impacts (Level 1 assessment) in various jurisdictions [16].

In Maptaphut area, AERMOD had been utilized by several studies. Chusai *et al.* (2012) used AERMOD to evaluate dispersion of NO₂ and SO₂ and relative roles of emission sources over this area. It was found that predicted data of both pollutants were under-estimated when compared with those observed data [17]. Results also indicated that petrochemical industry played the major contribution in annual average area-wide concentrations of NO₂ and SO₂ in this area. As well as a study of AERMOD tiering approach for NO₂ prediction in this industrial area was conducted by [18]. Three methods were tested for their performance in modelling NO₂ concentrations (Tier I: total conversion of NO_x to NO₂; Tier II: NO₂/NO_x ratio of 0.60 and Tier III: ambient O₃ concentrations were used for calculation using the plume volume molar ratio method (PVMRM)). The results indicated that Tier 1 provided less bias with those measured data as compared with other tiers. It also performed very well in predicting the extreme end of NO₂ concentrations. This study

recommended that Tier 1 was appropriate for prediction of the average as well as in determining the maximum ground level concentration of NO₂ in the Maptaphut industrial area [18].

In reference to these findings and given that the expansion and establishment of several industrial plants, including oil refineries, petroleum products, and power plant are major emission sources of NO_x particularly in the industries areas. Well as scientific researches focus on the analysis of the relative contributions from different activities to air quality reported that it is very important because such data are necessary to determine the types of sources that are most effective on average air pollutant concentrations in industrial complexes [19]. Specifically, air quality models have proven useful for determining the spatio-temporal distribution of air pollutants and for developing emission control policies that allocate limits to air pollutant emissions [20-22]. Furthermore, dispersion modeling describes the transport and dispersion of air pollutants, as well as chemical and physical processes within the plume. Such data enable researchers to better analyze air pollutant concentrations in various areas [19]. According to the statement contained and taking into consideration the fact that the establishment and development Oil Companies by ministry of petroleum in Isfahan has led to the construct complex dependent industries that cause NO_x pollutants cumulative release. Whereas AERMOD is the preferred model for dispersion of a wide range of application such as determination of the daily contribution 1-hour average NO₂ concentration, in this research in order to reach this goal Tier 1 applied to the 1-hour NO₂ contribution without any additional justification.

This study aims to (i) estimate, first of all, the highest maximum daily contribution 1-hour NO₂ values averaged over 1-year for source groups, including the refinery, power plant, and petrochemical and dependent industries for 13 randomly selected receptors, (ii) determine temporal and spatial distribution of 50 highest 1-hour NO₂ ground-level concentrations in all receptors during 2012, and (iii) compare 1-hour NO₂ concentrations using 1-year meteorological data with NAAQS (188 µg/m³).

MATERIALS AND METHODS

Esfahan oil companies

Esfahan Oil Refining Company (E.O.R.C.) started its activities in the refining of raw oil and production of oil-related products on 1980 and now

produces more than 22% of each of the oil-related internal products. E.O.R.C. supplies light lube cut fee to Sepahan Oil, vacuum bottom to JEI Oil, platformate to Esfahan Petrochemical and straight-run kerosene to L.A.B, which is located to the northwest of Esfahan city, at an altitude of 1,685 metres above the sea, in an area of about 340 hectares along the Esfahan-Tehran highway. The Esfahan refinery has seen much progress in crude oil refining per day-there were so many products in the early 1990s-and the crude oil refining capacity of the company increased 85% compared to the design capacity of 200,000 barrels per day, which has increased to more than 375,000 barrels [23].

Esfahan Shahid Montazeri Power Plant is located above the Esfahan Refinery on 2.2 million m² land, while L.A.B is located on the western side of the power plant. Esfahan Petrochemical is the first producer of the aromatic line of chemicals in an area of 170 hectares, located to the south of Esfahan refinery.

AERMOD model

AERMOD is a steady-state Gaussian plume model that contains algorithms to simulate plume rise and turbulent atmospheric mixing and dispersion processes [14]. It is recommended by the US EPA to examine the effects of sources on receptor that are generally within 50 km of the source [24] (US EPA, 2004). AERMOD is the current preferred model for 'a wide range of regulatory applications in all types of terrain' for purposes of estimating ambient concentrations of NO₂, based on NO_x emissions, under Tiers 1 and 2 and 3 [25].

Two pre-processors-AERMAP and AERMET-are required in order running AERMOD. AERMAP is a terrain pre-processor that characterizes the terrain and generates receptor grids, discrete receptors, and elevation for AERMOD. In AERMOD, when specifying discrete receptors, it is necessary to specify the position of a source relative to which the receptor is assigned [26]. Gridded terrain data are used to calculate a representative terrain-influenced height (hc), associated with each receptor location, and to calculate the dividing streamline height. The gridded data needed by AERMAP is selected from digital elevation model (DEM) data [27].

The surface and profile meteorological data, used in this study, were derived from NOAA/ESRL pre-processed meteorological data. Data periods read from meteorological data files were started on the first hour of 1st January to the 24th hour of 31st December 2012. The gridded data required by

AERMAP were selected from Digital Elevation Model (DEM) data and the terrain data were collected during the Shuttle Radar Topography Mission (SRTM). In this study, a comprehensive Uniform Cartesian grid-extending to 30 km from the centre of the emission source-was used in the AERMOD model. Also, the number of 13 Cartesian discrete receptors was selected randomly in all directions, at distances of 3, 6, and 30 km away from the centre-point, and the Esfahan receptor was chosen as a representative of the highest population point. In this study, Tier 1-full conversion of NO_x to NO₂ (100% conversion)-was used as an assumption for NO₂ prediction.

Emission data

Emission data were obtained from (DOE) database, measured by a reliable laboratory. These data were reported by each factory annually for air pollution monitoring. This study classified industry into six source groups: 1) Linear Alkyl Benzene (L.A.B), 2) JEI Oil, 3) Sepahan Oil 4) Petrochemical industry, 5) Power plant and 6) Refinery. In toto, there were 74 stack data, which were located in this industrial area. Characteristics of stack emission source were as summarized in Table 1.

RESULTS

The output of the pre-processors

The AERMET pre-processor presented the wind rose, which shows the frequency of occurrence of winds as well as their strength and direction. The wind rose depicts whether the predominant wind direction is respectively from the West, North West and South West. Calm winds comprised 55% of the total winds (Fig. 1). Terrain contours were processed by the AERMAP (Fig. 2) and showed elevation dense isoclines located in the west to west north of the study area. In general, isoclines of altitude varied from 1,539 to 2,527 metres.

Contributions of 1-hour average concentration of Source Groups

The first highest maximum daily contribution 1-hour values averaged over 1-year of emission sources assessed by AERMOD in this connection-the contribution of emission sources were determined in the receptor groups in the four directions at a distance of approximately 3 km (North, East, South and West), 6km (North2, East2, South2, and West2), 30 km (NE, SE, SW, and NW) and Esfahan (Table 2). Hence, in the table, it can be seen that total 1-hour average concentrations were 184.69 µg/m³ in north receptor, with contributions

Table 1. Characteristics of stack emission sources by Mean+S.D.

Stack emission group	Number of stack	Stack height (m)	Stack diameter,(m)	Stack exit Temperature,(°C)	Stack exit Velocity, (m/s)	NO _x Emission rates (g/s)
Petrochemical Refinery	8	27±9.33	1.5±0.44	255.69±226.6	10.4±5.13	1.5±1.74
Power plant	49	59.6±13	2.4±0.99	420±153	7.9±1.9	11.5±11.3
JEI Oil	8	210±0	3.1±0	171.5±50	19.3±1.24	43.97±14.6
Sepahan Oil	2	38.5±2.12	1.2±0	686 ±22.12	15±0	1.72±0.14
LAB	3	35±2	1.45±0.25	344.8±90.6	15±0	6.97±2.1
	4	59.75±5.44	1.87±0.62	344.85±47.3	15±0	5.2±2

S.D. = Standard deviation

Table 2. First highest maximum daily contribution 1-hour values averaged over 1-year for all source groups

Discrete Receptor (Group Name)	X (m)	Y(m)	Elevation (ZELEV)	Hill Heights (ZHILL)	CONT JEI OIL [µg/m ³]	CONT L.A.B [µg/m ³]	CONT PETRO [µg/m ³]	CONT PLANT POWER [µg/m ³]	CONT REF [µg/m ³]	CONT SEPAHAN OIL [µg/m ³]	CONT ALL [µg/m ³]	%
North	546925.5	3630332	1640.29	2526	0.23561	0.04735	11.3748	0	165.780	7.25565	184.694	8.3
East	546925.5	3630332	1640.29	2526	3.15125	1.50935	0.36899	0.00825	209.243	9.43619	222.773	10
South	547120.7	3624085	1794.42	2526	0.0001	0.00468	15.8384	0	555.088	1.04225	571.973	25.8
West	544414	3626636	1807.19	2526	0.38789	0.00893	0.02263	0	579.938	10.30572	590.663	26.7
North2	547110.7	3638536	1616.74	1616.7	0.58976	0.27812	17.6655	0	58.6756	5.60856	82.8176	3.7
East 2	558065.1	3626728	1576.95	1576.9	0.84557	1.67287	5.43941	0.04105	70.4750	3.94699	82.4209	3.7
South 2	558065.1	3626728	1576.95	1576.9	0.84557	1.67287	5.43941	0.04105	70.4750	3.94699	86.24	4.1
West 2	534875.8	3626159	1946.52	2505	0.49562	0.86692	3.90764	1.46207	75.8932	3.6684	86.2938	4
Isfahan	560483.6	3619188	1573.05	1573.0	0.58413	2.51605	12.0487	0.00133	53.6184	4.94582	73.7145	3.1
NE	572979	3653737	1669.94	1669.9	0.42388	1.36825	5.82224	0.11335	51.7918	2.5278	62.0473	2.7
SE	575570.7	3600175	1544.06	1717	0.63363	1.62129	20.0115	0	27.8567	5.66221	55.7855	2.5
SW	519244.6	3600866	1997.95	2421	1.00E	0.00092	0.00443	50.6914	0.01218	0.0013	50.7102	2.3
NW	519244.6	3654773	1841.35	2473	0.23313	2.30216	6.73962	5.3768	51.0010	2.54016	68.1928	3.1
TOTAL					7.95	13.2	111.7	58.3	1966.4	60.6	2218.329	
%					0.36	0.59	5	2.6	88.6	2.7	100	
Average					0.62298	1.0167	8.5946	4.48624	151.266	4.6652	170.6407	

Standard concentration =188 µg/m³

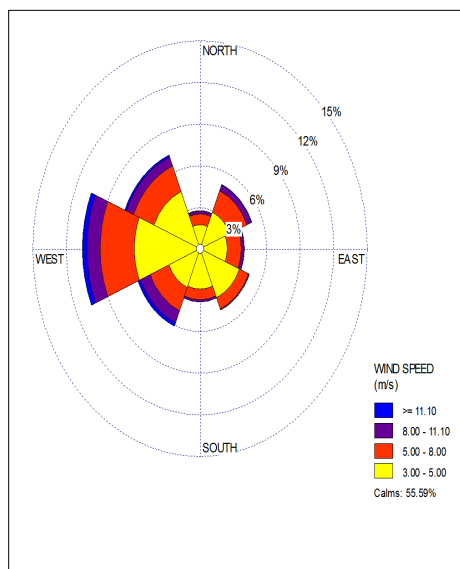


Fig. 1. Wind rose of meteorological data.

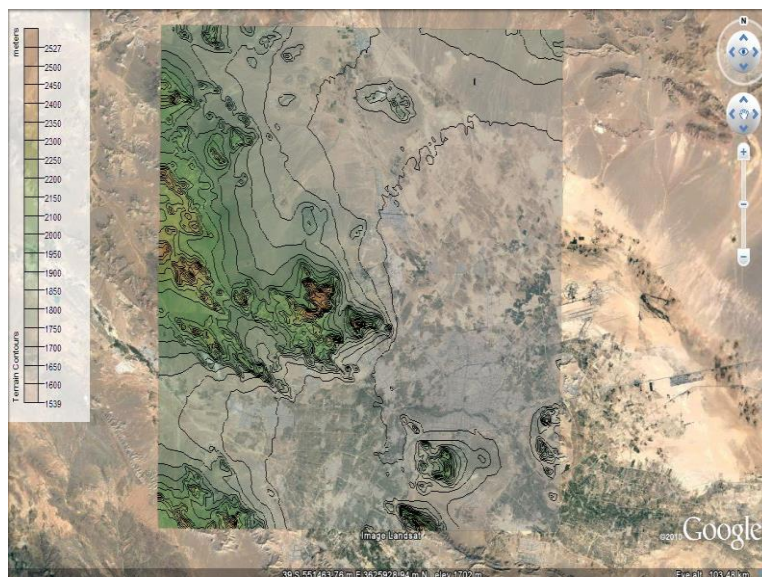


Fig. 2. Terrain contours by the AERMAP.

of 8.3%. NO₂ ground-level concentrations of Eastern receptor, portioned by 10%, were equal to 222.8 µg/m³. Moreover, the south receptors had a 25.8% contribution from total NO₂ ground-level concentration and equalled 571.97 µg/m³. The highest predicted concentration of NO₂ related to the west receptor up to 590.7 µg/m³ and, consequently, the total contribution of this receptor

was 26.7%. All four receptors (distance of 6 km of centre-point) and receptors located 30 kilometres away from centre-point were contributed by 15.5% and 10.7% (respiratory). The more than 99% concentration in SW recaptures (30 km) caused by power plant and 2.5% total contribution of SE receptor divided to 36 and 50% of Petrochemical and refinery respectively.

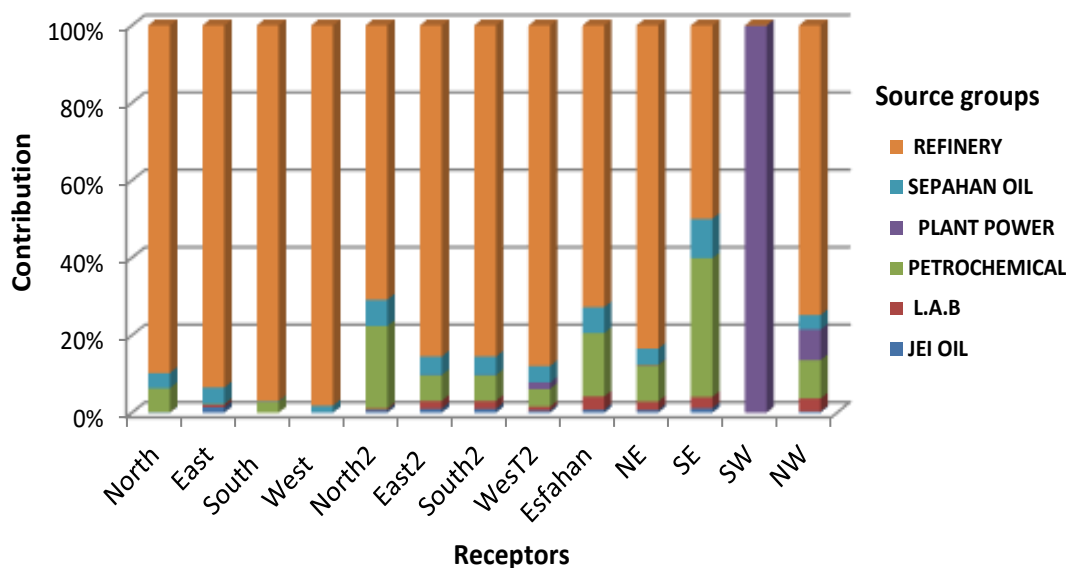


Fig. 3. The contribution of source groups in each receptor.

In relation to the predicted results, the refinery with NO₂ ground-level concentration equalling 1,966.4 µg/m³ had the highest contribution. The other corresponding source for SE receptor concentrations was Petrochemical. About 99% of the predicted concentration was contributed by emission of power plant in SW receptor. Also, the power plant did not have any effect on receptor groups in the four directions at a distance of approximately 3 km (North, South and West), North2 and SE. Total source group concentrations indicated highest to lowest polluted industries by respectively the refinery, Petrochemical, Sepahan Oil, power plant, L.A.B and JEI Oil (Table 2 and Fig. 3).

Temporal-spatial distribution of maximum 1-hour NO₂ from 1 to 50

As can be deduced from Table 3, 50 maximum highest 1-hour NO₂ ground-level concentrations were ranked for all receptors located only in South and West receptors. The highest concentration ranges for South receptor equalled 571.9739 µg/m³ to 268.011µg/m³ accrued in 34 cases. Also, 16 cases of ground level concentrations were corresponded to by the West receptor. Regarding Fig. 4, the temporal distribution maximum ranked concentrations explained that the south receptor throughout the months of a year received maximum 1-hour NO₂, with the highest frequency in March and October. However, this state was not observed in April, June, September, November and December for the West receptor. Overall, the greatest frequency of the highest 1-hour NO₂ occurred in March, October and February respectively.

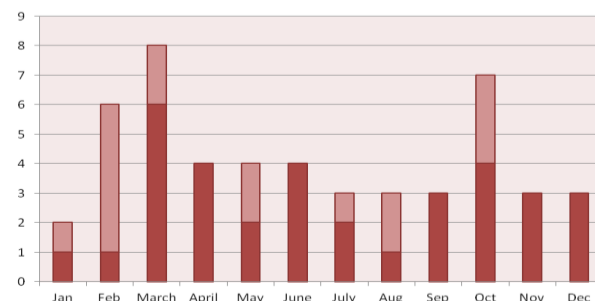


Fig. 4. Temporal-Spatial distribution of 50 cases of highest 1-hour NO₂ in all receptors.

Also, it was emphasized that the West receptor was mainly responsible for 50 highest 1-hour NO₂ this year.

Comparison with EPA standard

Based on comprehensive uniform Cartesian receptors grid extending 30km, the range of highest daily 1-hour values was between 16 and 1080 µg/m³, illustrating some exceeding 1-hour concentrations in 2012. In fact, Fig. 5 represented that the maximum 1-hour NO₂ ground-level concentrations is 5.7 times more than EPA standard 188 (µg/m³) that occurred in the vicinity of the centre-point. With regard to the predicted values, the refinery corresponded to 3.1, 2.95 and 1.1 times exceeding first maximum concentration 1-hour more than the EPA standard in West, South and East receptors respectively (Table 2). According to Table 3, all 50 first maximum 1-hour NO₂ values exceeded 188 µg/m³ in West and South receptors as well with violation of 3 to 1.4 times more than standard. For the Esfahan receptor, total concentration was acceptable, and so it did not have any effect on the resident population in this area.

Table 3. Temporal-spatial distribution of 50 cases of highest 1-hour NO₂ in all receptors

RANK	CONC [µg/m ³]	(YYMMDDHH)	Receptor	RANK	CONC [µg/m ³]	(YYMMDDHH)	Receptor
1	571.9739	12022021	S	26	365.811	12110217	S
2	525.0401	12032924	W	27	363.4813	12110323	S
3	500.4069	12120319	W	28	360.0639	12022204	S
4	481.108	12121617	W	29	358.0189	12060922	S
5	476.112	12102102	W	30	355.9894	12021218	S
6	451.2863	12092620	W	31	355.5398	12102805	W
7	448.8831	12032304	S	32	347.6009	12030322	S
8	447.6141	12032520	W	33	344.8019	12052701	W
9	438.6483	12011102	S	34	344.0498	12031918	W
10	427.841	12100623	S	35	340	12103122	S
11	423.7533	12082701	S	36	336.9673	12091901	S
12	423.4426	12022124	W	37	315.2369	12041005	S
13	411.571	12122007	S	38	307.0502	12040202	S
14	410.1798	12070104	W	39	296.8948	12030521	W
15	406.9648	12081923	S	40	296.4295	12022320	W
16	403.5412	12021720	S	41	295.4345	12042701	S
17	403.2453	12080404	S	42	290.1612	12072202	W
18	398.2622	12050622	S	43	287.7539	12011822	S
19	393.9638	12101222	S	44	283.7877	12062804	S
20	386.7081	12050423	S	45	272.786	12100805	S
21	384.4575	12061322	S	46	270.9455	12040319	S
22	380.131	12031320	W	47	269.5393	12100919	S
23	378.2883	12031322	W	48	268.8437	12061001	S
24	371.6585	12090702	S	49	268.295	12052420	S
25	366.0983	12073101	S	50	268.011	12111607	S

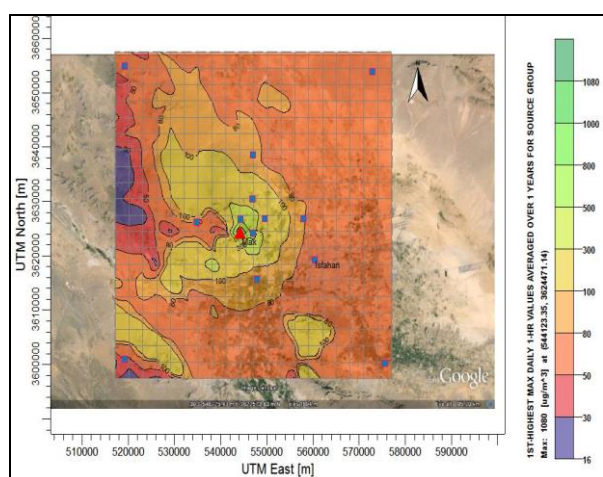


Fig. 5. Simulation of first Maximum 1-hour NO₂ concentration (blue squares represent location of receptors).

DISCUSSION

In this study, the highest maximum daily contribution 1-hour NO₂ associated with the emission rates of petroleum industrial sources was estimated. The contributions of discrete receptors that were located within approximate distances of 3, 6 and 30 km (receptor groups) keep away from a centre-point determined by AERMOD modelling. The results revealed that the cumulative 1-hour NO₂ concentrations (all sources) decline with increasing distance from the sources; however, the largest number of locations with the highest

concentrations was in the West and South receptors as well as more concentration of SE receptor affected by the Petrochemical and the refinery, while power plants had a 99% contribution in the SW receptor. However, the contribution of the refinery in the other receptors was more than 80%. Even though the power plant had an emission rate (43.97 ± 14.6) greater than that of the refinery (11.5 ± 11.3). According to Table 1, it can be the cause of the number of stacks and their heights.

However, the power plant corresponded to about 100% of NO₂ concentration in SW receptor. It is clear that tall stacks exert a great influence on the dispersion of pollutants. Based on some works, the contributions of the Petrochemical industry to annual concentrations of SO₂ and NO₂ were larger than the power plant, even though its emissions were almost four times lower than the emissions of power plant factory. They supported the scientific arguments that the taller stacks of the power plant group enhanced the dilution abilities of emissions from power plants, reducing concentrations of air pollutants at the ground surface level. Furthermore, plumes emitted may travel longer distances before reaching the ground level [28].

Also, the spatial distribution revealed that of 50 maximum highest 1-hour NO₂ ground-level concentrations, only governed by South and West receptors, all of them exceeded the EPA standard. In the interpretation of this result, it can be said that though the main wind direction is from the West,

North West and South West, it is, however, recognized that the highest concentration by prevailing winds do not match necessarily (Fig. 1, 5). In this type of prediction, the maximum highest 1-hour ground level concentrations in different spaces are considerable, and it can-in some cases-be different with the prevailing wind in our area. Furthermore, the results discovered that the higher concentrations can be seen at the place where the terrain begins to elevate at the feet of mountains (Figures 2 and 5). In this regard, referring to the behaviour of the plume, the pollutants can be trapped around the mountains such as that which occurred for south and west receptors, especially when calm winds occurred more than 50% in most cases. In Esfahan city, calm winds included 55% of the total winds [29]. According to the overall most temporal frequency of the highest 1-hour NO₂ occurred in March, October and February, it may be justified through the meteorological parameters and seasonal changes over a year.

Generally, the refinery with 88.6% values NO₂ ground level concentration had the most and Sepahan Oil, JEI Oil and L.A.B industries made the least contributions in the whole area. Concentrations in Esfahan receptor as a dense population point did not have any concern about special NO₂ effects on health.

CONCLUSIONS

The majority of the emissions of the oxides of nitrogen (NO_x) from emission sources are in the form of nitric oxide (NO). The NO gases in the emission plume mix with the atmosphere and react with ozone and other oxidants to oxidize a portion of the NO to NO₂. EPA describes a three-tiered screening approach to calculate NO₂ concentrations, based on dispersion model predictions of NO_x concentrations. Tier 1 approach, which assumes full conversion of NO to NO₂, could be overly conservative in many cases, and may also be prone to reflecting source-oriented impacts from nearby industrial sources located in a single corporate area.

Based on the use of AERMOD modelling, the total source group's concentrations indicated highest to lowest polluted industries by the refinery, Petrochemical, Sepahan Oil, power plant, L.A.B and JEI Oil respectively. Therefore, efforts should be more focused on NO_x emissions advanced control instruments for refinery. The AERMOD model is also a useful tool for identifying emission and contribution sources by discrete receptors.

To conclude, it can be supposed that the ranking of 50 highest 1-hour NO₂ ground-level

concentrations used in this survey showed some cases with maximum concentrations, which can help decision-makers, identify the most polluted receptors. Also, the determination of receptor contributions on different sides is important because in some receptors, corresponding pollutant sources are diverse, such as the SW receptor in this study.

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