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Air Emission Factors and Emission Rates in Asphalt Roofing Manufacturing

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ABSTRACT

This study was focused on measuring air pollutants emission from factories producing the asphalt related products in Delijan Industrial Estate, which is the main center of asphalt related products in Iran. Emissions of four factories were analyzed for air pollutants using a stack analyzer, Testo 360, Testo Co., and Germany. The average concentration of all measured stacks for PM, THC, CO, NOx, SO_2, and CO_2 was 698.0, 195.4, 109.3, and 30.7, 15.1 mg/m^3 and 5.17%, respectively. Estimation of the emission rate for the studied factories with total annual production of 46,500 tons of shingles and rolling asphalt revealed the annual emissions of 32.9, 8.9, 5.0, 1.4, 1.1, and 4,423.4 tons/yr for PM, THC, CO, NOx, SO_2, and CO_2 respectively. It was also shown 435.4, 118.2, 66.7, 189.4, 14.1, and 58,503.1 tons/yr for those pollutants by all factories in Delijan Industrial Estate with annual production of 615,000 tons. The emission factors for the above-mentioned pollutants were 708.1, 192.2, 108.4, 30.8, 22.9, and 95,127 g/ton respectively. Comparing the results with national standard showed that, the PM emission concentration was 2 times greater than Iranian emission standard (250 mg/m^3) and its emission factor exceeded the USEPA emission factor. The rest of pollutants concentrations and emission factors were in the range of both standards. In conclusion, all studied factories need to reduce their particle emission using appropriate air cleaners.

KEYWORDS: *Air pollution, Particle Matter, Hydrocarbon, Asphalt Roofing, Emission Factor*

INTRODUCTION

Asphalt products are extensively used for residential construction and commercial roofing. Six million tons of asphalt per year is produced in roughly 100 manufacturing facilities in USA [1]. Emissions from the Asphalt Roofing Industry (ARI) consist primarily of particulate matter (PM) andvolatile organic compounds (VOC).

Polycyclic Aromatic Hydrocarbons (PAHs), benzene, ethyl benzene, acetaldehyde, formaldehyde, toluene, and xylene are the most frequent hydrocarbons seen in ARI emissions [2]. They emit from asphalt storage tanks, blowing stills, saturators, coater-mixer tanks, and coaters. The PM from these operations is produced primarily from condensing asphalt fume. Wolfgang et al. [3] characterized the organic compounds present in emitting fine particulate of roofing asphalt using GC/MS techniques. They found the compound mass consists of n-alkanes (73%), PAHs and Thia-arenes (S-PAH) contribute nearly 8% of the organic compound mass and account for 0.57% of the total mass of emissions. This is corresponding with that of emitted from catalyst-equipped automobiles (0.5%) but more than heavy-duty diesel trucks with 0.1% [3]; whereas the inorganic compounds were found the most constituent of fine particle in urban air pollution [4]. Study on health damage costs of air pollution has

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been estimated tens of thousands US $ per each unit increase on it [5].

Epidemiological studies of exposure to PAHs have indicated an increased cancer risk in exposed workers and roofers [6-10]. The increasing in mortality among workers who exposed with fumes from heated bitumen in asphalt industry has also seen in numerous researches [11, 12]. Determining the concentration of total emissions arising from handling hot bituminous substances has revealed the amount of about 10 mg/m^3 for processes, which the asphalt is heated 180 °C, and up to 50 mg/m^3 for processes with 250 ^oC heating temperature [13]. Heating the liquid asphalt at about 300 ^oC releases organic compounds trough the exhaust gas, (gaseous and liquid aerosols) that may differ in type and amount from one process to another based on primary asphalt elements and additives. The analysis showed that the asphalt compounds in most asphalt include 79-88% carbon, 7-13% hydrogen, 0-8% sulfur, 2-8% oxygen and 0-3% nitrogen, so, it depends on the type crude oil [14].

The addition of polymers to asphalt in small quantities (at least 0.32%) reduced the asphalt fumes from built up roofing kettles [15]. The polymer forms a steady-state surface layer that reduces the release of fumes from the asphalt. The pilot plan demonstrated statistically significant reductions from 55 to 95% in both opacity and benzene soluble particulate with this technology. This reduction was seen not only in monitoring area near the asphalt kettle but also in limited personal monitoring of workers fume exposure in the workplace [15].

US Environmental Protection Agency (EPA) included the ARI in list of the industrial sources of hazardous air pollutants so it requires reducing its emission. The roofing manufacturing and asphalt processing industry section of the ARI have also been identified in EPA's emission inventory as sources of Polycyclic Organic Matter (POM) [16].The processes which contribute to emissions from asphalt roofing manufacturing include: the roofing manufacturing line; the delivery, transfer, and storage of asphalt and mineral products used in the manufacture of roofing products; and the blowing of asphalt [17].

The ARI emissions are estimated by its Emission Factors (EF) for air dispersion modelling. EFs have long been used as a cost-effective means to develop area–wide emission inventories. Emission inventories are fundamental tools for air quality management. They are used for identifying major contributors of atmospheric pollutant, developing emission control strategies, determining applicability of permitting programs, and other related applications [18]. An EF is a tool that is used to estimate air pollutant emissions to the atmosphere. It relates the quantity of pollutants released from a source to some activity associated with those emissions [19]. Hence, calculating EF for asphalt roofing manufacturers could be of high important.

Field of Study: Extensive using of roofing isolators for the coverage of the building roofs lead to establish more factories to produce asphalt-roofing rolls, so, 35 major factories are producing asphalt-roofing products in Delijan Industrial Estate (DIE). DIE as the main place of ARI in Iran with the area of 438 hectares has located in 5 km far from the southeast of Delijan City, a city with about 100,000 population in South-East of Tehran [20]. Nowadays, DIE is going to become a heath concern for people and health authorities due to its emission of large amount of PM and gaseous pollutants. Although settlement of new producers of ARI in the DIE and in the areas near the Delijan City has prevented by Health Council of Markazi Province, which Delijan city belong it, since April 2005 and they also set more limitations for production activity of existing factories as well, but there is still no useful data about type and exact amount of air emissions [21].

Process Description*:* The first step of asphalt roofing manufacturing is asphalt blowing. It involves the oxidation of hot asphalt flux, which is achieved by the bubbling air of the blowing still. Air is forced through holes in the sparger into a tank of hot asphalt flux. The result is an exothermic oxidation reaction, which raises the softening temperature of the asphalt, as well as modifying other characteristics. The process is highly temperature dependent, as the rate of oxidation increases rapidly with increases in temperature. Since the reaction is exothermic, the temperature rises as blowing proceeds. Temperatures must be kept safely below the flash point of the asphalt. The temperature is therefore kept at an optimum level of 260^oC during blowing by spraying water onto the asphalt surface.

Inorganic salts such as ferric chloride ($FeCl_3$) may be used as catalysts to achieve the desired properties and/or to increase the rate of reaction, thus decreasing the blowing time. Blowing times may vary in duration from 30 minutes to 12 hours, depending on the desired characteristics of the asphalt (softening point, penetration rate). Process parameters influencing emissions include the blowing temperature, air rate, design/configuration of the still, and the type of product desired.

To prepare desire mixture of asphalt for covering the tissue, oxidized asphalt is mixed with recovered (second hand) Polypropylene granule, Talk powder and $CaCO_3$ in the decoction drum (Bonza) and is heated up to about 300 ^{o}C until the suitable asphalt is obtained.

For asphalt-saturated felt, a typical manufacturing line consists of a tissue and polyester feed roll, a dry lopper section, a saturator spray section (may not be used), a saturator dipping section, steam-heated drying-in drums, a wet lopper, water cooled rollers, a finish floating lopper, and a roll winder.

For asphalt shingles, smooth rolls and mineral-surfaced rolls, the manufacturing line is similar to the felt line, with the addition of a filled asphalt coater, a granule applicator, a press section, water cooled rollers, a finish floating lopper, and either a roll winder or a shingle cutter and stacker. After preparing, the primary material turns over to the storage mixer. Primary materials go to the hot asphalt basin and then tissue and polyester go through with 1 meter width into the basin, filled with asphalt and then enter to the cold-water basin for chilling. Then, one side covered with nylon to avoided stickiness of layers each other during rolling and the other side covered by grout. Some factories use aluminum foil instead of grout, therefore roof's surface is shiny.

This study was focused on measuring air pollutants emission from factories producing the asphalt related products in Delijan Industrial Estate, which is the main center of asphalt related products in Iran.

MATERIALS AND METHODS

Sampling methods: This work was focused on measuring air pollutants emission from factories were producing the asphalt roofing shingles in DIE in 2008. Four factories were randomly selected among 35 active manufacturers situated in DIE namely Parsian Dej (PD), Sadaf Gostar (SG), Nemoone (N), and Yekta Bam (YB). They all work in one working shift in a day from 8:00 to 16:00, 6 days in a week and just 8 months in a year. Their activities start from mid August until Mid April. Each one had 2 exhaust stacks, one for decoction drum and another for their mixer, except the YB that had just one exhaust stack, so that all emissions conducted to that. It had a very traditional process system.

Air pollutants include: Particle Matter (PM), total Hydrocarbon (THC), Carbon Monoxide (CO), Nitrogen Oxide (NOx), Sulfur Dioxide (SO_2), and Carbon Dioxide (CO_2) were measured using a Testo 360 stack analyzer, which was calibrated, by manufacturing factory few months ago. A sampling hole was prepared in final straight section of each stack at more than 6D distances far from any fitting and before the end (EPA method 1). To minimize the effect of work fluctuations on results the measurements were repeated 2 times in a working shift at morning and afternoon with about 4 hours interval between them. The temperature and velocity of flue gas as well as the stack diameter were also measured and the other essential data about shift duration, holidays and production rate was gathered.

Method of EF calculation: EFs are usually expressed as the weight of pollutant emitted divided by a unit weight, volume, distance, or the activity emitting the pollutant (e.g., pounds of particulate matter emitted per ton of coal burned) [17]. EPA published the estimated EFs for productive industries as the weight of pollutants, kg or g, per tons of products [17-22]. Therefore, in order to calculate EFs, the emission estimate and production data by plant or for the sector are required. There are several available methods for estimating emissions from industries. Generally, methods that use site-specific data, such concentrations in the exhaust gas and the exhaust gas volumetric flow rate were used to determine the Efs [19].

Total exhaust air volume of a shift calculated by multiplying the stack air velocity in the stack area and shift duration. It was also normalized for temperature and local barometric pressure. Then, the emission rate was calculated by multiplying the normalized exhaust air in the pollutant concentration.

RESULTS

The essential characteristics of the stacks and their flows needed for emission calculations in the studied factories are presented in Table 1.

Narrowness the stacks and inefficient ventilation caused going out and dispersion a part of pollution in the ways other than stacks, so it was not included in this calculation.

Table 1. Stacks, height (m) and its flow rate characteristics in asphalt roofing factories in DIE (2008)

	Factory		**PM**	**THC**	**CO**	**NO_x**	**SO_2**	**$\%CO_2$**
1	Parsian Dej	Bonza	780	195	92	39	19	5
		Mixer	732	143	65	31	10	6.5
2	Sadafgostar	Bonza	745	160	112	24	26	7
		Mixer	626	180	120	29	24	3.8
3	Nemune	Bonza	631	216	180	35	12	4
		Mixer	639	232	104	28	11	4.1
4	Yektabam		733	242	92	29	4	5.8
	Mean		698	195.4	109.3	30.7	15.1	5.17

Table 2 shows the average concentration of the measured pollutants include PM, THC, CO, NOx, SO_2, and CO_2 in emission stacks of 4 sample factories in DIE. PD bonza with the 780 mg/m^3 and SG mixer with 626 mg/m^3 average concentration of PM had the highest and the lowest emission among the factories respectively. This has seen for THC emission in YB and PD mixer with 242 mg/m^3 and 143 mg/m^3 respectively.

Table 2. Concentration of air pollutants (mg/m^3) in the emission of asphalt roofing factories in DIE (2008)

Factory		**Stack (m)**	**exhaust velocity (m/s)**	**Flow rate (m^3/min)**	**Normalized flow rate (m^3/min)**
Parsian Dej	Bonza	0.35	15.7	90.43	73.3
	Mixer	0.35	15.7	90.43	73.3
Sadafgostar	Bonza	0.50	10.5	123.6	83.3
	Mixer	0.50	11	129.5	86.6
Nemune	Bonza	0.45	9.7	92.5	54.6
	Mixer	0.45	9.7	92.5	54.6
Yektabam		0.50	9.5	111.9	63.5

Total environmental air emission of 4 studied factories were calculated using average concentrations of pollutants and stacks flow characteristics. As it is shown in Table 3, after CO_2 emission with 4,423.4 tons/year which has a globally importance pollutant due to its greenhouse effect, PM with emission of 32.9 tons/year was the major pollutant dispersed over the around environment. The THC, CO, NOx, and SO_2 emission with 8.939, 5.042, 1.432, and 1.066 tons/year were in the later degrees regarding weight of total emission.

Table 3. Emission rate ($E_{Tons/Year}$) of air pollutants from different process of asphalt roofing factories in DIE (2008)

Factory (Process)		E_{PM}	E_{THC}	E_{CO}	E_{NOx}	E_{SO2}	E_{CO2}
Parsian Dej	Bonza	5.489	1.372	0.644	0.274	0.134	631.9
	Mixer	5.150	1.006	0.922	0.218	0.07	821.4
	Total	10.639	2.378	1.566	0.492	0.207	1453.3
Sadafgostar	Bonza	5.958	1.279	0.9	0.192	0.286	1,005.3
	Mixer	5.204	1.496	0.998	0.241	0.306	567.4
	Total	11.162	2.775	1.898	0.433	0.592	1,572.7
Nemune	Bonza	3.307	1.094	0.472	0.183	0.063	376.5
	Mixer	3.349	1.216	0.545	0.147	0.058	386
	Total	6.656	2.31	1.017	0.33	0.121	762.5
Yektabam (Total)		4.468	1.476	0.561	0.177	0.146	635
Total of 4 Factories		32.925	8.939	5.042	1.432	1.066	4,423.4
DIE (35 Factories, 615,000 tons of production)		435.420	118.202	66.666	18.942	14.099	58,503.1

Table 4 shows the EFs of air pollutant for 4 studied factories. The results here have the same order of other tables. CO_2 had the highest amount and then PM was the second one and so on. The average EF for PM was 3.5 times higher than that of THC and more than the others too.

Table 4. Emission Factors (g/ton product) of air pollutants from different process of asphalt roofing factories in DIE 2008

Factory		EF_{PM}	EF_{THC}	EF_{CO}	EF_{Nox}	EF_{So2}	EF_{Co2}
Parsian Dej	Bonza	407	102	47.7	20.3	10.0	46,807
	Mixer	381	74.5	68.3	16.1	5.2	60,844
	Total	788	176.5	116	36.4	15.2	107,651
Sadafgostar	Bonza	441	94.7	66.7	14.2	21.2	74,467
	Mixer	385	11.1	73.9	17.9	22.7	4,203
	Total	826	105.8	140.6	32.1	43.9	78,670
Nemune	Bonza	368	121.1	52.4	20.3	7.0	41,833
	Mixer	372	135.1	60.6	16.3	6.4	42,889
	Total	740	256.2	113	36.6	13.4	84,722
Yektabam (Total)		426	140.6	53.4	16.9	13.9	60,476
Average		708.1	192.2	108.4	30.8	22.9	95,127

Table 5. Emission Factors from asphalt roofing (17)

Pollutant	EF (g/ton product)	95% Confidence Interval	
		Lower	Upper
PM	600	200	1600
NMVOCs	5	4	160
CO	18	3	30
NOx	38	NA	NA
SO_2	28	NA	NA

Comparing the calculated EFs for DIE with this table showed that in all cases unless NOx the amounts were exceeded than these levels. Noticing the 95% confidence interval for EFs in the table revealed although the EF for PM was higher than that of EPA but still was in the range while the others such as THC and CO not only exceeded the EFs but also were not in the 95% confidence interval. The only normal case for calculated EFs was the NOx emission that was under the both limits. As it mentioned in the beginning of this section all of the emissions in the studied factories as well as in the others in DIE were not conducted in to the stacks, so the results were less than the real amounts.

DISCUSSION

Minimum CO emission with 65 mg/m^3 was seen in PD mixer and its maximum amount with 180 mg/m^3 was seen in N bonza. The average concentrations of NOx and SO_2 in the factories were rather low, so the ranges of concentration were 24 – 39 mg/m^3 and 4 – 26 mg/m^{3}for them respectively. Finally, the highest average emission of CO_2 with the amount of 7% was seen in SG bonza section and the lowest one with 3.8% was seen in the same factory but in the other section, SG mixer. This variation in CO_2 emission while the process system is similar may due to weakness of system maintenance and dilution of exhaust emission with workplace air. The total average concentration of PM, THC, CO, NOx, SO_2, and CO_2 emission in 4 factories which used for emission calculation were 698.0, 195.4, 109.3, and 30.7, 15.1 mg /m^3 and 5.17% respectively

Total emissions of pollutants were estimated for all 35 factories of DIE using average EFs calculated for 4 factories and their total annually production. While CO_2 with 58,503.1 tons/year was the most emission gas from the DIE, the PM emission with 435.4 tons/year was the second major emission and 4 other gaseous pollutant with sum of 217.9 nearly correspond with half of the PM emission. This was the real concern of the environmental health authorities. The PM emission may contains hazardous materials such as aromatic hydrocarbons and settled on the around environment, so, the area will be polluted for long time. However, the gaseous pollutants dilute in the air and could travel more distance rather than PM so their effects appear only in short term after emission.

Iran Department of Environment (IDE) has not established separate emission standards for air pollutants emission by asphalt roofing factories but there were general standards for those factories, which had not separate standards, and the ARI must pass those. They required the factories to maintain their air pollutants emissions include: PM, CO, and SO_2 less than 250 mg/m^3, 350 ppm, and 800 ppm respectively [23]. These standards had two major shortages; firstly they mentioned the standards as maximum concentration of air pollutants in the stacks of factories so the factories could easily pass the standards not by reducing their pollution but by blowing excess air in to the stacks or conduct a part of their pollution in other ways such as general ventilation via windows or wall axial fans. Secondly, there were not detailed standards for elements of emissions such as benzene, toluene, xylene, and etc.

However comparing the averages of pollutants emissions with IDE general standards has revealed that the PM concentration was exceeded the standard in all studied factories but the CO and SO_2 were not exceeded the proposed levels. Although some researchers had estimated a very high level of EFs using engineering calculations for air pollutants (540 g/ton for VOCs) but using data from more than 100 asphalt related manufacturers [24]; EPA estimated EFs for shingle (rolling asphalt) producing in asphalt sectors as table 5 [22].

The important issue other than the amounts of pollutants is their constituents. Unfortunately due to the limitation in sampling equipment in this work, the analysis of PM and THC to their constituents were not possible. Both of them may consist of hazardous compounds such as benzene, ethyl benzene, formaldehyde, acetaldehyde, quinone, toluene, xylene and naphthalene, which many of them have carcinogenic effects.

As the results showed, the aggregation of these factories in one industrial estate may cause to emit a large amount of such toxic pollutants which could face people's health on dangerous, so the policy of establishment for ARI must be noted and the minimum distance of 10 km from the populated areas for these estates should be specified.

Another accepted way to minimize the adverse effects is using control devices. A number of add on technologies exist that are aimed at reducing the emissions of specific pollutants. Electrostatic Precipitator and High Energy Air Filter are capable of more than 97% abatement of particle in ARI emission [22]. Process controls can also be used including vertical rather than horizontal stills, asphalts that inherently produce lower emissions, higher flash point asphalts, and lower asphalt blowing temperatures [25].

A common method for controlling emissions from the saturator, including the wet looper, is to enclose them completely and vent the enclosure to a control device. The coater may be partially enclosed, normally with a canopy-type hood that is vented to a control device. Full enclosure is not always practical due to operating constraints. Fugitive emissions from the saturator or coater may pass through roof vents and other building openings if not captured by enclosures or hoods. Control devices for saturator/coater emissions include low-voltage electrostatic precipitators, high-energy air filters, coalescing filters (mist eliminators), afterburners (thermal oxidation), fabric filters, and wet scrubbers.

Particulate matter associated with mineral handling and storage operations could capture by enclosures, hoods, or pickup pipes and controlled by fabric filtration with removal efficiencies of approximately 99 percent. Other control devices that may be used with mineral handling and storage operations are wet scrubbers and cyclones.

In the industry, closed silos and bins should be used for mineral storage, so open storage piles are not an emission source. To protect the minerals from moisture pickup, all conveyors that are outside the buildings must cover or enclosed. Fugitive mineral emissions may occur at unloading points depending on the type of equipment used and the mineral handled. The discharge from the conveyor to the silos and bins could normally controlled by a fabric filter.

CONCLUSION

The studied factories emitted a large amount of air toxics, which can lead to adverse effects in environment and could endanger the human life in populated areas near them. Minimum 10 km distance from the residential areas, inherently cleaner production, prevention of establishment of more than 10 factories (or 200,000 tons of production capacity) in one area, and install suitable air cleaners in ARI could be useful for reducing the adverse effects of them.

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