

# Air Pollution from a Large Steel Factory: Polycyclic Aromatic Hydrocarbon Emissions from Coke-Oven Batteries

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## ABSTRACT

A systematic investigation of solid and gaseous atmospheric emissions from some coke-oven batteries of one of Europe's largest integrated steel factory (Taranto, Italy) has been carried out. In air monitoring samples, polycyclic aromatic hydrocarbons (PAHs) were consistently detected at concentrations largely exceeding threshold limit values. By means of PAHs speciation profile and benzo(a)pyrene (BaP) equivalent dispersion modeling from diffuse sources, the study indicated that serious health risks exist not only in working areas, but also in a densely populated residential district near the factory.

## INTRODUCTION

The steel factory in Taranto, Italy is one of the largest in Europe and represents a very heavy pollution source.<sup>1</sup> Although poor detailed information exists about the results of air quality monitoring in the surrounding area, this plant is commonly blamed for being a major contributor to severe air pollution facing the local population. Epidemiologic data show that this area experiences a 22% death rate of cancer in excess of the regional average, with >40% of cases because of lung disease.<sup>2</sup> Taranto was listed among "Italy's 15 areas at high risk of environmental crisis" according to the Italian laws No. 349/86 and 305/89.

To collect more information and experimental evidence, a 7-month investigation on major toxic pollutants, namely, total suspended particles (TSP), fine particulate matter with aerodynamic diameter  $\leq 3.5 \mu\text{m}$  ( $\text{PM}_{3.5}$ ), and Benzene emitted from the factory, particularly from the old batteries no. 3-4 and 5-6 of the coke-oven plants, has been carried out. In previous articles, it has been shown how the aforementioned toxic pollutants from coke-oven emissions are responsible for health risks among workers.<sup>3,4</sup> In this paper, specific attention is dedicated to the

emission of polycyclic aromatic hydrocarbons (PAHs), because general concern of these persistent organic pollutants is increasing worldwide because of their genotoxic, mutagenic, and carcinogenic health effects in humans.<sup>5</sup>

## PAH Pollution

Coke-oven batteries, where coal is pyrolysed at  $\sim 1100^\circ\text{C}$  to produce coke and pyrolytic gas, both necessary for other sections of the process, are a major source of toxic emissions in the steelmaking integrated plants (see Figure 1, taken from ref 6). These emissions, predominantly diffuse, originate from oven leakages, as well as from cyclic operations of coal loading and coke unloading.

PAHs are a large group of organic compounds made by fused benzene rings with linear, angular, or clustered arrangements resulting from incomplete combustion of organic matter. Airborne PAHs with less than three aromatic rings (mol wt 128-178) are gaseous, whereas PAHs with five or more rings (mol wt  $>228$ ) are merely bound to airborne particulates. In particular, 80-100% of these latter PAHs are reported to be associated with fine particulate matter with aerodynamic diameter  $\leq 2.5 \mu\text{m}$  ( $\text{PM}_{2.5}$ ).<sup>7,8</sup>

Major routes of PAHs exposure are through inhalation and cutaneous absorption, which could be very significant and reach 50% of the total body dose for coke-oven workers.<sup>9</sup> Near coke-oven batteries, the levels of benzo(a)pyrene (BaP) may range from 100 to 200  $\mu\text{g}/\text{m}^3$  on the machinery and discharge side of a battery roof and  $\sim 400 \mu\text{g}/\text{m}^3$  at the battery top.<sup>10</sup> These figures should be compared with much lower existing BaP limits for occupational exposure, such as the 5  $\mu\text{g}/\text{m}^3$  German Technische Richtkonzentration (TRK) proposed by the Federal Institute for Occupational Safety and Health, reportedly matched by modern coking plants fully equipped with best available techniques (BAT).<sup>11,12</sup>

In living ambient, on the other hand, the annual mean level of BaP observed in major European cities is in the range of 0.5-3.0  $\text{ng}/\text{m}^3$ ,<sup>13,14</sup> whereas the World Health Organization (WHO) has proposed a guideline as low as 0.01  $\text{ng}/\text{m}^3$ .<sup>15</sup> In Italy, the limit for BaP in living ambient is set at 1  $\text{ng}/\text{m}^3$  by the Ministerial Decree on November 25, 1994.

## IMPLICATIONS

This study demonstrates that air quality in a residential area surrounding Europe's largest steel-making factory exceeds PAHs threshold limits in force, as well as BaP World Health Organization guidelines in living ambient, even after full equipment of existing installations with the best available techniques in the sector.

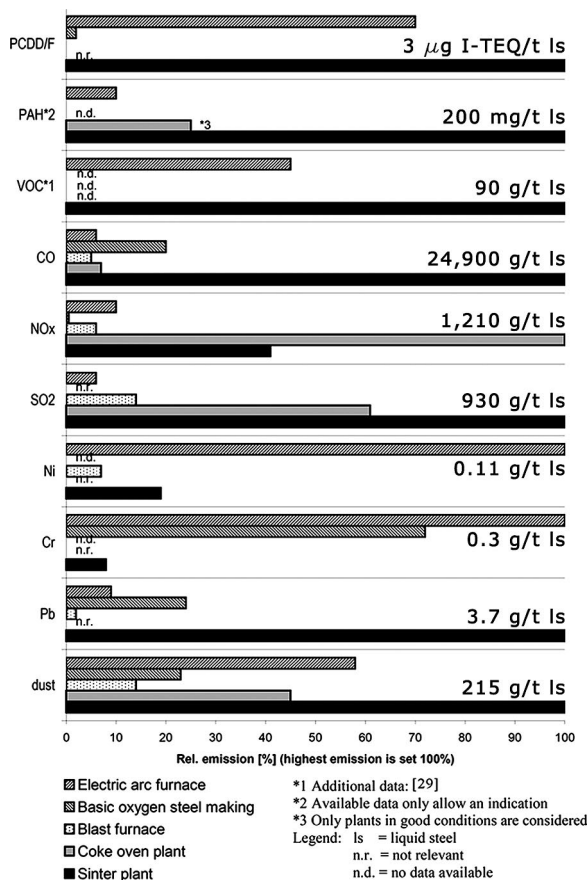


Figure 1. Relative atmospheric emission of selected pollutants from steel making.<sup>6</sup>

### Coke-Oven Plants

Coking plants at the Taranto factory consist of 12 batteries. The present investigation concerned four batteries (No. 3–4 and 5–6, see Figure 2), each made by 45 ovens,

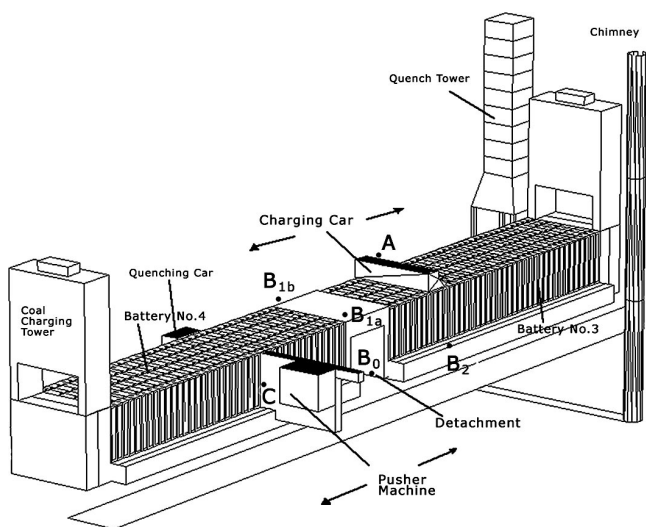


Figure 2. Sketch of coke-oven batteries No. 3–4 (or 5–6) and sampling stations (A and C mobile sampling stations, placed on the charging and pusher machine, respectively; B<sub>0</sub>, B<sub>1</sub>, and B<sub>2</sub> fixed sampling stations placed alternatively at the batteries detachment or at the top of battery or on the pusher machine side).

built between 1964 and 1970, never submitted to revamping since then, with (partial) refreshing carried out on batteries No. 5–6 at the end of the 1980s. Such a situation is representative of a large number of similar cokerries working around the world.<sup>6,16</sup>

The coking process in each oven occurs batch-wise according to the following cycle (see Figure 2): (1) the charging car, previously loaded with coal at the charging tower, feeds the oven with coal by the charging holes at the battery upper desk (~15 min); (2) the holes are closed with lids hermetically sealed, and the pyrolytic distillation of coal at ~100 °C within the oven occurs (~21 hr); (3) the pusher machine evacuates the oven by transferring the coke remained therein into the quenching car at the intermediate desk (~15 min); (4) the quenching car reaches the quenching tower where the hot coke is cooled at ambient temperature with fresh water (~15 min). Once over (~21.5 hr), the cycle starts again at that same oven. Batch operation of all ovens occurs through a specific sequence (i.e., oven No. 93, 98, 103, 108 etc.) to maintain thermal asset of the batteries and to ensure continuous plant operation.

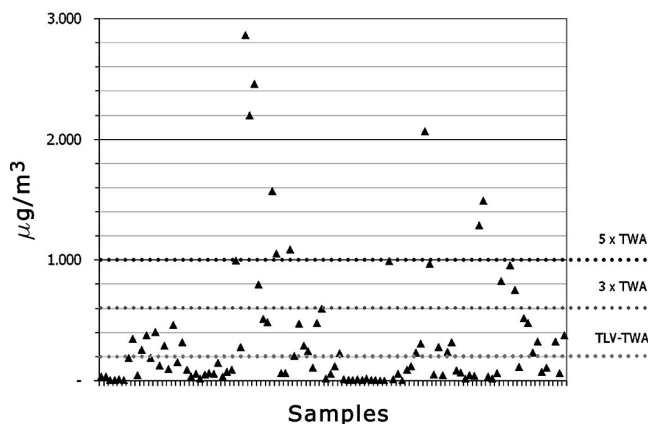
On average, each oven is loaded with 20.5 t (30 m<sup>3</sup>) of pit coal and produces 16.1 t of coke per cycle. A total of 36,370 loading and unloading sessions per year (~100 daily) occur on batteries No. 3–4 treating 746,585 t of pit-coal and producing 585,076 t of coke and 264,749 kNm<sup>3</sup> of pyrolytic gas (similar performances on batteries No. 5–6). Specific productivity averages 0.78 t of coke and 355 Nm<sup>3</sup> of gas per metric ton of coal, a performance quite acceptable for this type of plant. In addition to unavoidable gas emissions during oven charging and discharging, after ~40 yr of continuous operation, batteries No. 3–4 and 5–6 produce diffuse emissions of toxic gaseous and particulate compounds from numberless wall breaks, as repeatedly evidenced during this study.

### Monitoring Strategy and Program

Environmental monitoring at batteries No. 3–4 and 5–6 discussed in this paper was carried out through fixed and semimobile sampling stations. In particular, three air sampling stations were used in each session (see Figure 2), two (mobile) on the charging car (A) and the pusher machine (C), respectively, one (fixed) alternatively placed at the detachment area (B<sub>0</sub>), at the top of the battery (coke side, B<sub>1a</sub> or pusher machine side, B<sub>1b</sub>), or at the pusher machine (B<sub>2</sub>), monitoring alternatively batteries No. 3–4 or 5–6. Because of the continuous movement of cars along the batteries, samples collected therein were considered representative of average air characteristics around loading (upper desk) and unloading (intermediate desk) working areas, respectively, whereas samples from the fixed station were assumed representative of overall diffuse emissions at the three desk levels.

The monitoring program was carried out through six distinct sessions. Five sessions were held November 2–3, 14–15, and 21–22; and December 5–6 and 12–13, 2001, respectively, involving both coke-oven batteries No. 3–4 and 5–6, whereas the sixth session (May 15–16, 2002) involved batteries No. 3–4 only.

Each monitoring session lasted ~12 hr (from 9:00 a.m. to 8:00 p.m.) except for the last session, extended for



**Figure 3.** PAH total concentration (particulate + vapor) measured at batteries No. 3–4 and 5–6.

~40 hr (from 9:00 a.m. of May 15 to midnight of May 16, 2002) to monitor one complete coking cycle. According to sampling recommendations, each sampling lasted ~2 hr continuously. Six samples were taken on the average at each station during a typical 12-hr session.

#### EXPERIMENTAL WORK

Air samples were collected by a sampling train made by a cassette filter holder (47/12 mm) equipped with a Teflon filter (2 µm) followed by a trap filled with 100 mg (front) and 50 mg (back) of adsorbent resin (Amberlite XAD-2) connected to the filter by a short polyvinyl chloride tube. Sampling pumps (Zambelli mod. ZB2 - PLUS 6000 and Tecora mod. Bravo R/PRG) equipped with dry volume meter, flow rate meter, rotary pump with linear flow-forced circulation, and cooling circuit with heat exchange serpentine were set at proper flow rate, that is, 8–10 L/min for particulate and PAHs. All of the samples were protected from sunlight and frozen immediately.

Dust collected on filters was weighed with an electrical balance ( $\pm 0.01$  mg), then sent to the laboratory together with the sorbent trap to determine separately particle-bound and gaseous PAH concentration. To this aim, each sample collected was extracted with 5 mL of acetonitrile in an ultrasonic bath for 30 to 60 min. PAHs were analyzed by high-performance liquid chromatography-UV according to National Institute for Occupational

Safety and Health (NIOSH) method 5506, modified to determine PAHs through  $PM_{3.5}$  filters.<sup>17,18</sup>

Polyaromatic hydrocarbon recovery efficiency, determined by processing a standard spiked filter with the same experimental procedure used for samples, varied between 0.74 and 1.10 (average of 0.85) with 19% mean relative standard deviation and 20% analysis uncertainty. Calibration and quality controls, as well as measurements and calculations, including peak identification and quantification, occurred according to NIOSH method 5506.<sup>18</sup>

## RESULTS AND DISCUSSION

### Occupational Exposure to PAHs

According to Italian Ministerial Decree, August 20, 1999, total PAH concentration (particle + vapor) detected around coking plants was matched with air standard in workplace addressed by the American Conference of Governmental Industrial Hygienists (ACGIH), which recommends a 200 µg/m<sup>3</sup> threshold limit value (TLV)-time weighted average (TWA) occupational exposure to coal tar products for an 8-hr workday within a 40-hr workweek. Air concentration may reach 600 µg/m<sup>3</sup> (i.e., three times the TLV-TWA) for 30 min cumulatively in a workday, but under no circumstances it may exceed 1000 µg/m<sup>3</sup> (i.e., five times the TLV-TWA).<sup>19</sup>

Figure 3 reports PAH experimental concentration determined during the first five monitoring sessions (104 samples). From these data, it results that the TLV-TWA was exceeded in 46% of cases, in 18% and 11% of cases more than three and less than five times, respectively. In ~27% of cases, the 137.45 µg/m<sup>3</sup> geometric mean was exceeded by 3.13 times, indicating poor process stability according to the ACGIH.

Table 1 specifies where the excess PAHs concentrations occurred, confirming poor coal distillation in the ovens (mobile sampling stations) and uncontrolled leaks from oven brick walls during the entire cycle. These data confirmed heavy occupational exposure at all of the working areas around these batteries (upper, intermediate, and lower desks).<sup>3,4</sup>

### PAH Speciation Profile

To ascertain the total PAH speciation profile, the occurrence and abundance of single PAHs was determined for each of the 91 samples collected in the sixth session. The

**Table 1.** Summary of PAHs average concentration determined at batteries No. 3–4 and 5–6 during the first five sessions.

Session	Location of Sampling Station	Type	No. of Samples	Total PAHs (g/m <sup>3</sup> )	
A	Charging car	Mobile	19	<b>358</b>	<b>513</b>
B <sub>1a</sub>	Top—pusher machine side	Fixed	16	<b>628</b>	95
B <sub>1b</sub>	Top—coke side	Fixed	16	<b>426</b>	—
C	Pusher machine	Mobile	19	<b>274</b>	171
B <sub>2</sub>	Pusher machine side	Fixed	16	<b>329</b>	<b>1389</b>
B <sub>0</sub>	Batteries detachment	Fixed	18	126	<b>442</b>
		Average		<b>357</b>	<b>522</b>
		Standard Deviation		<b>611</b>	<b>622</b>

Notes: Bold values exceed TLV-TWA.

**Table 2.** Average speciation profile and carcinogenicity factor of PAHs found in emissions from batteries No. 3–4.

Compound	Symbol	CAS Number	Abundance (%)	Occurrence (%)	Carcinogenicity factor
Naphthalene	Nap	91-20-3	28.70	89	—
Pyrene	Pyr	129-00-0	15.58	69	0.021
Benzo(b)fluoranthene <sup>c</sup>	BbF	205-99-2	10.46	65	0.099
Indeno(1,2,3-cd)pyrene <sup>c</sup>	IND	193-39-5	8.36	69	0.091
BaP <sup>a</sup>	BaP	50-32-8	6.96	74	1 <sup>b</sup>
Acenaphthylene	AcPy	208-96-8	6.77	71	—
Acenaphthene	Acp	83-32-9	4.82	60	—
Dibenz(a,h)anthracene <sup>a</sup>	DBA	53-70-3	3.70	47	1.8
Fluorene	Flu	86-73-7	2.98	37	—
Fluoranthene	FL	206-44-0	2.51	24	0.088
Anthracene	Ant	120-12-7	2.22	37	0.0026
Phenanthrene	PA	85-01-8	1.89	33	0.0044
Chrysene	CHR	218-01-9	1.35	10	0.13
Benzo(e)pyrene	Bep	192-97-2	1.26	12	0.002
Benzo(a)anthracene <sup>a</sup>	BaA	56-55-3	1.09	29	0.067
Benzo(ghi)perylene	BghiP	191-24-2	0.82	1	0.017
Benzo(k)fluoranthene <sup>c</sup>	BkF	207-08-9	0.52	12	0.061

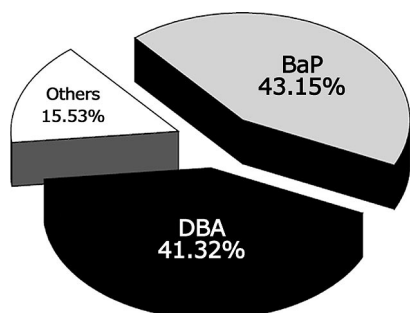
<sup>a</sup>Class 2A (probable human carcinogen) by the International Agency for Research on Cancer. <sup>b</sup>Equivalent concentration to BaP. <sup>c</sup>Class 2B (possible human carcinogen) by the International Agency for Research on Cancer.

results are reported in Table 2 for each PAH found together with its relative carcinogenicity factor to BaP, which allows for converting total PAHs into a BaP equivalent concentration.<sup>14</sup>

Several toxic PAHs were detected, namely: BaP, benzo(a)anthracene, and dibenz(a,h)anthracene, listed among class 2A (“probable human carcinogens”), as well as benzo(b)fluoranthene, benzo(k)fluoranthene, and indeno(1,2,3-cd)pyrene, listed among class 2B (“possible human carcinogens”) by the International Agency for Research on Cancer.<sup>5</sup> Reference to most frequently found PAHs, called “indicatory” PAHs, has long been used for identifying pollution sources.<sup>20,21</sup>

Accounting for their predominant concentration, the following six PAHs, contributing to ~77% of total PAHs detected, were considered “indicatory” in the situation investigated: Naphthalene (Nap), Pyrene (Pyr), Benzo(b)fluoranthene (BbF), Indeno(1,2,3cd)pyrene (IND), BaP, and Acenaphthylene (AcPy; see Table 2).

BaP and Dibenz(a,h)anthracene (DBA) were responsible for ~85% of the total carcinogenicity content of the emissions investigated (see Figure 4). The BaP equivalent concentration of total PAHs averaged 6.82, 9.30, and 10.65  $\mu\text{g}/\text{m}^3$  at pusher machine, detachment batteries,

**Figure 4.** Carcinogenicity contribution of PAH mixture at batteries No. 3–4.

and charging car, respectively, with an overall average value of 9.81  $\mu\text{g}/\text{m}^3$ .

These figures always exceeded the 5  $\mu\text{g}/\text{m}^3$  TRK limit, even when the total PAH concentration was below TLV–TWA. This confirms that the total PAH concentration may not be an appropriate indicator for the effective carcinogenicity potential of the emission investigated, as stated by other investigators who suggested using BaP to that aim.<sup>14</sup>

### Emission Factors

Emissions from industrial plants may fall into two broad categories: stack (e.g., via a chimney) and fugitive emissions (e.g., via coke-oven seals and wall breaks), with the latter much more difficult to control. Emission factors are a useful guide for estimating air pollution and setting environmental regulations. By reference to the 96/61/CE European Directive for Integrated Pollution Prevention and Control, a systematic investigation in the iron and steel sector among major European plants permitted to evaluate the range of emission factors (EF; grams of pollutant per metric ton of crude steel) for the main components of coke-oven emission with or without BAT adoption, shown in Table 3.<sup>6,22</sup>

Because fugitive emission from coke batteries No. 3–4 and 5–6 cannot be measured experimentally, their EFs were assumed to lay in the upper half of the European range  $\{EF = [(EF_{\max} - EF_{\min})/2 + EF_{\min}]\}$ . The corresponding absolute emissions (t/yr) with or without BAT values (t/yr) in Table 3 were accordingly calculated for each pollutant on the basis of the yearly production level of these plants already described. These data permit the quantification of the relevant environmental impact of the plants under consideration. As shown in Table 3, their absolute emissions would be appreciably reduced by adopting steel sector BAT, already introduced in the 1980s by the German TA-Luft<sup>23</sup> and by the Italian Ministerial



**Table 3.** Emission Factors from coke ovens.<sup>6</sup>

Pollutant	European Range (g/t of crude steel)		Emissions Estimated from Batteries No. 3-4 and 5-6 in Taranto (t/yr)	
	With BAT	Worst Case	Present	With BAT
Particulate	17	75	167-270	61
CO	130	1,500	2900-5400	468
VOC	4	8	22-29	14
Benzene	0.3	15	29-54	1
PAHs	0.17	0.5	1.2-1.8	0.6
H <sub>2</sub> S	4	20	43-72	14
NH <sub>3</sub>	0.8	3.4	08-12	3
HCN	0.02	0.04	0.11-0.14	0.07
SO <sub>2</sub>	27	950	1759-3420	97
NO <sub>x</sub>	230	600	1494-2160	828

Decree July 12, 1990, updated by Italian Legislative Decree August 4, 1999 No. 372, which is still waiting for national regulation.

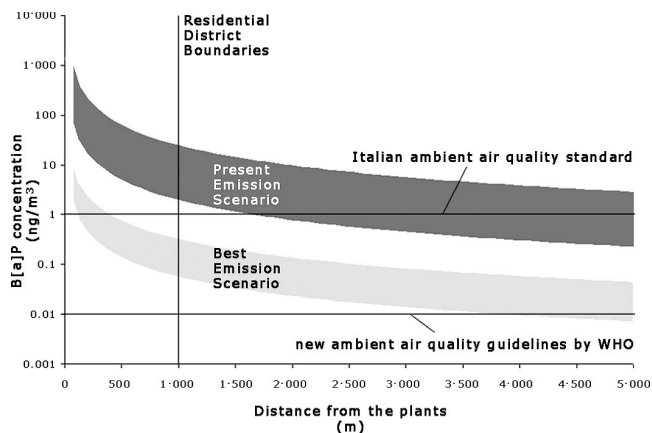
### BaP Dispersion and Population Exposure

Total PAH emission factors for the Taranto coking plants No. 3-4 and 5-6 has been estimated to be in the range of 0.8-1.2 g/metric tons of coal. Based on this figure, the dispersion of airborne pollutants in surrounding areas along different wind directions, particularly toward the southeast, where a densely populated residential suburb ("Tamburi") is found, has been calculated.

The dispersion model used in this study was aimed at assessing the background concentration of total PAHs in the particle-bound phase, expressed as toxicity equivalent to BaP. The EPA Fugitive Dust Modeling (FDM, 93070 version) program was used, an analytical air quality model specifically designed for analyzing the dispersion of fugitive dust. It incorporates the transport, dispersion, and deposition of pollutants in the atmosphere, using particulate matter (particle radius, density, etc.) and air-flow characteristics (mean velocity, wind direction, etc.) as input data.

The diffusion equation of pollutant transport is solved with the use of several simplifying assumptions. In particular, the pollutant is assumed to be characterized by different classes of uniformly sized particles, and the diffusion along the x direction is considered negligible in comparison with the advection term along that direction. Furthermore, it is assumed that eddy diffusivities depend on downwind distance and not on travel time from the source. Based on these assumptions, the concentration and deposition rates at several receptor sites are calculated.

The deposition mechanism in the FDM is based on the Ermak equations (1977). In the development of the deposition treatment, two velocities were considered: settling because of gravity (Stokes' law) and deposition velocity, which accounts for removal of particles by all methods (e.g. turbulent diffusion) from the atmosphere to the earth's surface ( $z = 0$ ). This can be expressed as:

**Figure 5.** BaP dispersion downstream coke-oven batteries No. 3-4.

$$\left[ K \frac{\partial c}{\partial z} + Wc \right]_{z=0} = vc|_{z=0} \quad (1)$$

where  $c$  is the pollutant concentration ( $\text{g}/\text{m}^3$ ),  $K$  is the eddy diffusivity ( $\text{m}^2/\text{sec}$ ),  $W$  is the gravitational settling velocity, and  $v$  the deposition velocity (both  $\text{m}/\text{sec}$ ).<sup>30</sup> A source extending along the entire surface of the coke-oven batteries was assumed.<sup>24</sup> Because BaP was present  $\sim 100\%$  in particulate phase, prevalingly in the fine fractions, the simulation assumed the  $\text{PM}_{2.5}$  particulate fraction (with an average density  $1 \text{ g}/\text{m}^3$ ) as the mean carrier agent, as reported in the literature.<sup>25</sup>

Different meteorological scenarios were considered either favorable or unfavorable to dispersion. Local prevailing wind direction is from the northwest (i.e., perpendicular to the longitudinal front of the batteries), upstream to the residential suburb, with a typical intensity of 5-10  $\text{m}/\text{sec}$  (at 10 m above the ground level) and with atmospheric stability class (Turner's formulas) A, D, and F with air temperature of 25 °C as reported by the local meteorological station.

Simulations were conducted according to estimated particulate emission (167-270 t/yr) as well as assuming 60 t/yr emission after full BAT adoption (see Table 3). Resulting BaP concentrations were calculated at 50 ground-level receptors placed at 100 m distance and at 0, 10, and 20 m above ground-level.<sup>26-30</sup>

Results of FDM simulation under the conditions discussed above for such plants, without (present situation) or with BAT (possible future situation), are reported in Figure 5 and compared with BaP limits in living ambient in force in Italy, as well as with WHO guidelines.<sup>15</sup> Although the estimated BaP concentration decreases rapidly with distance, in the residential area near the factory, it remains quite high (2-25  $\text{ng}/\text{m}^3$  at 1000 m from the source), largely exceeding the Italian limit in force.

### CONCLUSIONS

On the basis of a 7-month investigation on diffuse air emission from some coke-oven batteries of one of Europe's largest steel factories (Taranto, Italy), total PAH concentrations as gases and solids (i.e., adsorbed onto fine dust particles) were found largely exceeding TLV-TWA set

by ACGIH for occupational exposure. This agrees with heavy emissions of other toxic pollutants (TSP, PM<sub>3,5</sub>, and benzene) from the same plants reported previously.<sup>3,4</sup>

The speciation profile evidenced the presence of six indicative PAHs, namely Nap, Pyr, BbF, IND, BaP, and AcPy, which account for 77% of the total PAH concentration, with BaP and DBA mostly responsible (85%) for the powerful carcinogenicity of the emissions investigated. Using EPA FDM model, dust dispersion outside steel factory boundaries near the city of Taranto has been found to exceed existing Italian BaP limits in living ambient, as well as the WHO guidelines, even assuming full adoption of steel-sector BATs.

Preliminary experimental evidence of excess PAH concentration in the urban area has been achieved through a public monitoring network (14 automatic air sampling stations) installed recently in Taranto's metropolitan area to overcome the chronic leak of air quality data. These data will be presented in a paper now in preparation. After the results of this investigation, in September 2002, the coke-oven plants investigated were shut down by the local authorities.

#### ACKNOWLEDGMENTS

The authors thank the Breda Research Institute (Bari, Italy), in particular Giuseppe Vitucci and Vincenzo Cagnazzo, who carefully carried out analytical sampling and measurement activities.

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