

Research Article

Indoor/Outdoor Air Quality Assessment at School near the Steel Plant in Taranto (Italy)

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Received 9 January 2017; Accepted 20 February 2017; Published 22 May 2017

Academic Editor: Pedro Salvador

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This study aims to investigate the air quality in primary school placed in district of Taranto (south of Italy), an area of high environmental risk because of closeness between large industrial complex and urban settlement. The chemical characterization of PM_{2.5} was performed to identify origin of pollutants detected inside school and the comparison between indoor and outdoor levels of PAHs and metals allowed evaluating intrusion of outdoor pollutants or the existence of specific indoor sources. The results showed that the indoor and outdoor levels of PM_{2.5}, BaP, Cd, Ni, As, and Pb never exceeded the target values issued by World Health Organization (WHO). Nevertheless, high metals and PAHs concentrations were detected especially when school were downwind to the steel plant. The *I/O* ratio showed the impact of outdoor pollutants, especially of industrial markers as Fe, Mn, Zn, and Pb, on indoor air quality. This result was confirmed by values of diagnostic ratio as B(a)P/B(g)P, IP/(IP + BgP), BaP/Chry, and BaP/(BaP + Chry), which showed range characteristics of coke and coal combustion. However, Ni and As showed *I/O* ratio of 2.5 and 1.4, respectively, suggesting the presence of indoor sources.

1. Introduction

Children spend most of day in indoor environments and especially in school classrooms [1]. Indoor air quality (IAQ) in schools has been identified as an important factor that can affect children health, because of a large exposure to indoor air pollutants in that environment.

Airborne particles in classroom have been recognized as the principal class of contaminants that can affect indoor air quality and students health [2, 3]. Furthermore, these particles consist of several compounds such as mineral fibers, heavy metals, and toxic chemical substances which may pose health concerns to occupants [4]. Numerous epidemiological studies have pointed out that exposure to PM is correlated with a large variety of adverse acute and chronic health effects, including heart and respiratory diseases [5, 6]. Those repercussions on health are even more adverse for children that have a respiratory system yet non-fully developed. The health effect of particles depends strongly on their size,

specific surface area, number, and chemical composition. PM size is a key factor affecting the percentage of particle deposition in different parts of the respiratory system. Along with PM size, chemical components (including polycyclic aromatic hydrocarbons (PAHs), other organic carbons, elemental carbon, sulfate and nitrate salts, and metals) also affect the toxicity of PM [7]. Several studies have highlighted that PAHs and metals represent the two main PM classes correlated with the adverse health effects [8–10]. The PAHs bound to PM represent an increasing concern because of their carcinogenicity and mutagenicity [8]. Studies have shown that particle-bound metals are relevant in causing mitochondrial damage [11] and induction of oxidative stress [12], which ultimately leads to an increase in cardiovascular mortality and morbidity [13]. These potentially toxic elements may originate from various industrial or urban sources. The indoor PM concentration variability is determined by the activities that occur inside the building. However, outdoor air contaminants may intrude indoors by two ways: by means of

opened windows or doors and by penetration through rifts in the building envelope [14].

Until now, only few studies have characterized the chemical composition of indoor particles and little is known regarding the school environment [4]. More studies on the PM concentrations and their composition in schools are essential to assess the exposure of children to airborne particles. Generally, the typical indoor PM sources, such as heating combustion, cooking, and smoking, are missing in school buildings. Nevertheless, classrooms are characterized by a complex environment that depends on several factors such as number of occupants, activities, ventilation system, outdoor PM concentration and composition, tightness, and furniture and materials of school building [15]. Besides, the site-specific characteristics of the investigated area play an important role in the indoor PM concentration when the school is located nearby an industrial area. Indeed, the PM is considered an outdoor pollutant that can be found in indoor environment by means of intrusion or in presence of discontinuous sources such as smoking of cigarettes, candles burning, chalks use, and resuspension [16].

The aim of the present study is to characterize IAQ in a primary school building located nearby the high-impact industrial sites such as the industrial area of Taranto (south of Italy).

The industrial area of Taranto is, in fact, considered one of the areas of high environmental risk and it is included in the list of polluted sites of national interest because of the presence of a large industrial complex near the urban settlement.

2. Material and Methods

2.1. PM Sampling Campaign. PM_{2.5} samples were collected inside the atrium of the school building located in a district of Taranto city, called Tamburi, few kilometres away from the biggest European steel plant. Taranto (40°28'N 17°14'E) is the third most populated city of the Southern Italy and its industrial area includes the biggest steel plant in Europe, a refinery, a quarry, a cement plant, a composting plant, and military and trade harbors. Because of the proximity of this large industrial complex to the urban settlement (Tamburi district), Taranto is one of the areas identified at high environmental risk in Italy and it has been included in the list of the polluted sites of national interest (Figure 1).

Eleven daily samples of PM_{2.5} were collected from 11 to 22 December 2014 on quartz fiber filters (Whatman) using low-volume air sampler (Skypost by TRC TECORA) and FAI sampling heads operating to 2.3 m³ h⁻¹. According to UNI EN 1234-1, the collected PM filters were conditioned for 48 hours in a climatic chamber provided with a control system for the temperature and the humidity (20 ± 1°C and 50 ± 5% RH) (Activa Climatic, Aquaria, Milan, Italy) and then, the particulate mass on the filter was measured by using an analytical balance (Sartorius Genius series, mod. SE2, Germany) with a sensitivity of 0.0001 mg.

Then, the PM filters (47 mm diameter) were cut in two parts: half of the filter was analysed in order to determinate

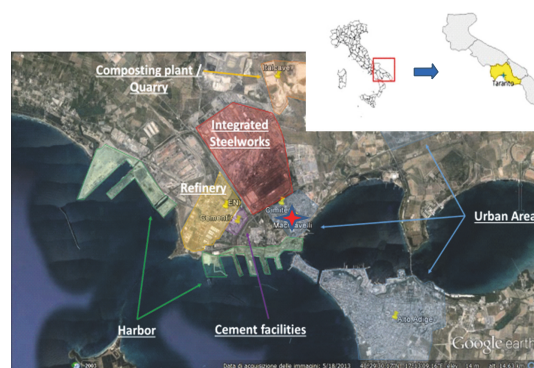


FIGURE 1: Map of Taranto (south of Italy). Red star shows sampling site.

elements (Al, V, Cr, Fe, Mn, Co, Ni, Cu, Zn, As, Cd, and Pb) and the other half was analysed for PAHs determination (Benzo[a]anthracene (BaA), benzo[b+j]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IP), benzo[ghi]perylene (BgP), and dibenzo[a,h]anthracene (DBA)).

Simultaneously, real-time monitoring of total PAHs was carried out by means Ecochem PAS monitor (Saras S.p.A.) in order to study the activation of sources during the monitored days. This monitor, using an Excimer lamp at a determined wavelength, ionizes only the carbon aerosols, while gas molecules and noncarbon aerosols remain neutral. The positively charged particles are collected on a filter inside an electrometer, where the charge measured is proportional to the concentration of total PAH. Indoor/outdoor (*I/O*) comparisons, useful to evaluate the different *I/O* contributions and to study the intrusion phenomena, were guaranteed by proximity between the selected school and the regional air quality station called Machiavelli, on the same street and 200 meters ahead with respect the investigated school. Only data about European regulated pollutants as PM, BaP, Ni, Cd, Pb, and As, about total PAHs monitored in addition to meteorological data, were obtained for Machiavelli station by regional air quality network.

2.2. Chemical Characterization of PM_{2.5}. Half of PM_{2.5} filters were extracted with a mixture of acetone/hexane by means of a microwave assisted solvent extraction (Milestone, model Ethos D). The extracted samples were analysed using an Agilent 6890 PLUS gas chromatograph (Agilent Technologies, Inc., Santa Clara, CA, USA) equipped with a programmable temperature vaporization injection system (PTV) and interfaced to a mass spectrometer, operating in electron impact ionization (Agilent MS-5973 N). Benzo[a]anthracene (BaA), benzo[b+j]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IP), benzo[ghi]perylene (BgP), and dibenzo[a,h]anthracene (DBA) were determined using the signals corresponding to the molecular ions: BaA (228), BbF (252), BkF (252), BaP (252), IP (276), BgP (276), and DBA (278). Perylene-D12 (PrD, 264) was used as internal standard (IS). The analytical performances (extraction recovery,

TABLE 1: Mean, minimum, and maximum concentration of PM2.5 ($\mu\text{g}/\text{m}^3$), PAHs, and metals (ng/m^3) measured inside the investigated school.

Mean (min–max) ng/m^3	This study	Viana et al., 2014 [28]	Zwoździak et al., 2013 [23]	Hassanvand et al., 2015 [24]	Oliveira et al., 2017 [30]
Location	Taranto, Italy	Barcelona, Spain	Wroclaw, Poland	Tehran, Iran	Oporto, Portugal
BaA	0.15 (0.11–0.23)			24.5	0.046 (0.041–0.051)
Cr	0.21 (0.17–0.28)			39.26	0.13 (0.11–0.15)
BbF	0.51 (0.33–0.71)			23.75	0.7 (0.59–0.83)
BkF	0.29 (0.20–0.39)			12.22	0.12 (0.10–0.15)
BaP	0.22 (0.14–0.32)			5.42	0.27 (0.21–0.33)
IP	0.34 (0.24–0.52)			2.23	0.44 (0.36–0.52)
DBA	0.25 (0.18–0.34)			13.75	1.5 (1.3–1.8)
BgP	0.42 (0.24–0.68)			10.43	0.61 (0.51–0.74)
Al	635.3 (521.8–703.2)		561	39.72	
V	1.17 (0.81–1.92)	3.4			
Cr	14.5 (11.9–19.6)	3.1	6.06	5.55	
Fe	212 (93.9–693)	200	525	102.20	
Mn	4.93 (2.36–8.58)	5.9	46	15.25	
Co	0.18 (0.13–0.25)				
Ni	5.24 (3.20–10.6)	2.3	2.04	4.66	
Cu	13.9 (6.7–26.8)	9.1	50	22.17	
Zn	15.5 (5.20–30.5)	42.1	267	68.90	
As	0.64 (0.42–0.81)	0.34	4	3.01	
Cd	0.09 (0.05–0.15)	0.69		0.29	
Pb	3.81 (1.65–10.6)	3.73	85	60.60	
PM2.5 ($\mu\text{g}/\text{m}^3$)	16.6 (10.6–27.1)	29.9	59.8 (18.2–86.6)	20 (3–81)	25.8 (9.2–66.6)

extraction linearity, analytical repeatability, and LOD) were verified in our previous work [17].

In addition, the other half of PM2.5 samples were digested in 8 ml of nitric acid and 2 ml of hydrogen peroxide solution by using a microwave system (Milestone mod. Ethos D). The extract was analysed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Perkin Elmer NexION 350) in order to determine Al, V, Cr, Fe, Mn, Co, Ni, Cu, Zn, As, Cd, and Pb using Rh as internal standard. Recoveries varied from $91.4 \pm 2.8\%$ to $95.8 \pm 1.1\%$, for all investigated metals. Limits of detection (LODs) ranged from $0.02 \text{ ng}/\text{m}^3$ for Cd to $0.27 \text{ ng}/\text{m}^3$ for Pb. Standards were analysed daily, as well as filter blank, every 5 samples to verify instrumental performance. Each quantification was performed in triplicate.

3. Results and Discussion

3.1. PM Mass Concentrations. The basic statistics (mean, maximum, and minimum) of mass and chemical composition of PM2.5 determined for the investigated period and school are summarized in Table 1.

During the investigated period, no significant differences can be observed for PM2.5 concentrations determined inside and outside the school. The mean PM2.5 concentrations

were 16.6 and $15.9 \mu\text{g}/\text{m}^3$ for indoor and outdoor environments, respectively. These values were lower than the European target settled ($25 \mu\text{g}/\text{m}^3$) and comparable to those obtained in other regions of the world (e.g., between 10.7 and $14.8 \mu\text{g}/\text{m}^3$ in three schools in Quito, Ecuador [1]; between 12 ± 2.3 and $24.7 \pm 16 \mu\text{g}/\text{m}^3$ in Italian classroom [18]; between 2.8 and $13.9 \mu\text{g}/\text{m}^3$ in Swedish schools [19]). Moreover the values determined in this study were lower than those found inside classrooms located in centre of European metropolis in London ($30 \mu\text{g}/\text{m}^3$, [20]); Athens ($82 \pm 56 \mu\text{g}/\text{m}^3$, [16]); Barcelona ($8.3\text{--}95 \mu\text{g}/\text{m}^3$, [21]); Munich ($19.3\text{--}105.9 \mu\text{g}/\text{m}^3$, [4]); Antwerp ($11\text{--}166 \mu\text{g}/\text{m}^3$, [22]); and Wroclaw ($44\text{--}143 \mu\text{g}/\text{m}^3$, [23]).

The mean I/O ratio was 1.08 ± 0.37 ; thus, PM levels slightly higher were found inside the school suggesting a contribution of indoor sources to PM2.5 components. Moreover, the indoor/outdoor correlation for PM2.5 (Figure 2) showed a zero coefficient of determination (R^2) for the collected data. The flat slopes (0.21) was due to the high intercepts obtained (13.25) that it indicated a contribution of indoor sources to PM2.5 [21]. This finding could be attributed to extreme physical activity leading to the resuspension of coarse particles in the school.

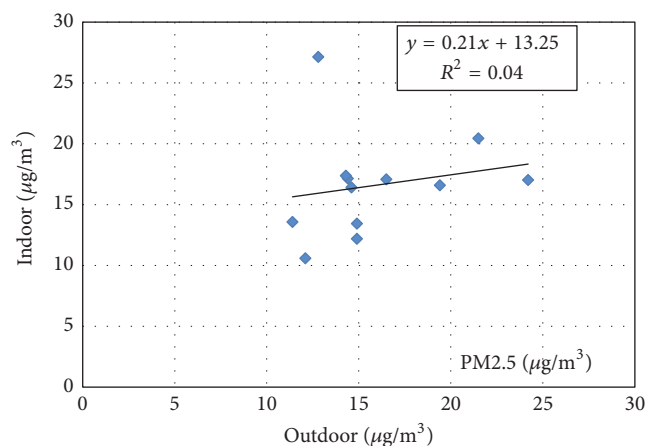


FIGURE 2: Scatterplot showing the indoor/outdoor correlation for PM2.5.

3.2. Chemical Characterization of PM

3.2.1. Metals. The mean levels of metals measured in indoor PM2.5 samples during sampling campaign are summarized in Table 1. The most abundant metals detected were Al (521.8–703.2 ng/m³) and Fe (93.9–693.5 ng/m³). These values were comparable with those determined in a secondary school located in the centre of Wrocław, Poland [23], but higher than those measured inside schools at Tehran [24]. Several studies have reported that the presence of Fe and Al in PM is due mainly to road dust resuspension and also to industrial sources [25, 26]. In fact, during the sampling campaign the indoor levels of metallurgical industry markers as Fe, Mn, Cd, As, and Zn reached their maximum values (693, 8.6, 0.15, 0.81, and 30.5 ng/m³, resp.) on 18 and 21 December, when strong wind blew from north-northwest transporting air pollutants from the nearby steel plant to the investigated school. Fe and Mn are related to park mineral dusting or blast furnace charging, Cd and Zn are emitted by sintering processes, and, finally, As is released by mining and smelting of base metals or fuel combustion [27].

The concentrations of Pb determined in this study were comparable to those measured in Spain [27] and lower than those found in Poland and Tehran [23, 24]. Although Pb has been found to be usually emitted from sources such as fuel and motor oil combustion, in the developed world the use of leaded gasoline decreased since 1990s due to national regulations, determining a drastic reduction of Pb levels in atmosphere. Therefore, the indoor and outdoor concentrations of lead determined in this work (1.65–10.6 ng/m³ for indoor and 3.3–37 ng/m³ for outdoor) were lower than the annual national ambient air quality standards (500 ng/m³). However, considering the trend of Pb concentrations like that of industrial markers, and maximum values of Pb concentration registered in correspondence of N-W wind direction, an industrial source could be hypothesized for Pb. It could be probably involved in sintering processes [24].

In addition to lead, also Ni, Cd, and As, classified by the IARC as carcinogenic to humans (Group 1), never exceeded

standard annual outdoor levels (20, 6, and 5 ng/m³, resp.) settled by European Commission (EC) [28]. However, Ni and As, unlike the other metals, showed *I/O* ratio higher than 1 (2.5 and 1.4 in average, resp.) suggesting the existence of some indoor sources. School equipment made from treated wood or paints and dyes used for Christmas gifts hand-made by children inside school could be probably the indoor sources of these pollutants [23].

Although V concentrations resulted lower than those measured in Spanish schools [27], the highest values were determined on 13 and 19 December 2014, when wind blowing from west-southwest probably allowed the transport of this pollutants from refinery to the investigated school.

Finally, Cr levels resulted up to 5 times greater than those obtained in other works (Table 1), probably due to a joint effect of two sources: industrial and traffic ones. In fact, Cr is emitted from road traffic and it mainly derives from both break wear [29, 30]. However, metallurgical industry has been also identified as anthropogenic source of Cr [24].

3.2.2. PAHs. Among individual PAHs bound to PM, benz[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]luoranthene, benzo[a]pyrene, dibenz[a,h]anthracene, and indeno[1,2,3-cd]pyrene were designated by International Agency Research on Cancer as possible or probable carcinogens (IARC, 2002, 2010) and were analysed in this work. The levels of these carcinogenic PAHs (i.e., ΣPAHs) at the indoor air of the studied school ranged from 1.51 and 2.36 ng/m³ accounting for 14.3% of PM2.5 concentration. The indoor concentrations of the eight carcinogenic PAHs, that is, BaA, Chr, BbF, BkF, BaP, BgP, DBA, and IP, were similar to those found in a primary school of Portugal [28] and up to two orders of magnitude lower than the values determined in Tehran [22]. The most abundant indoor PAHs bound to PM2.5 were BbF, BgP, IP, BkF, and DBA, accounting for about 22%, 18%, 14%, 12%, and 10% of the total PAHs concentration, respectively. High abundances of DBA and BgP highlight vehicular traffic source (light-duty gasoline) while BbF and IP abundances suggest emissions from coke oven [31–33]. In fact, according to trend of Fe, Mn, Zn, and Pb concentrations, PAHs showed the highest values in correspondence of wind blowing from industrial area to the investigated school.

As concerns benzo[a]pyrene, indicator of carcinogenic PAHs, the current European legislation on ambient air (Directive 2004/107/EC, 2005) sets annual target value of 1 ng m⁻³ for carcinogenic PAHs in PM10. The mean concentration of this pollutant during the sampling period was 0.22 ± 0.07 ng m⁻³ and 0.32 ± 0.13 ng m⁻³ in indoor and outdoor environments, respectively; and they never exceeded the target value.

One of the most important aspects of the air quality management process is the identification of the source of pollutants. Indoor-to-outdoor (*I/O*) concentration ratios of individual PAHs provide a rough identification of the pollution origin. The PAH concentrations in outdoor and indoor air determined in this work were not statistically different and the mean *I/O* ratio for BaP was 0.82±0.38, indicating outdoor

TABLE 2: DRs range determined in literature and in this study.

	BaP/Chry	B(a)P/B(g)P	IP/(IP + BgP)	BaA/(BaA + Chry)
Traffic [34, 35]		0.5–0.6	0.17 [35]	
Diesel emissions		0.45–0.83		
Gasoline cars		0.30–0.40	0.21–0.22 [33, 40–43]	0.76 [33, 45]
Lead smelter (coke burning)		0.45 [36]	0.36 [36, 38]	
Coke combustion	>0.35 [33, 45]	≥1.25 [33, 37]	0.33 [39]	
Coal burning		0.9–6.6 [38, 39]	0.56 [34, 38]	0.27–0.65 [33, 45]
Wood combustion			0.62	0.43 [33, 45]
This study's mean (min–mx)	1.06 (0.76–1.32)	0.64 (0.30–1.35)	0.45 (0.31–0.55)	0.41 (0.37–0.52)
Taranto, outdoor [31]		0.77 (0.27–1.55)	0.47 (0.29–0.56)	

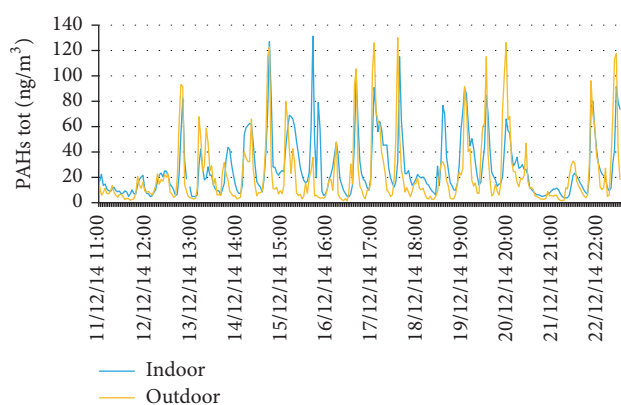


FIGURE 3: Trend of real-time PAHs concentrations monitored inside and outside the investigated school.

origin of these contaminants. This finding was confirmed by real-time monitoring of total PAHs. In fact, as shown in Figure 3, concentrations and trends of total PAHs monitored inside and outside the school were comparable except for some morning hours on 15–16 December 2014, when higher indoor concentrations were detected. In the same days, the BaP *I/O* ratio also reached its maximum value (ranging from 1.2 to 1.4), suggesting potential contribution of indoor sources. Probably, the use of artistic materials as wax melting to prepare Christmas gifts hand-made or the burning candles during preparative Christmas show contribute to high indoor concentration of PAHs [34].

In addition, analysis of diagnostic ratios of individual PAH congeners can provide further insight regarding PAHs origin. In fact, because of different sources show specific concentration profiles of PAHs, several studies focused on PAHs diagnostic ratios, especially on B(a)P/B(g)P and IP/(IP + BgP) ratios [34–46]. The value of B(a)P/B(g)P, IP/(IP + BgP), BaP/Chry, and BaA/(BaA + Chry) ratio inside the investigated school ranged from 0.30 to 1.35, from 0.31 to 0.55, from 0.76 to 1.32, and from 0.37 to 0.52, respectively (Table 2). These results were comparable with those determined in

Taranto industrial area for outdoor [31] and, except for B(a)P/B(g)P ratio, showed the strong impact of industrial area and especially of steel plant on indoor and outdoor air quality in Taranto. However, even if lower mean values were determined for B(a)P/B(g)P ratio, the analysis of these depending on the wind direction showed that the highest values of B(a)P/B(g)P ratio ranging from 1.18 to 1.35, typical of coke combustion, were determined when winds blew mainly from the NW sector. On the contrary, these DR values decreased to 0.5–0.6, typical of vehicular traffic contribution, when the schools located near the steel plant were upwind of industrial area.

4. Conclusions

The main goal of this work was to assess the impact of the biggest European steel plant on nearby residential area of Taranto (South of Italy) and, in particular, on indoor air quality inside school. Therefore, a PM_{2.5} sampling campaign was carried out from December 11 to 22, 2014, in a primary school located in Tamburi district of Taranto. The chemical characterization of PM provided PAH and metal concentrations. In addition, real-time total PAHs were monitored inside and outside the investigated school. The results showed that the indoor and outdoor levels of PM_{2.5}, BaP, Cd, Ni, As, and Pb were lower than guidelines issued by World Health Organization (WHO). Nevertheless, high metals and PAHs concentrations were detected especially when schools were downwind to the steel plant. The *I/O* ratio showed the impact of outdoor pollutants, especially of industrial markers as Fe, Mn, Zn, and Pb, on indoor air quality. This result was confirmed by diagnostic ratio as B(a)P/B(g)P, IP/(IP + BgP), BaP/Chry, and BaA/(BaA + Chry), which showed range values typical of coke and coal combustion. However, Ni and As showed *I/O* ratio of 2.5 and 1.4, respectively, suggesting the presence of indoor sources. These results could be probably due to the use of school equipment made from treated wood or paints and dyes for the Christmas gifts hand-made by children.

Disclosure

This manuscript is based on a paper presented to TECH-AIR 2016 conference.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

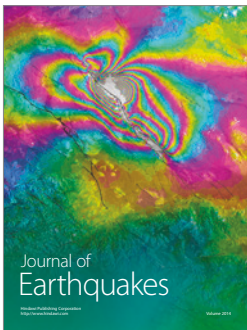
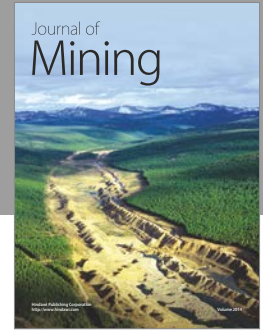
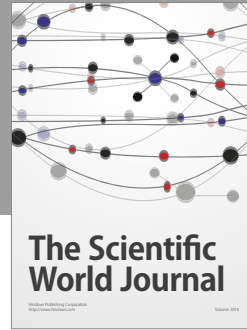
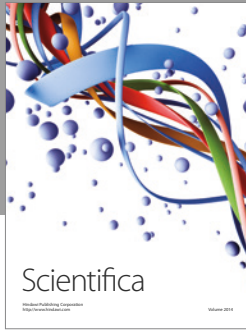
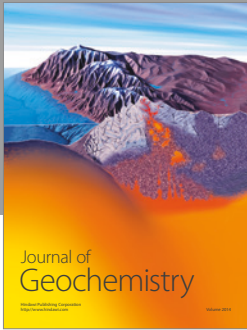
Acknowledgments

Special appreciation goes to ARPA Puglia, especially to Dr. Dambruoso Paolo Rosario and Dr. Placentino Claudia Marcella for the collaboration in sampling campaign.

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