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Oxidant generation capacity of source-apportioned PM_{2.5}

Polina Maciejczyk¹, Mianhua Zhong², Morton Lippmann², and Lung-Chi Chen²

¹Eckerd. College, St. Petersburg, Florida, USA

²New York University, Department of Environmental Medicine, Tuxedo, NY, USA

Abstract

While many studies found associations between ambient particulate matter (PM) and morbidity or mortality outcomes, it is unclear whether these associations are dependent on the composition of PM, which varies with the source of that PM. We addressed this knowledge gap by conducting a time-series PM-health effects assessment that specifically investigated the role of source-apportioned fine PM (PM_{2.5}) on the oxidant generation capacity that might be responsible for respiratory and cardiovascular health outcomes. Daily PM_{2.5} composition speciation and black carbon (BC) measurements, conducted in rural New York for 303 days between March 2003 and January 2005, were analyzed by a factor analysis source apportionment model, and five source categories (transported aerosol/secondary sulfate, resuspended soil, metals, residual oil combustion, industrial/incineration) were identified. After the exposure of human epithelial cells (BEAS-2B) to our PM_{2.5} samples, cellular NF-κB activation showed a relatively significant association with the Ni concentration averaging 38 ng/m³, and weaker but still significant correlations with Ba (13 ng/m³), Mn (9 ng/m³), and Fe (500 ng/m³). The single-source category regression analysis of NF-κB signal showed a significant association only with the metals source. Thus, three metals, which together constitute only 60 ng/m³ to the PM_{2.5} mass (107 μg/m³), appear to be more influential on lung cell responses than BC (6 μg/m³) and sulfate ions (36 μg/m³) that are present at much higher mass concentrations.

Keywords

ambient particles; PM_{2.5}; oxidative stress

INTRODUCTION

There have been many studies that find an association between ambient particulate matter (PM), specifically PM_{2.5} (particulate matter with an aerodynamic diameter of less than 2.5 μm) pollution, and adverse health outcomes in humans. As the specific physical and chemical properties of PM, as well as their sources, remain unclear, it is unlikely that any single chemical component of PM_{2.5} is responsible for all adverse health outcomes. Most recent reviews agree that the human health effects should not be attributed simply to the total mass concentration but endorse the need to ascertain the toxicity associated with PM

Correspondence address to: Lung Chi Chen, Ph.D., Department of Environmental Medicine, NYU School of Medicine, 57 Old Forge Road, Tuxedo, NY 10987, lung-chi.chen@nyumc.org.

components (Fanning et al. 2007), specifically metallic elements (Lippmann and Chen, 2009), inorganic compounds (Schlesinger, 2007) and carbonaceous PM components (Mauderly and Chow, 2008). Source apportionment receptor models provide estimates of the contributions of various source categories to ambient air pollution at a given location at a given time. Various sources are distinguishable by the source apportionment because they have distinctive tracers, and correlations that are closely associated with each source profile. With several methods used to resolve the composition of PM into components related to emission source categories, identification of the most causal source categories and/or specific PM components responsible for most of the adverse health effects would permit more focused control of more casual PM_{2.5} components and their effects.

As inhaled particles deposit upon epithelial cells of the respiratory tract, they facilitate an inflammatory response via activator protein-1 (AP-1) and nuclear factor kappa B (NF- κ B) transcription factors (Levis and Mossman 2007; Porter et al. 2007), which then induce a variety of proinflammatory cytokine factors and adhesion-molecule genes. Transition metals have long been suspected to be major components of ambient PM in producing adverse health effects because of their reduction-oxidation potential and the production of reactive oxygen species (ROS). Many studies specifically have shown that metals in ambient air PM altered intracellular redox state, resulting in activation of these transcription factors (U.S.EPA, 2004). We have previously reported an *in vitro* exposure technique to compare the daily variations of the cellular responses to fine concentrated ambient particles (CAPs) collected in Tuxedo, NY, a rural area upwind of New York City (NYC) (Maciejczyk and Chen, 2005). Detailed chemical characterization of the CAPs allowed us to investigate the source categories and individual components of ambient PM that were associated with the induced cellular response NF- κ B. In that first study, we found that NF- κ B expression in BEAS-2B lung cells was significantly associated with a residual oil combustion source factor which represented only 2% of total PM_{2.5} mass and was significantly associated with vanadium (V) and nickel (Ni). This current paper provides comprehensive dataset of additional two campaigns of sub-chronic daily exposure of normal mice and a mouse model of atherosclerosis. The results of the mice exposure is described elsewhere (Chen et al 2010, 2005). Here we attempt to address whether daily compositional changes in ambient PM_{2.5} altered cellular response in human epithelial cells using an expanded dataset.

METHODS

Particulate Sample Collection and Analysis

The exposure PM_{2.5} CAPs were produced using a versatile aerosol concentration enrichment system (VACES) in Sterling Forest, Tuxedo, NY, in a largely undeveloped woodland New York State Park that is about 30 miles northwest of the NYC urban center in Manhattan. The NYU laboratory is located near the center of the Sterling Forest State Park, on a relatively lightly traveled two-lane road that bisects the park. There are no large power generators or industrial operations within 20 miles of the site. The overview methods of monitoring of the exposure atmosphere and ambient aerosol are discussed in Lippmann et al. (2005) and Maciejczyk et al. (2005). Daily 6-hr particulate air samples (ambient and nominally 10X concentrated, CAPs) were collected with VACES exposure system on

Teflon filters (GelmanTeflo, 37mm,0.2 μ m pore) for the period of 9 a.m. to 3 p.m. on weekdays only in three separate campaigns, for 303 days between March 2003 and January 2005. Study dates were March 4 – September 5, 2003; February 10 – May 7, 2004, and July 16, 2004 – January 12, 2005.

The outdoor monitoring included semi-continuous measurements of ambient black carbon (BC), as a surrogate index of elemental carbon (EC) concentrations, recorded every 5 min throughout the study using an Aethalometer (Thermo Electron). We had previously reported a good correlation between the BC concentrations measured by Aethalometer and Sunset Lab EC measured on quartz filters ($r^2 = 0.67$), with Aethalometer's concentration 2.8 $\times$ higher than those of EC as measured by Sunset Lab (Chen et al. 2010). Meteorological parameters relevant to this manuscript included wind speed and wind direction as measured on the roof of the laboratory. Filter samples were stored at constant temperature and relative humidity ($21 \pm 0.5^\circ\text{C}$, $40 \pm 5\%$ RH) until analyzed. Filter masses were measured on a microbalance (model MT5, Mettler-Toledo Inc., Highstown, NJ). Analyses for 34 elements followed by nondestructive energy dispersive x-ray fluorescence, ED-XRF (model EX-6600-AF, Jordan Valley) using five secondary fluorescers (Si, Ti, Fe, Ge, and Mo), and spectral software XRF2000v3.1 (U.S. EPA and ManTech Environmental Technology, Inc.), as described in Maciejczyk et al (2005).

Detection of NF- κ B

The VACES system was also attached to a liquid particle collector (BiosamplerTM, SKC Inc, Eighty-four, PA, USA) to collect samples for *in-vitro* analyses into aqueous suspensions for indirect cell exposure. In total, 247 samples were collected. In these three exposure campaigns, we used the BEAS-2B cells (American Type Culture Collection, Manassas, VA) rather than primary cells to eliminate the confounding factor of biodiversity, and ensured uniform and reproducible responses to PM_{2.5} exposure of a homogeneous cell population. Prior to cell exposure, each Biosampler sample was diluted with cell media for a final concentration of 300 μg PM/mL. The protocol for NF- κ B detection using a Luciferase assay was described previously (Huang et al., 2001; Maciejczyk&Chen, 2005). Briefly, BEAS-2B cells stably transfected with NF- κ B luciferase reporter gene are cultured in DMEM (Dulbecco's modified Eagle's medium, Bio-Whittaker) supplemented with 10% FBS (fetal bovine serum, Cambrex), 100 IU/ml penicillin (Cellgro), 100 μg /ml streptomycin (Cellgro), and 2 mM L-glutamine (Gibco). The cells were cultured at 37°C in a 5% CO₂ incubator. The cultures were dissociated with trypsin–ethylenediamine tetraacetic acid (Gibco) and transferred to new 35-cm² flasks once a week. The cells were seeded to 96-well plates at a density of about 9×10^4 /well, and cultured until 95% confluence. Cells were treated with an aliquot of the PM suspension for 24 h. The negative control was medium-treated cells; the positive control was vanadyl sulfate trihydrate- (200 nM, 100 nM, 50 nM, 12.5 nM) and TPA (10 ng/ml)-treated cells. After applying the lysis reagent and luciferase assay system reagents (Promega), the cell responses in terms of NF- κ B were determined with a luminometer, and are discussed in terms of the signal increase compared to the negative control.

Source Apportionment and Cell Response

To identify major particle source categories, Na, Mg, Al, Si, S, Cl, K, Ca, V, Mn, Fe, Ni, Cu, Zn, Se, Br, Ba, Pb, and BC were used as independent variables in source apportionment. The selection of species was based on findings from previous source apportionment studies conducted for the New York area, known elemental tracers, and XRF detectability. Nitrates and organic carbon (OC), major constituents of PM, were only measured during first campaign March 4 – September 5, 2003, and used for source apportionment published previously (Maciejczyk and Chen, 2005). However, for the second and third campaign nitrates and OC were not available, and thus were not modeled in overall three-campaign source apportionment. Factor Analysis model with varimax rotations were applied to the PM trace constituents and BC. To obtain the daily mass contribution from each source, the “absolute” scores were regressed onto the gravimetric mass concentrations (Thurston and Spangler, 1985). The elemental dataset was also verified by alternate factor analysis oblimin rotation and with Positive Matrix Factorization (PMF-2) source apportionment model.

The NOAA HYSPLIT (hybrid single-particle Lagrangian integrated trajectory) backward trajectory on-line model was judiciously used to investigate the origination of air masses associated with particular days when unusually high PM mass or elemental concentrations occurred. Meteorological data for backward trajectory modeling were downloaded from the EDAS archive (horizontal resolution of 40 or 80 km). The 24-h or 48-h backward trajectories starting at the height of 20 m and 100 m above the ground level were computed for key days of interest. The combination of source apportionment, and back trajectories provided information on the source categories for integrated samples, and indicated the air mass origin. Finally, daily sources’ mass contributions, as well as individual elements, were correlated with daily NF- κ B levels.

RESULTS

CAPs mass and elemental concentrations

In total, 303 CAPs samples and 19 variables were used for modeling purposes. The average concentration with standard deviation of 19 elements, BC, and total mass are shown in Table 1. Based on gravimetric mass analysis, VACES concentrating factor between ambient PM and CAPs was 8.9 in 2003, and 9.9 in 2004. The overall average mass concentration was $107 \pm 83 \mu\text{g}/\text{m}^3$. During the summer months (June–August), CAPs mass concentrations were generally higher (summer average $152 \mu\text{g}/\text{m}^3$) versus other times of the year (annual average $88 \mu\text{g}/\text{m}^3$). The higher summer concentrations are associated with secondary sulfate aerosol formations in the summertime and, as found in previous work, is primarily associated with transported aerosols from the Midwest (Qin et al, 2006; Lall et al, 2006). In fact, the 10 highest mass contributions of transported aerosol source category in this dataset occurred in summer between June and August. The largest measured contributors to the PM mass were S and BC, followed by Si, Na, and Fe.

Source apportionment

All three source-apportionment models (Factor analysis using varimax and oblimin rotations as well as PMF analysis) identified five source categories. Between three and seven factors

were tried with each source apportionment model. The disagreement between the models was primarily on mass contribution of each source to overall CAPs mass concentration. In order to distinguish among source type contributions, the measured chemical characteristics must be such that they are present in different proportions in different emission sources. Traffic factor was difficult to separate due to lack of a specific tracer, sampling period that most likely missed the rush hour, and lack of traffic in general at this rural site. The number of factors in the final analysis was based on the evaluation of the interpretability of the resulting source profiles; and in seventh source could not be clearly identified. In a six-source model in factor analysis, one factor was identified as non-significant ($p > 0.01$) by the statistical analysis of individual source coefficients. Thus, the five-solution model was selected. The factor loadings for each factors source category by varimax rotation is shown in Figure 1. Mass contributions of each source to overall mass were computed by each factor score regression onto daily CAPs mass, and are the averages of daily mass contributions per source are shown in Figure 2.

We assigned the factor rich in S and BC to long-range transported aerosol. Sulfate is mostly from coal combustion power plants, but also can result from vehicular emission, residual oil combustion, steel and coke complexes, and from smelters. Se is mainly a coal-fired power plant emission, but is also emitted in lower levels from residual oil combustion. This source category had the largest mean contribution to overall mass of $70.5 \mu\text{g}/\text{m}^3$ (or 66%).

The re-suspended soil factor (rich in Si, Ca, Al) had a mean contribution $16.7 \mu\text{g}/\text{m}^3$ (or 16% of total mass). It also had substantial loading of Zn which in urban areas may derive primarily from worn tires and diesel lubricating oil, both indicative of traffic source. However, with this dataset, traffic and soil were difficult to separate, and what we were observing is the surface soil that is aerosolized by motor vehicle tires passing over the roadway.

The third factor (assigned as 'metals'), which was characterized by high concentrations of Ni, Fe and Mn, with the highest levels on December 15, 2004, and November 29, 2004. The mean daily contribution from the 'metals' source was rather small, i.e., only $2.2 \mu\text{g}/\text{m}^3$ (or 2.1%). This source category was consistent with the distant upwind Sudbury Ni refinery plume enriched in Fe and Ni that was described in our earlier studies (Lippmann et al 2006, Chen et al 2010).

The fourth factor was rich in V, Ni, and Br, and was assigned as residual oil combustion emitted from power plants of the eastern United States and from ocean-going ships (especially those docked in the Port of New York, whose major facilities are at Port Newark and Port Elizabeth in New Jersey)). Mass contribution from this source was $0.17 \mu\text{g}/\text{m}^3$ (or 0.16%). The highest mass contribution for residual oil combustion source was primarily under north and north east winds and occasional southeast winds. The average Ni concentration ($38 \text{ ng}/\text{m}^3$) was much higher than the concentration of V ($23 \text{ ng}/\text{m}^3$), consistent with the report by Peltier and Lippmann (2010) that attributed, a high Ni/V ratio to emissions from residential and commercial buildings within the northern areas of NYC. They attributed the low Ni/V ratio in the southern parts of NYC to vessel bunkering, which constitutes the majority of residual oil consumption in New Jersey at the large port facilities

at Port Elizabeth and Port Newark. Presence of Br in this source category is most likely an unresolved traffic component brought in to our site with NYC emissions under south-easterly winds. Due to the lack of unique tracer constituent(s) associated with the traffic source, NO₂ (a marker of local combustion) is commonly included in the source apportionment to assist in the identification and separation of a “traffic” factor. Unfortunately, we did not have a complete dataset of NO₂ or nitrate ion for this analysis.

The last factor contained Pb, Br, Se, Zn, and Cu, and was assigned as an industrial/incineration source. Its mean contribution to CAPs mass concentration was 7 µg/m³ (6.5%). The highest concentrations of this source were observed mainly under southerly winds that brought emissions from New Jersey and NYC. Although Zn mostly originates from tire wear, it is also attributed to incinerators and metal working. This miscellaneous industrial category could have also contained fresh coal-fired power plant emissions, given the very high Se levels under wind direction from the northeast, where a coal-fired power plant is located at Stony Point, NY, about 20 miles from Sterling Forest.

Cellular responses

The average NF-κB response was 11.4 ± 8.9 (in terms of the arbitrary luminescent signal increase compared to the negative control), and its daily variation are shown in Figure 2. We first considered the CAPs composition concentrations as a covariate for estimating cellular effects. As shown in Table 1, using single-component regression analysis daily NF-κB signal showed a significant association with daily changes in Ni concentration, although the correlation coefficient was small ($r = 0.256$, $p < 0.001$). Cellular response also had weak but still significant correlations with Ba ($r = 0.194$, $p = 0.0022$), Mn ($r = 0.181$, $p = 0.0043$), and Fe ($r = 0.17$, $p = 0.0075$). Then, we considered the daily mass contributions from individual trace element-based source category as covariates for estimating acute effects. Again, daily NF-κB signal was regressed with each source daily mass contribution. As shown in Table 3, only the ‘metals’ source category correlated significantly with NF-κB, with $r = 0.305$, $p < 0.001$.

DISCUSSION

We used an *in vitro* exposure technique to compare daily relative response of cells to concentrated fine ambient particles collected from a rural area upwind of NYC. NF-κB belongs to a family of transcription factors that are involved in the regulation of a large number of normal cellular processes, such as immune and inflammatory responses, cellular growth, and apoptosis. Thus, the activation of NF-κB is thought to be part of a stress response as it is activated by a variety of stimuli that include growth factors, cytokines, radiation, and pollution. The NF-κB activation is indicative of the oxidative stress that eventually triggers the release of various cytokines. The cytokines produced by the bronchial epithelial cells after exposure to PM are capable of contributing to the local inflammatory response in the lung. Further, the release of these cytokines into the circulation could induce a systemic response that includes stimulation of the bone marrow to release leukocytes and platelets (Fujii et al., 2002; Goto et al., 2004).

The advantage of the source-apportioned approach is that it allowed us to examine the association between cell responses and categories of source-related pollutants that vary together (due to the same source type, or weather condition), rather than trying to separate the effects of individual pollutants. In our previous analysis (Maciejczyk and Chen, 2005) we used the subset of chemical composition data for CAPs used in current paper (March – September 2003, 126 samples and 22 variables) in factor analysis with varimax orthogonal rotation to determine four particle source categories contributing significant amounts of mass to CAPs at Sterling Forest. These source categories were: (1) regional secondary sulfate, characterized by high S, Si, and OC; (2) resuspended soil, characterized by high concentrations of Ca, Fe, Al, and Si; (3) ROFA emissions from ships (especially in the Port of New York City) and power plants of the eastern United States, identified by the presence of V, Ni, and Se; and (4) unknown other sources. The latter was most likely a combination of incineration rich in Pb and Zn, and long-range transported motor vehicle traffic due to presence of nitrate (nitrate data was available during the first campaign). To estimate the mass contributions of each individual source category, the CAPs mass concentration was regressed against the factor scores. Regional sulfate was the largest contributor to mass (65%), followed by soil (20%), residual oil combustion (2%), and the other sources contributing 13%. In that *in vitro* exposure study involving source apportionment, the NF- κ B response was notably correlated with source containing V and Ni. These elements were correlated with oil combustion source category, as r-value was 0.30 and 0.29 for Ni and V, respectively. Thus, not surprisingly, the NF- κ B response correlated with oil combustion source category.

In contrast to that previous study, in this combined source apportionment analysis, the dataset was extended to 303 samples of 19 variables from three campaigns which provided us with enough statistical power to identify five source categories: transported aerosol/secondary sulfate, resuspended soil, metals, residual oil combustion and industrial/incineration, that contributed to particle mass concentrations. The difference between the sum of average daily contributions and CAPs mass average daily concentration was 10.6 $\mu\text{g}/\text{m}^3$, which means that 9.9% of mass was not explained. Note that mass contribution of each source to overall mass was computed by each factor score regression, which could be negative, onto daily CAPs mass. Since we could not perform mass reconstruction due to lack of data on OC, nitrate, and ammonia, these mass contributions reflect only components measured, and should be used for references with caution.

The notable separation of a metal source in this new analysis was mainly driven by high concentrations of Ni, Fe and Mn, and had a significant implication to the cellular function correlation. The backward trajectory analysis tracked the highest mass contributions of this source to the distant upwind Sudbury Ni refinery that was described in our previous study (Lippmann et al., 2006), as well as residual oil combustion sources in northern New Jersey and NYC as shown on Figure 3. Nickel (but not V) can also come from Ni smelters, and there can be local sources of both metals. Generally, fine particle Ni and V are often associated with the combustion of residual fuel oils either in ships or in power plants. In our analysis the correlation coefficient between Ni and V was $r = 0.547$, indicating that these elements were not always emitted from the same source(s). Large spatial and seasonal variations of fine particle Ni are observed through the NYC metropolitan region (Peltier and

Lippmann, 2010; Peltier et al., 2009) where space-heating boilers residual fuel oil used for space heating contains much more Ni than V in NYC, whereas the power plant and shipboard combustion in the Port of New York are the major source of V. Since the source loadings for metal source category did not contain V, we assume that the contribution of oil combustion to metals category was negligible. Our analysis indicated that no correlation between residual oil and metals source categories.

We have found that NF- κ B response correlated with Ni but not V as shown in Table 1, and with the metals source category but not residual oil combustion as shown in Table 2. These results indicate that the NF κ B activating component is primarily attributable to Ni. And in the absence of metals sources, the residual oil combustion source category induces the cellular effect due to presence of Ni. Zhang et al (2009) considered all findings on Ni and V of exposure, epidemiological, and toxicological studies, and concluded that Ni and V are positively correlated with human cardiovascular and respiratory diseases. The recent review by Lippmann and Chen (2009) implicated residual oil fly ash as the most toxic source-related mixture, and Ni and V, as particularly influential components in terms of acute cardiac function changes and excess short-term mortality. However, in contrast to the previous study (Maciejczyk and Chen 2005), with a larger datasets it was statistically possible to separate a 'metals' source, and that is what we observed. The implication of our analysis were also confirmed in the subchronic inhalation studies of ApoE^{-/-} mice predisposed to atherosclerosis where highly significant increases in heart rate variability (HRV) and decreases in HRV were observed during fourteen days characterized as having much higher than average concentrations of Ni, Cr, and Fe, but much lower than normal concentrations of fine particles, S, Al, and V. The observed cardiovascular health effects were significantly associated with effects of human exposure to fine ambient PM rich in Ni. Lippmann et al. (2006) regressed the daily average mortality coefficient against PM_{2.5} and its chemical component concentrations reported for 2000–2003 by the U.S. Environmental Protection Agency (EPA) Speciation Trends Network (STN) for the 60 US cities with such data among the 90 cities in the National Mortality and Morbidity Study (NMMAPS; Dominici et al., 2003), and reported that only Ni and V were significantly associated with the significantly higher daily mortality in NYC than in other US cities. Dominici et al. (2007) then extended the regression analyses of NMMAPS mortality and STN composition data out to 2005 for 69 US communities, and found that their results were consistent with those of Lippmann et al. (2006), but that when the NYC data, which were much higher in Ni concentration than any other U.S. city, were excluded from their analysis, the overall associations of mortality coefficient with Ni and V were no longer statistically significant. Together, these findings suggest that responses to Ni exposure have a threshold for measureable effects.

CONCLUSIONS

Human epithelial cells (BEAS-2B) exposed to PM_{2.5} samples collected in a rural area upwind of NYC were analyzed for cellular NF κ B activation, and the associations with the concentrations of PM_{2.5} mass, its elemental components, black carbon (BC), and source-related mixtures were analyzed. A relatively strong and significant association was exhibited with the Ni concentration (averaging 38 ng/m³), and there were weaker, but still significant

correlations with Ba (13 ng/m³), Mn (9 ng/m³), and Fe (500 ng/m³). The source category regression analysis of NF- κ B signal showed a significant association only with the 'metals' source. Thus, three metals, which together constitute only 60 ng/m³ to the PM_{2.5} mass (107 μ g/m³), appear to be much more influential on lung cell responses than BC (6 μ g/m³) and sulfate ions (36 μ g/m³) that are present at much higher mass concentrations. The ultimate goal of current PM research is to identify the components most responsible for adverse effects so regulations can be more targeted (Lippmann 2010). Our results showed that metals, specifically Ni, in PM_{2.5} were the important source for cellular oxidant generation and may be responsible for subsequent health effects associate with particle air pollution.

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Figure 1.
Factor loadings of five factor scores determined by Factor Analysis with varimax rotation.

Figure 2.

Daily mass contribution of each source category (in $\mu\text{g}/\text{m}^3$). The NF κ B response is in terms of the signal increase compared to the negative control.

Figure 3. Examples of NOAA HYSPLIT back trajectories on days with highest mass contribution of (a) metals source category under north-west winds; (b)) metals source category under other winds; (c) residual oil combustion category.

Table 1Concentrations of selected elements (in ng/m³), and correlation coefficient r with NF-κB response

	Average (ng/m ³)	Std. dev. (ng/m ³)	Corr. coef. r with NF-κB
Na	560	940	0.075
Mg	110	180	0.01
Al	350	450	-0.16
Si	940	830	-0.05
S	12000	12000	-0.14
Cl	66	260	-0.03
K	250	190	-0.07
Ca	180	140	-0.05
V	23	43	0.089
Mn	9.1	17	0.181 *
Fe	500	440	0.17 *
Ni	38	56	0.256 *
Cu	12	19	-0.12
Zn	81	78	-0.14
Se	11	14	-0.11
Br	27	23	-0.13
Ba	13	46	0.194 *
Pb	28	52	-0.12
EC	6000	4200	0.104

*
p<0.01

Table 2Correlation coefficients (r) of the cellular response NF- κ B with the sources identified by factor analysis

Source	r-value	p-value
Resuspended soil	-0.0525	0.411
Metals	0.305	<0.0001
Industrial/inciner	-0.144	0.0233
Transported	-0.090	0.161
Residual oil	0.093	0.146