PM_{10} chemical characterization and seasonal variations in a high density urban area nearby Venice, Italy

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An air quality monitoring network with mobile stations has been arranged to evaluate the impact of a new highway building in the Venice hinterland, an area characterized by the presence of several anthropogenic pollutant sources. PM_{10} samples collected on filters (low-volume samplers) from November 2005 to August 2007 in rural and urban background areas and in construction sites were subsequently analyzed for PAHs (BaA, BaP, BbF, BkF), water-soluble ions (sulphate, nitrate, ammonium, calcium, potassium, magnesium) and trace elements (arsenic, cadmium, nickel, lead). The evidence of a seasonal winter-summer trend is noticed only for a few parameters, mainly PM_{10} well correlated to a reference urban background fixed station. Principal components analysis applied to average data from PM_{10} chemical characterization points out well-defined clusters among monitoring sites. Some interesting differences are observed with reference to building activities, anthropogenic combustion sources and local industrial emissions, highlighting the significant role of meteorology on atmospheric pollution.

1. Introduction

In North Italy the Po Valley represents a unique case due to the situation of severe atmospheric pollution with regards to particulate matter PM_{10} (EEA, 2007, pp. 39-41). Every year limit values set by the 99/30/EC directive are exceeded and critical events frequently occur especially during autumn and winter in this basin, geographically bordered by mountain chains and a closed sea. Scientific evidence of chronic and acute effects on human health attributable to different levels of PM_{10} in 5 North Italian cities is acknowledged in terms of mortality, morbidity and years of life lost (Martuzzi et al., 2006). Similarly to the rest of the Valley, several pollutant sources insist on the Veneto Region, where densely populated towns coexist with suburban areas, industrial activities, agricultural practises and road traffic. In particular the city of Venice-Mestre is subjected to the daily flux of heavy duty vehicles moving along the so-called European “Corridor 5”. Under these conditions, a new highway was projected in order to remove traffic away from the urban centre. Building activities started in 2005 and are going to be completed in 2009. Air quality evaluation has been carried out from the beginning of the project paying great attention to the key-pollutant PM_{10}, before and during building activities. The aim of this study was to investigate the complexity of urban aerosol and its different pollutant sources by means of PM_{10} chemical characterization concearning PAHs, ions and trace elements, thus improving local knowledge on the contribution of various species to PM_{10} in the Venice hinterland.

2. Experimental

2.1 Monitoring strategy and sites description

The study area was represented by the new highway future path in the Venice hinterland within a range of about 10 km from the city centre of Venice-Mestre (Figure 1). Near-city and suburban background stations were chosen rather than urban ones, since the aim of the network was direct to monitor air quality in construction and control sites, with regards to human health and ecosystem targets. Instead of fixed stations, a mobile monitoring was experimented by means of campaigns carried out in fall-winter (1st Oct-31 Mar) or spring-summer (1st Apr-30 Sep), before and during building activities. Every campaign is identified in Table 1 with initials indicating the site in chronological starting order and the season (winter or summer). A “C” is added with reference to construction sites and “2” if the same site is monitored for the second time.

2.2 Sampling and analysis

Daily PM_{10} samples were collected on filters using low-volume samplers carrying out 24 campaigns in 18 sites from November 2005 to August 2007. Every investigation lasted 30-45 days, except in site A where sampling was carried out quite permanently. Two types of PM_{10} sampler were used: Skypost PM HV (TCR Tecora, Italy, flow rate 2.3 m^3 h^{-1}) and SWAM5a (FAI Instruments, Italy, flow rate 2.3 m^3 h^{-1}), both equivalent to standard method EN 12341 (CEN, 1998) followed to determine PM_{10} concentration. Depending on further chemical analysis, several types of membrane filters (47 mm diameter) were used: binder free glass fibre filters (PALL and Millipore) for PAHs, teflon membrane filters (PALL, 2 μm pore size) for ions and mixed cellulose ester filters (Sartorius and MFS, 0.8 μm pore size) for trace elements. In each campaign the filters frequency was every 4 days for mixed cellulose ester filters and every 11 days for teflon membrane filters, glass fibre filters in remaining days. Chemical analysis on filters were performed with inductively coupled plasma mass spectrometry according to EN14902 (CEN, 2005) for trace elements (As, Cd, Pb, Ni), ion chromatography (Xiu et al., 2004) for water soluble ions (K^+, Ca^{2+}, Mg^{2+}, NH_4^+, NO_3^-, SO_4^{2-}) and HPLC-Fluorescence (Noël et al., 1996) for PAHs: benzo(a)anthracene (BaA), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP).

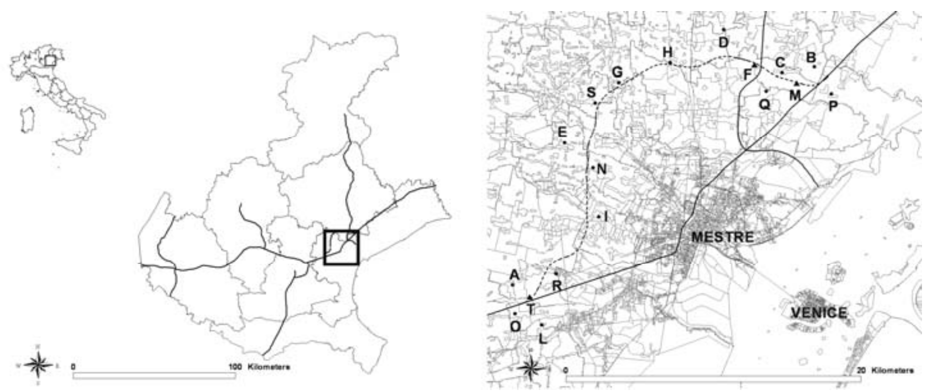

Fig. 1. The study area with the new highway future path (dashed) and monitoring sites.

Table 1. Experimental results obtained for each sampling campaign from November 2005 to August 2007. PM_{10} average, standard deviation (SD), minima and maxima concentrations; PAHs, trace elements and ions average concentrations (±SD).

Site	PM_{10} (µg m^{-3})			BaP (ng m^{-3})	BaA (ng m^{-3})	BbF (ng m^{-3})	BkF (ng m^{-3})	As (ng m^{-3})	Cd (ng m^{-3})	Ni (ng m^{-3})	Pb (ng m^{-3})	Mg^{2+} (µg m^{-3})	NH_4^+ (µg m^{-3})	NO_3^- (µg m^{-3})	SO_4^{2-} (µg m^{-3})	K^+ (µg m^{-3})	Ca^{2+} (µg m^{-3})
	Average ±SD	Min.	Max.														
Aw	65 ± 33	13	157	6.20 ± 3.28	5.37 ± 3.54	5.90 ± 2.89	3.15 ± 1.59	5.7 ± 3.5	2.0 ± 0.9	4.7 ± 0.6	36.9 ± 8.3	0.08 ± 0.06	2.53 ± 1.06	7.28 ± 2.65	2.86 ± 1.18	0.77 ± 0.34	0.96 ± 0.79
Bw	50 ± 33	12	127	2.43 ± 2.18	1.76 ± 1.85	2.71 ± 2.16	1.32 ± 1.05	8.2 ± 6.2	4.2 ± 2.2	3.8 ± 1.4	18.9 ± 12.7	0.06 ± 0.04	2.63 ± 1.17	9.69 ± 5.27	3.60 ± 0.69	0.32 ± 0.13	0.71 ± 0.62
Cw	50 ± 20	15	85	0.80 ± 0.49	0.63 ± 0.31	1.09 ± 0.62	0.53 ± 0.28	6.0 ± 3.6	4.4 ± 3.0	3.8 ± 1.3	16.7 ± 9.8	0.13 ± 0.10	3.67 ± 1.39	13.50 ± 8.56	5.06 ± 1.15	0.31 ± 0.05	0.58 ± 0.25
Ds	29 ± 10	5	42	0.05 ± 0.03	0.02 ± 0.01	0.09 ± 0.05	0.03 ± 0.02	4.3 ± 2.8	2.6 ± 2.0	4.7 ± 2.6	11.4 ± 4.9	0.08 ± 0.06	1.55 ± 1.39	5.00 ± 4.21	3.41 ± 0.42	0.12 ± 0.08	0.54 ± 0.33
Es	31 ± 13	5	50	0.08 ± 0.05	0.07 ± 0.06	0.12 ± 0.06	0.04 ± 0.03	3.7 ± 2.5	2.5 ± 2.1	5.3 ± 3.1	13.1 ± 6.0	0.13 ± 0.07	1.78 ± 1.21	4.47 ± 3.63	3.67 ± 2.66	0.18 ± 0.15	0.72 ± 0.46
As	36 ± 17	7	87	0.07 ± 0.05	0.05 ± 0.03	0.11 ± 0.06	0.05 ± 0.03	1.9 ± 1.3	1.1 ± 1.0	4.0 ± 2.1	12.7 ± 6.4	0.12 ± 0.07	1.74 ± 0.87	3.26 ± 3.22	3.23 ± 2.18	0.16 ± 0.07	0.75 ± 0.47
Fs**C**	61 ± 30	9	122	0.05 ± 0.02	0.04 ± 0.01	0.08 ± 0.03	0.03 ± 0.01	4.5 ± 2.8	5.2 ± 4.8	5.6 ± 2.0	9.8 ± 2.5	0.24 ± 0.09	2.02 ± 0.69	4.72 ± 3.65	4.21 ± 1.80	0.19 ± 0.06	3.10 ± 0.84
Gs	36 ± 18	9	71	0.06 ± 0.03	0.03 ± 0.01	0.09 ± 0.04	0.04 ± 0.02	2.7 ± 1.2	5.5 ± 3.4	2.2 ± 1.3	6.3 ± 2.6	0.09 ± 0.06	1.62 ± 0.64	3.17 ± 1.91	3.35 ± 1.88	0.16 ± 0.05	1.07 ± 0.21
Hs	30 ± 17	8	82	0.03 ± 0.01	0.02 ± 0.01	0.06 ± 0.01	0.04 ± 0.02	1.8 ± 1.0	3.2 ± 2.1	4.3 ± 2.7	10.0 ± 3.2	0.08 ± 0.07	2.32 ± 0.29	2.77 ± 1.71	6.21 ± 1.82	0.14 ± 0.02	0.53 ± 0.28
Is	31 ± 14	8	61	0.03 ± 0.01	0.02 ± 0.01	0.06 ± 0.03	0.02 ± 0.01	1.3 ± 0.6	0.5 ± 0.1	1.6 ± 0.8	10.5 ± 5.2	0.18 ± 0.08	1.63 ± 0.54	1.36 ± 0.76	4.85 ± 1.70	0.20 ± 0.09	1.18 ± 0.46
Ls	33 ± 20	6	74	0.03 ± 0.01	0.02 ± 0.01	0.06 ± 0.02	0.02 ± 0.01	1.9 ± 0.9	0.5 ± 0.1	2.6 ± 1.5	8.4 ± 2.9	0.15 ± 0.10	0.77 ± 0.29	0.87 ± 0.21	2.47 ± 1.04	0.06 ± 0.02	1.16 ± 0.60
Ms**C**	50 ± 29	8	145	0.13 ± 0.22	0.06 ± 0.07	0.18 ± 0.23	0.08 ± 0.12	5.9 ± 2.9	2.7 ± 2.4	3.1 ± 2.3	13.0 ± 8.9	0.20 ± 0.08	1.57 ± 0.95	3.17 ± 1.89	2.25 ± 0.94	0.22 ± 0.17	3.55 ± 0.89
Ns	35 ± 22	9	96	0.04 ± 0.01	0.02 ± 0.01	0.07 ± 0.03	0.03 ± 0.02	1.5 ± 0.8	0.7 ± 0.4	2.4 ± 1.8	9.5 ± 3.8	0.14 ± 0.06	0.66 ± 0.43	1.27 ± 0.60	1.82 ± 0.74	0.10 ± 0.02	3.00 ± 2.56
Os	29 ± 15	4	58	0.10 ± 0.15	0.05 ± 0.05	0.29 ± 0.53	0.14 ± 0.27	2.1 ± 1.2	1.5 ± 0.9	3.7 ± 2.5	8.1 ± 4.2	0.15 ± 0.07	0.75 ± 0.19	1.07 ± 0.38	1.37 ± 0.43	0.22 ± 0.13	0.74 ± 0.36
Pw	61 ± 32	21	169	1.32 ± 1.14	0.84 ± 0.82	1.37 ± 1.19	0.73 ± 0.62	10.8 ± 3.5	9.4 ± 3.4	7.4 ± 2.3	28.3 ± 10.0	0.23 ± 0.13	5.78 ± 3.16	17.13 ± 8.08	4.46 ± 1.96	0.65 ± 0.32	3.30 ± 1.74
Qw	66 ± 32	14	142	3.98 ± 2.12	2.89 ± 1.04	4.09 ± 2.30	1.90 ± 1.10	4.0 ± 1.0	3.1 ± 2.6	4.4 ± 3.2	26.9 ± 13.4	0.04 ± 0.03	1.64 ± 0.97	5.51 ± 2.22	1.43 ± 0.33	1.03 ± 0.60	1.95 ± 0.82
Aw2	82 ± 43	10	199	4.38 ± 2.34	2.83 ± 1.23	4.14 ± 2.76	1.93 ± 0.94	4.1 ± 3.2	2.0 ± 1.8	4.2 ± 4.2	31.9 ± 19.4	0.23 ± 0.22	5.31 ± 2.33	14.73 ± 7.65	5.22 ± 5.13	2.20 ± 1.70	1.24 ± 0.62
Rw	73 ± 43	20	185	3.03 ± 1.92	1.89 ± 0.98	2.74 ± 1.66	1.32 ± 0.73	6.7 ± 0.4	2.2 ± 1.3	5.6 ± 2.1	37.3 ± 11.0	0.24 ± 0.19	5.65 ± 3.47	17.58 ± 8.80	4.53 ± 2.04	0.44 ± 0.13	1.67 ± 0.79
Sw	75 ± 39	22	223	5.85 ± 3.51	4.29 ± 3.09	5.29 ± 2.94	2.56 ± 1.41	3.0 ± 2.3	4.1 ± 4.0	5.1 ± 3.5	47.0 ± 43.8	0.26 ± 0.06	2.61 ± 1.96	7.61 ± 5.49	2.45 ± 1.65	0.80 ± 0.34	1.33 ± 0.67
Hw	67 ± 35	13	122	2.29 ± 1.53	1.67 ± 1.35	2.54 ± 1.67	1.15 ± 0.75	2.1 ± 0.8	1.9 ± 0.8	5.3 ± 1.4	22.0 ± 8.5	0.01 ± 0.01	3.83 ± 0.62	10.16 ± 2.63	2.75 ± 0.92	1.08 ± 0.75	0.63 ± 0.13
Tw**C**	64 ± 31	10	139	1.97 ± 0.82	0.95 ± 0.43	2.11 ± 1.62	0.92 ± 0.70	4.0 ± 2.8	2.3 ± 2.0	4.8 ± 2.2	24.4 ± 11.4	0.26 ± 0.13	3.51 ± 1.66	10.00 ± 5.13	2.97 ± 1.39	0.86 ± 0.44	1.27 ± 0.72
As2	31 ± 13	7	73	0.08 ± 0.18	0.04 ± 0.07	0.11 ± 0.19	0.05 ± 0.10	2.2 ± 2.1	1.1 ± 1.3	3.7 ± 2.7	10.4 ± 4.4	0.15 ± 0.17	2.26 ± 1.96	4.41 ± 6.36	4.33 ± 2.04	0.21 ± 0.15	0.85 ± 0.45
Hs2	26 ± 10	9	54	0.02 ± 0.01	0.01 ± 0.01	0.03 ± 0.01	0.01 ± 0.01	1.7 ± 1.6	2.3 ± 2.1	3.7 ± 3.2	8.7 ± 4.7	0.10 ± 0.08	1.56 ± 0.59	1.16 ± 0.48	3.99 ± 2.33	0.20 ± 0.11	0.92 ± 0.49
Es2	26 ± 8	11	41	0.03 ± 0.02	0.02 ± 0.02	0.04 ± 0.01	0.02 ± 0.01	1.8 ± 1.6	1.7 ± 1.6	2.9 ± 2.4	8.8 ± 3.4	0.12 ± 0.03	1.45 ± 0.65	1.43 ± 0.58	3.97 ± 2.31	0.21 ± 0.12	1.15 ± 0.68

s: summer period – w: winter period – Xx**C**: construction site – Xx2: replica

3. Results and discussion

3.1 PM_{10} chemical characterization

Average, minima and maxima PM_{10} concentrations are listed in Table 1 for each campaign, followed by PAHs, trace elements and ions average concentrations with their standard deviations. PM_{10} mean values range from 26 to 82 µg m^{-3}, in agreement with results recently obtained in stations close to Venice (Rampazzo et al., 2008). Winter (w), summer (s) and year 2006 mean concentrations, derived in Table 2 from the whole data set, are comparable with values obtained in Milan (Marcazzan et al., 2001), even though quite lower due to sampling at near-city background stations rather than at urban sites in the city centre. Average PM_{10} concentration for every w-campaigns (69 µg m^{-3}) is about double with respect to all s-campaigns (36 µg m^{-3}). Average PM_{10} value (44 µg m^{-3}), referred to samples collected in 2006, agrees with data from Bologna (Van Dingenen et al., 2004) and exceeds annual limit value of 40 µg m^{-3} set by the 99/30/EC directive. PAHs (BaA, BaP, BbF and BkF) concentrations range from detection limit levels in summer to 6.2 ng m^{-3} for BaP in winter (Table 1), and generally show good agreement with results obtained in Zagreb (Šišović et al., 2008) for winter, summer and annual values (Table 2). Mean BaP concentration for year 2006 (0.97 ng m^{-3}) is quite equivalent to annual target value of 1 ng m^{-3} set by the 2004/107/EC directive. Trace elements Ni, Pb (Table 1) show w/s-concentrations lower than Milan levels (Marcazzan et al., 2001). Average annual As, Cd, Ni concentrations (Table 2) are in agreement with some Spanish cities (Moreno et al., 2006) and do not exceed target values set by the 2004/107/EC directive. Ions concentrations in the Venice hinterland are comparable to results from near-city background stations in Europe, in particular for nitrates and sulphates (Putaud et al., 2004). A seasonal w/s-trend in the study area is evident only for a few parameters such PM_{10}, PAHs, Pb and NO_3^-, showing w-maxima and s-minima.

Average PM_{10} concentrations obtained for each campaign are compared to a reference urban background station in Venice-Mestre city centre, approximately 10 km far from monitoring sites. Figure 2 shows a good correlation with reference to control sites data, whereas construction sites are characterized by higher concentration levels.

Table 2. Average concentrations referred to the whole data set.

		Winter	Summer	2006
PM_{10}	µg m^{-3}	69	36	44
BaP	ng m^{-3}	3.70	0.06	0.97
BaA	ng m^{-3}	2.64	0.04	0.79
BbF	ng m^{-3}	3.60	0.10	1.00
BkF	ng m^{-3}	1.75	0.04	0.52
As	ng m^{-3}	4.7	2.5	4.0
Cd	ng m^{-3}	3.0	1.9	3.0
Ni	ng m^{-3}	4.7	3.7	4.1
Pb	ng m^{-3}	29.6	10.3	13.4
Mg^{2+}	µg m^{-3}	0.16	0.14	0.14
NH_4^+	µg m^{-3}	3.68	1.68	2.09
NO_3^-	µg m^{-3}	10.93	3.07	5.15
SO_4^{2-}	µg m^{-3}	3.63	3.55	3.41
K^+	µg m^{-3}	1.02	0.17	0.29
Ca^{2+}	µg m^{-3}	1.35	1.40	1.49

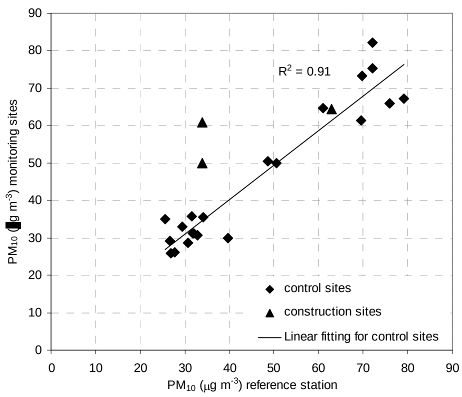

Fig. 2. Scatterplot of PM_{10} values.

3.2 Multivariate analysis

Principal components analysis was applied to PAHs, ions and trace elements mean concentrations to compare different monitoring sites, with respect to PM_{10} chemical characterization. All the 14 analytes determined on PM_{10} samples were involved in the multivariate analysis contributing to the setting of two significant principal components, with 70% of total variance explained. The relative load of each analyte represented in Figure 3 determines the projection of campaigns on the plane built up by the two significant principal components, as shown in Figure 4. In this plot the first component (PC1) distinguishes very well summer (s) from winter (w) sites: a well-defined cluster in the negative PC1 semi-axis includes s-sites whereas all w-sites are spread in positive PC1 semi-axis. This output can be explained by means of a higher level of some pollutants (PAHs, Pb, NO_3^-) in winter samples than in summer ones, according to PM_{10} concentration seasonal trend. In the summer cluster it is generally hard to distinguish among different sites, suggesting that in the study area the concentration of analytes approaches to a common background level. The position of summer construction sites on principal components plane suggests some interesting differences compared to other cluster sites, since MsC is a boundary cluster point and FsC is shifted towards positive semi-axis of PC2. They are characterized by relative high levels of calcium and magnesium, probably related to soil dust. The relative abundance of these analytes can be connected with excavation and building activities in construction sites, characterized by relevant soil movement.

A major difference among winter sites is generally observed, highlighted by points spread on the score plot plane (Figure 4). Aw and Sw show the highest relative concentration of all determined PAHs and important levels of lead and potassium. The replica of campaign in site A in the same period of the following year (Aw2) shows good similarity to the first one. High relative levels of these particular pattern of pollutants can be related to anthropogenic combustion sources, probably attributable to fine fraction of PM_{10} (Marcazzan et al., 2001). Generally good correlation between potassium and organic pollutants and relative high concentration of these analytes in PM_{10} in winter period is also compatible with wood combustion (Osàn et al., 2002), a common practise employed in domestic heating plants in the rural area around Venice-Mestre. Moreover, summer samples collected in As and As2 show concentrations of PM_{10} and pollutants comparable to other summer sites, supporting the hypothesis of a seasonal related pollutant source.

Pw is visibly different from other points, due to relative high levels of arsenic, nickel and cadmium. This pattern of elements could be related to emissions from local industrial sources (Ni and presumably part of As) combined with traffic emission from a nearby highway, as similarly shown in literature (Ragosta et al., 2008; Moreno et al., 2006; Thomaidis et al., 2003)

PM_{10} chemical characterization does not show a particular pattern for the winter construction site TwC, without evident markers related to building activities. This result could be linked to the outstanding effect of meteorology. In winter, stable atmospheric conditions and periods of persistent thermal inversion play a significant role in maintaining high levels of pollutants, that seem to override the contribute of construction activities.

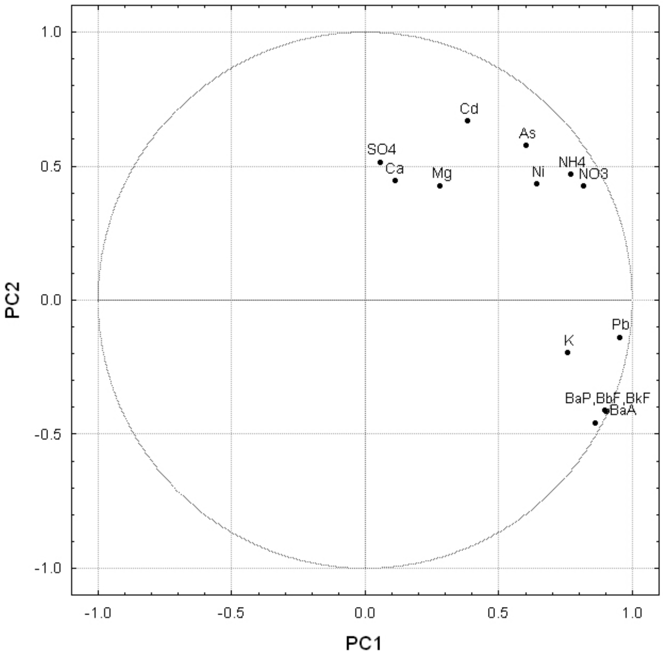

Figure 3. Projection of variables on the PC1-PC2 plane.

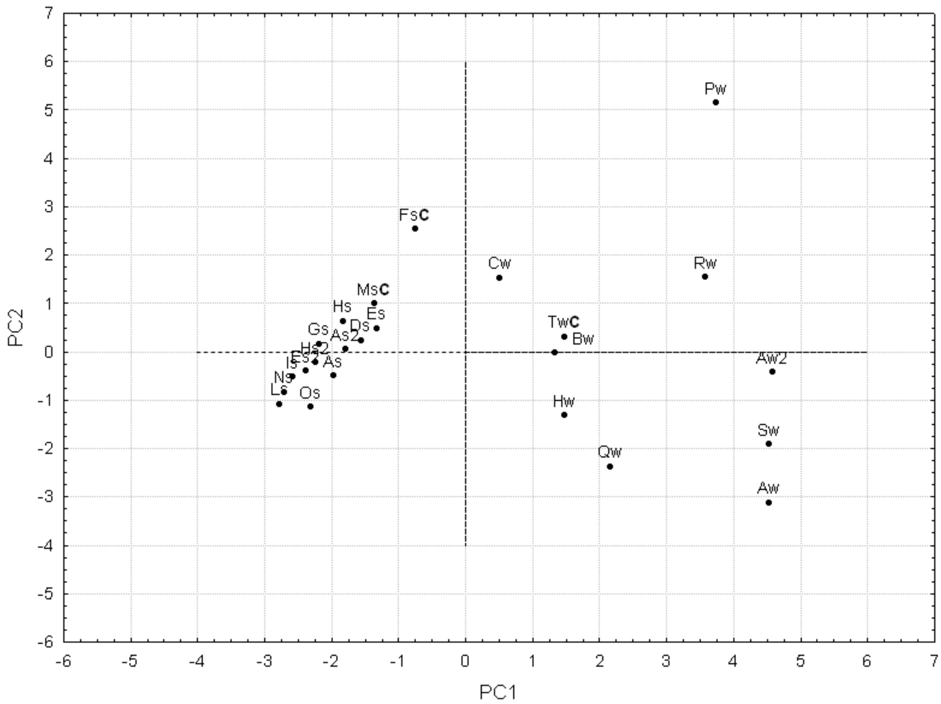

Figure 4. Projection of campaigns on the PC1-PC2 plane.

4. Conclusions

The investigation performed on 18 monitoring sites along the future path of a new highway in the Venice hinterland showed a good correlation between average PM_{10} concentrations in monitoring sites and in an urban background reference station. Data analysis pointed out the seasonal trend of PM_{10} according to some analytes as PAHs, Pb, NO_3^-, with the highest concentrations observed in winter and the lowest in summertime. Summer campaigns showed very similar results among them, suggesting the approach of pollutants concentration to a common background level. Summer construction sites revealed concentrations of PM_{10}, enriched by calcium and magnesium, significantly higher than other summer monitoring sites. This output is consistent with soil movement due to excavation activities in construction sites. In winter, periods of stable atmospheric conditions seem to override the contribute of construction activities to diffuse PM_{10} pollution, due to persistent thermal inversion that played a significant role in maintaining high levels of pollutants.
Moreover this study allowed to make further preliminary hypothesis about particular situations in winter period, probably related to local emission sources. On one side good correlation between potassium and PAHs found in A-site in presence of relative high concentration of PM_{10} could be compatible with wood combustion practises during winter period. On the other, relative high levels of arsenic, nickel and cadmium in P-site could be related to emissions from local industrial sources combined with important traffic emissions.

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