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Ecotoxicological and inorganic chemicals' characterization of rainwater in an urban residential area

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ABSTRACT

A survey of inorganic chemicals and acute ecotoxicity to *Daphnia magna* was performed in rain water samples collected in an urban area. Samples were collected from three types of origins namely ground surface contact, non-contact, collected directly from atmosphere, and roof contact. Chemical and ecotoxicological results varied with the locations and origins of the samples. Data were found to be distributed normally by means of **Kolmogorov–Smirnov** statistics test. All samples were found to be different from each other statistically (100% variance). None of the samples displayed any significant toxicity to *Daphnia magna* (<30% of immobilization after 48-h exposure time). Anyhow acute ecotoxicity data displayed statistically dependent parameters such as pH and EC, and performing chronic ecotoxicological tests would be useful to be able to assess the risk and to determine mitigation strategies for water quality control.

Keywords: Diffuse pollution; Daphnia magna; Ecotoxicity; Inorganic chemicals; Metals; Rainwater

1. Introduction

Water scarcity is becoming the most important problem in the world. The ongoing drought and increase in water demand of the growing population reduce water reservoirs. To tackle this problem, water authorities are adopting several measures including identifying alternative water sources such as stormwater harvesting, graywater, and wastewater reuse and desalination [1–3] among which rainwater results as the most economic and less risk posing one since graywater and wastewater contain more variety of organic pollutants and desalination processes are relatively expensive. In Sweden, analysis carried out on a rainwater collection system for domestic water supply revealed that a significant measure of potable water can be saved if rainwater tanks were re-included as part of a dual water supply solution [4]. The main source of constituents strongly affecting the composition of

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rainwater samples is directly related to the roofing materials (i.e. asphalt fiberglass shingle, galvalume metal, and concrete tile). Besides, the high concentrations of trace metals (Fe, Al, Zn, Cu, Cd, and Pb) in rainwater samples were recorded due to the long-range atmospheric transport of anthropogenic activities (industrial activity, coal combustion, and automobile exhaust), atmospheric pollution, and soil dust [5]. One of the major atmospheric pollutants is ammonia, which originates from livestock waste, fossil fuel combustion, fertilizer production, and agricultural waste [6].

In parallel to chemical analysis, performing ecotoxicity tests are very useful to assess the combined effects of wide variety pollutants instead of assessing their individual effects to conclude complex mixture toxicity. In such a study, toxicity of 36 rainwater samples collected in Achaia Prefecture, Greece, from urban and rural areas was monitored by 48-h acute toxicity tests with the cladoceran *Daphnia pulex*, in the form of the microbiotest kit *Daphtoxkit F pulex*. A 52% and a 46.7% of rainwater samples were found to be toxic in the rural and urban areas, respectively. Toxicity of the rainwater samples was reported to be moderate and was correlated to the presence of the insecticide only in the rural areas [7].

This study aimed to characterize inorganic chemicals and ecotoxicity of rainwater samples collected from different points (in the centrum, traffic-affected, and outer zones of the city), different types (ground surface contact, SC, non-surface contact, NSC, and roof contact, RC), and during different periods (from May to December 2012) in Silivri, İstanbul (Turkey). This study forms the first attempt to combine toxicity to rainwater characterization in Turkey, and it is one of those few studies worldwide.

2. Materials and methods

2.1. Sample collection and conservation

The rain water samples were collected from SC (road or garden ground), RC, and NSC drained from the atmosphere directly to the sample containers in a coastal residential area (Silivri, Istanbul, Turkey) as seen in Fig. 1. The coordinates and dates of sampling area are shown in Table 3. Meteorological data were provided from the State Meteorology Center of Turkey accordingly. Average rainwater fall in the sampling area varies from 500 to 700 mm/year during summer and winter seasons, respectively. The frequency of stormy days and wind velocity are moderately often in winter and rarely often in summer periods. During sampling, relatively moderate periods by means of intensive rain and wind flow days are considered.

Fig. 1. Location of the sampling site in the Maramara Region, Turkey (Source: http://wowturkey.com/forum/viewtopic.php?t=55403).

Samples were collected after the first flash period (after 15 min of the start of rain) in pre-rinsed plastic containers. Samples were collected on different dates to provide representative number of samples from the same sampling locations. Freshly collected samples were delivered to the laboratory, cooled, and kept refrigerated at +4˚C during analysis without adding any chemicals.

2.2. Chemical analyses

Inorganic analyses were performed according to USEPA method [29]. Metals were measured using ICP-OES (Spectro Arcos, Germany) in a Water and Soil Analyses Accredited Laboratory (Hayrabolu, Tekirdağ, Turkey). Since samples were transparent, they were only roughly filtered without any pre-extraction procedure. LOD and LOQ values for each parameter are given in Tables 4 and 5. pH and electroconductivity (EC) were also monitored in the samples using a multi-parameter measurement instrument (Thermo Orion 3 star). Ammonia and TKN were measured according to standard methods [30].

2.3. Ecotoxicity

Samples were submitted to acute ecotoxicity test using new born (<24-h old) daphnids exposed to samples for 24 and 48 h according to ISO6341 method [31]. Daphnids were obtained from the vivarium,

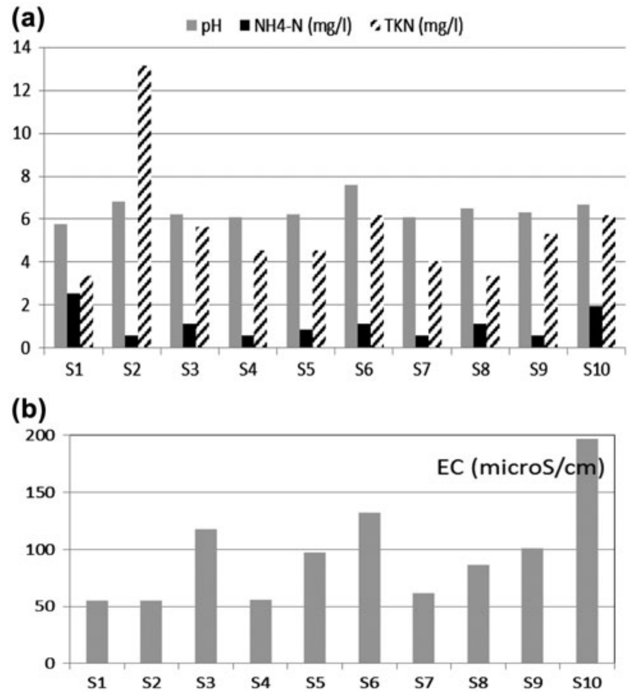

Fig. 2. Evolution of pH and nitrogen (a) and EC (b) in the rainwater samples collected from different origins (sample numbers are as indicated in Table 1).

where daphnids were grown in the laboratory at 16-h day light and 8-h dark periods, supplying a 3,000 lux illumination at 20°C room temperature and were fed with *Selenastrum capricornutum* (300,000 cell/ml) and baker's yeast (*Schizosaccharomyces cerevisiae*, 200,000 cell/ml). A minimum of 6 mg/l dissolved oxygen was supplied in the vivarium reactors by air pumps. Toxicity tests were performed quadruplicate using five daphnids in each test beaker with 50 ml effective volume of rainwater without dilution. pH of the rainwater samples was set to 8.0 according to standard test procedure. A negative control test was run in parallel to control the quality of test organisms (<5% of immobilization). The immobilization percentage was recorded as the ratio of the total number of immobile organisms divided by the total number of organisms tested [32].

2.4. Statistics

Data-sets were submitted to statistical evaluation by means of normal distribution and homogeneity. Kolmogorov–Smirnov test was used to test normal distribution. Accordingly, asymptotic sigma (two-tailed) value was set to be greater than 0.05. When test of homogeneity of variances was resulted greater than 0.05, Pearson and Spearman parametric test methods were used to find the correlation among the parameters. Furthermore, ANOVA test was used to evaluate independency among the samples, after which a relationship between pH (dependent variable) and NH_4^+ (independent variable), electrical conductivity (EC) (dependent variable) and calcium and magnesium concentrations (independent variable), and toxicity (dependent variable) and arsenic concentration (independent variable) was evaluated using one-way variation analysis ($p = 0.05$). Furthermore, a multivariate analysis was performed among pH, EC, other parameters, and toxicity using SPSS 15.0 data analysis program ($p = 0.025$).

Table 1
Chemical analyses in rainwater in various locations (EC is in μS/cm, others are in μeq/l)

Location	pH	EC	Ca^{+2}	Mg^{+2}	Na^+	K^+	NH_4^+	Reference
Jordan	6.91	95	165.3	93.12	130.6	85.21	75.36	[5]
Italy	5.18	–	70	77	252	17	25	[8]
Tirupati, India	–	–	150.7	55.51	33.08	33.89	20.37	[9]
New Mexico, USA	5.08	–	34.6	3.69	4.52	2.04	95	[10]
Melle, Belgium	5.19	–	26.87	9.25	36.98	1.96	65.65	[11]
Spain	6.4	–	57.5	9.8	22.3	4	22.9	[12]
Avignon, France	5.2	36.4	117.8	16.7	49.8	13.3	29.2	[13]
Kefalonia Island, Greece	8.31	103	189.6	12.34	260.9	61.38	0.56	[2]
Southern Taiwan	5.32–5.74	–	53.4	32.6	97.1	10.9	50.2	[14]
Newark, USA	4.6	–	6	3.3	10.9	1.3	24.4	[15]
Tokyo, Japan	4.52	–	24.9	11.5	37	2.9	40.4	[16]
Negev, Israel	7.24	390	1,215	188	406	19	47	[17]
Shanghai, China	4.49	–	204	29.6	50.1	14.9	80.7	[18]
Gangneung, South Korea	5.3	30	1.6	0.22	1.1	2.1	0.02	[19]
Istanbul, Turkey	6.96	24.2	32.9	9.17	61.74	3.85	–	[6]
Mugla, Turkey	6.9	–	174	–	17	3.5	30	[20]

Table 2
Concentrations of metals measured in rainwater in various locations all around the world (all units are in ppb)

Country	Fe	Al	Zn	Pb	Cu	Cd	Ni	Cr	Reference
Jordan	430	324	210	66	73	52	3.5	3.1	[5]
Mexico, USA	–	50.7	–	2.48	–	0.41	3.37	0.52	[10]
Kefalonia Island, Greece	11	–	10	<2.0	<2.5	0.05	<10	<1.3	[2]
Athens, Greece	4.38 ± 2.54	5.87 ± 8.67	33.46 ± 40.8	0.88 ± 1.0	15.41 ± 14.5	0.2 ± 0.14	4.14 ± 3.9	1.29 ± 0.9	[21]
Seoul, South Korea SK)									[22]
(wooden shingle roof)	154	227	135	10	34	–	–	–	
(concrete roof)	160	535	196	14	58	–	–	–	
(clay roof)	155	243	313	11	37	–	–	–	
(concrete roof)	302	622	428	12	59	–	–	–	
Gangneung, SK	–	100	60	20	35	ND	–	1	[19]
Jeju Island, Korea	30	–	–	–	–	–	–	–	[23]
Australia	2.66	–	98	73	2.45	0.01	ND	0.52	[24]
Monmouth County, USA	3,600	1,500	66.8	14.6	16.6	–	–	15.2	[25]
Turkey									
Mersin	743.2 ± 115	484.5 ± 49.5	50.2 ± 6.06	11.36 ± 0.8	3.94 ± 0.3	0.81 ± 0.09	7.23 ± 0.5	5.72 ± 0.4	[26]
Ankara	750 ± 2,370	980 ± 2,900	0.03 ± 0.03	19.1 ± 37.6	6.1 ± 9.5	9.5 ± 12.0	3.37 ± 0.6	0.52 ± 0.0	[27]
Antalya	–	580 ± 758	137 ± 510	10 ± 14	5.9 ± 6.4	4.9 ± 6.4	24 ± 26	9.0 ± 11.6	[28]
İstanbul	2,750	7,660	–	1.47	1,450	–	0.77	0.58	[6]

3. Results and discussion

3.1. Physicochemical parameters

The origin of the samples and the raining period affected significantly the values of pH, NH_4^+, TKN, and EC as seen in Fig. 2(a) and (b), respectively. The values of pH varied from 5.8 to 7.6 among the samples. Accordingly, pH values measured in this study were observed to be in the interval of the literature studies performed in different locations as seen in Table 1. Except for sample S1, the difference between NH_4^+ and TKN parameters was significant in the samples. This difference indicated a strong organic nitrogen contribution in the samples. A peak value was observed in the S2 sample which was a SC sample collected in a relatively dry season when a high quantity of organics, probably originated from microbiological pollution contribution, could be present [33]. The concentration of NH_4^+ was relatively high in all samples compared to the literature values shown in Table 1.

EC varied from 54.9 to 197.1 μS/cm among the samples as seen in Fig. 2(b). Measured EC values were found to be close to the results of the previous studies performed worldwide shown in Table 1. According to the data shown in Table 4, calcium and magnesium concentrations were measured varying in a wide range. However, those results were seen to be in the interval of the previous literature findings shown in Table 1 except for sample 10 of which the concentrations were close to the study performed in Israel [17]. The high concentrations of calcium and magnesium parameters are agreed by the higher measured value of EC in S10 sample, which was collected from SC close to the road of the residential area.

3.2. Metals

Concentrations of the metals were measured varying from one location to another as seen in Table 5. All concentrations were found to be significant considering the LOD values of each parameter. The varying metal concentrations of the samples are attributed to the atmospheric deposition, meteorological events by means of intensive rains and winds as well as traffic load and sample collection medium [5,6]. These facts are clearly seen in Table 5 that the concentrations of the metals varied in the same group of samples (RC, NSC and SC) versus sampling dates on which meteorological and environmental (on-heating system) conditions.

Some of the metals (Pb, Zn, and Ni) measured were seen to be in the range of the other studies performed in the other cities worldwide [10,21]. If the results of the samples collected from roof are compared to the study performed in South Korea [22], except for the lead parameter, all other metals were measured less in our samples, even from the same type of roof material (Clay). When the results of previous studies conducted in other cities in Turkey (Table 2) are considered, it can be emphasized that the values of the parameters were found to be at the same range except copper metal measured in Istanbul [6] and iron measured in Mersin and Ankara cities, respectively [26,27]. Significant higher levels of some metals, such as iron metal in samples S6 and S10, are to be associated to the origin of the samples (SC).

3.3. Ecotoxicity

Because standard deviations were less than 5% in both rainwater samples and negative control tests,

Table 3
Coordinates of the rainwater sampling points in this study

Sample no	Date	Time	Origin*	Coordinates
S1	18.05.2012	19:00	NSC	41 04 00.56 N–28 15 20.60 E
S2	18.05.2012	19:00	Roof contact	41 04 02.30 N–28 15 22.20 E
S3	29.10.2012	23:00	NSC	41 04 02.30 N–28 15 22.20 E
S4	21.09.2012	08:00	NSC	41 04 00.56 N–28 15 20.60 E
S5	01.12.2012	12:00	Roof contact	41 04 01.71 N–28 15 22.17 E
S6	01.12.2012	11:30	SC	41 04 37.12 N–28 14 19.96 E
S7	01.12.2012	15:00	NSC	41 04 02.30 N–28 15 22.20 E
S8	01.12.2012	14:30	SC	41 04 02.30 N–28 15 22.20 E
S9	01.12.2012	14:00	Roof contact	41 04 21.79 N–28 15 03.11 E
S10	01.12.2012	10:15	SC	41 04 49.30 N–28 13 10.61 E

SC: ground surface contact; NSC: non-surface contact.

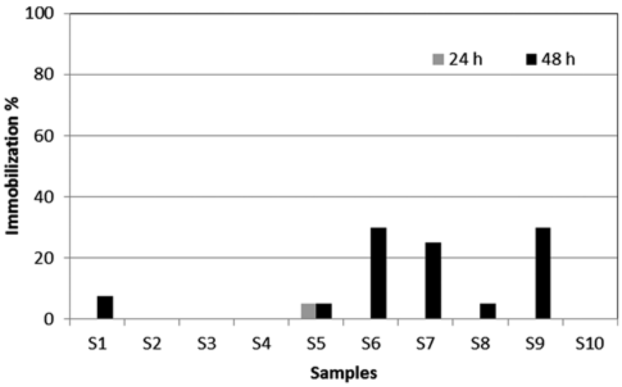

Fig. 3. Toxicity evolution in the rainwater samples tested (standard errors of quadruplicate experiments were less than 5%).

Table 4
Calcium and magnesium analysis in the rainwater samples analyzed in this study[a,b]

Sample no	Mg^{2+} (ppb)	Ca^{2+} (ppb)
S1	881.609	2,647.61
S2	430.349	2,841.36
S3	1,267.21	1,101.42
S4	506.297	414.170
S5	2,035.81	1,202.60
S6	1,970.8	2,838.46
S7	792.074	515.728
S8	967.903	1,338.83
S9	1,072.94	1,877.93
S10	5,517.33	2,682.95

[a] Standard deviation of six measurements for each sample was less than 3%.

[b] LOD and LOQ values were 8.316 and 25 ppb for magnesium and 40 and 100 ppb for calcium, respectively.

average acute toxicity results of quadruplicate tests are shown in Fig. 3. As seen in Fig. 3, none of the samples resulted in toxicity to *D. magna* after 24-h exposure time (except S5), whereas some incremental increases in toxicity were noted in some of the samples (S1 (15%), S6 (30%), S7 (25%), and S9 (30%)) after 48 h of exposure. Toxicity seemed to be independent from the origin of the samples (S6, S7, and S9 samples were SC, NSC, and RC collected samples, respectively) (Fig. 3). Toxicity was observed to be independent from NH_4^+ parameter since this parameter was measured at low levels (<1 mg/L), to cause significant toxicity to *Daphnia magna* [32].

3.4. Statistics

Asymptotic sigma value was proved to be greater than 0.05 which indicated that the data were normally distributed. The correlation analyses' results are shown in Table 6. When whole samples were considered, high and positive correlations were only observed between EC and magnesium parameters. When the data obtained were submitted to variance analysis using SPSS 15.0, it was observed that the results measured in whole samples were significantly independent from each other (0<0.05) with the exception observed between EC and magnesium parameters (Table 7). However, when only RC samples were evaluated, a full (100%) dependency was noted between pH and NH_4^+, EC and magnesium and between acute toxicity (48 h) ($p < 0.025$). High concentrations of lead parameter in samples S6, S7, and S9 were noted to relate to the higher acute toxicity observed as seen in Fig. 3. However, statistically there was no evidence of that kind of correlation while acute toxicity was found to be dependent on arsenic

Table 5
Mean concentrations of metals in the collected rainwater samples in this study (all units in ppb)*

Sample no.	As	B	Cd	Co	Cu	Mn	Mo	Ni	Cr	Pb	Zn	Fe
S1	1.772	0	1.060	0	5.920	6.326	0.411	4.520	2.694	0.625	31.871	16.547
S2	2.003	0	0.587	0	3.387	0.179	0.467	1.481	2.733	1.954	4.465	9.732
S3	2.788	0	3.251	1.453	3.341	9.881	2.631	2.399	2.396	0	23.665	11.866
S4	2.723	0	3.223	0.583	3.967	5.363	2.326	1.390	2.315	9.717	33.528	10.415
S5	2.246	0	5.274	1.285	6.367	4.365	0.055	3.643	2.735	12.459	17.088	18.953
S6	3.043	0	5.186	1.959	6.623	5.598	0.293	6.724	3.112	18.903	13.464	110.00
S7	2.739	0	5.205	0	5.482	5.583	0	4.603	2.691	15.324	16.896	9.658
S8	3.146	0	4.792	1.040	5.143	4.064	0	3.315	2.710	10.181	10.201	22.079
S9	2.646	0	5.090	1.496	5.983	4.252	0	3.387	2.513	31.428	9.091	17.435
S10	3.035	0	4.827	2.019	7.957	5.686	0	6.310	2.741	19.456	9.897	117.99
LOD (ppb)	1.38	1.61	0.5297	0.5713	2.009	0.1779	0.05	1.098	2.31	0.612	1.566	1.056
LOQ (ppb)	10	10	10	10	10	10	25	10	10	25	10	10

*Standard deviation of six measurements for each sample was less than 3% for all parameters.

Table 6
Statistical correlation results among the parameters in the rainwater samples tested in this study

Parameters	Correlation method	Regression coefficient	Significance level
pH–NH_4^+	Pearson/Spearmans	−0.138/0.032	0.01
EC–Mg^{2+}	**Pearson/Spearmans**	**0.913/0.891**	**0.01**
EC–Ca^{2+}	Pearson/Spearmans	0.355/0.103	0.01
Acute ecotoxicity (48 h)–Arsenic	Pearson/Spearmans	−0.279/−0.290	0.01

Note: Values in bold character shows a strong correlation between the tested parameters.

Table 7
Statistical analyses to determine independency levels among tested parameters

Related parameters	Sample type	Dependency level (%)	Significance level (Confidence level 95%)
pH–NH_4^+	Whole samples	1.9	(p 0.70 > 0.025)
	NSC	6.67	(p 0.183 > 0.025)
	Roof contact	100	(***p* 0 < 0.025**)
	SC	3.5	(p 0.812 > 0.025)
EC–Mg^{2+}	Whole samples	83.5	(***p* 0.02 < 0.025**)
	NSC	90.7	(p 0.305 > 0.025)
	Roof	100	(***p* 0 < 0.025**)
	SC	99	(p 0.028 > 0.025)
Toxicity-Arsenic	Whole samples	7.8	(p 0.4 > 0.025)
	Roof	100	(***p* 0 < 0.025**)

Note: NSC and SC are to be as indicated in Table 3; values in bold character shows a strong correlation between the tested parameters.

parameters, when RC samples were only considered (Table 7). Lack of any correlation among other samples can be due to the presence of other pollutants which may interfere with the toxicity results [7,32].

4. Conclusion

The results of this survey showed that rainwater quality varies depending on the sampling origin and meteorological events.

None of the samples resulted in severe toxicity to *Daphnia magna* after 24-h exposure, while three samples (S6, S7, and S9) exhibited moderate toxicity (20–30% immobilization) after 48-h exposure. These low acute toxicity results seemed to be confirmed by longer exposure time-based biotests since metals were observed at meaningful ranges to be present in each sample.

Furthermore, it is recommended to monitor rainwater quality for the content of other chemicals and microbiological parameters to control the risk in the aquatic environment.

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