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Research Paper

Appraisal of rainwater characteristics in and around historical monuments of 16-18th century, Haryana, India

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Abstract: Monuments of Haryana date back to around 16-18th century are large masonry structures built using burnt-clay bricks (*lakhauri*) and lime-crushed bricks aggregate (*surkhi*) mortars. These monumental buildings have great importance due to their identity of transferring old era information to the present and future generations. In recent years, the alterations in natural environment of Haryana state negatively affected the monumental buildings. Climate related deteriorations on the monumental buildings resulted from temperature differences, water movement in the buildings due to capillary action, abrasive effects of rain water, salt and chemicals involved in water, particles carried by wind and air pollution etc. Among these, rainwater serves as a chemical agent for many geological processes and geo-chemical changes of the environment. The rainwater analyses help to reveal the chemical state of the environment in which historical structure is situated. The rainwater attributes depend on the concentrations of anionic as well as the cationic species. The ionic concentrations in rainwater of Haryana showed following order for anions SO_4^{2-} > NO_3^- > Cl^- > HCO_3^- > F^- while cations showed Ca^{2+} > Mg^{2+} > NH_4^+ > K^+ ≥ Na^+. The sources of these ions were soluble components of the soil-derived and anthropogenic aerosols dissolve in the cloud column during cloud formation and subsequent rain formation. Ionic constituents influence the pH of rainwater.

Keywords: *Climate, Rain fall, Ionic strength, pH.*

INTRODUCTION:

The historical monuments and cultural values are an expression of accumulation

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and a bridge from past to present. The unchecked infrastructure developments, industrial pollution, increased tourism and population explosion accelerated the rate of decay of heritage components at an alarming rate and exert catastrophic effects on cultural heritage at global level (Camuffo, 1986, 1991).

The damaging effects of extreme weather conditions are obvious, but constant damage caused by everyday weather conditions which has a cumulative and synergistic effect and in the long run it is more destructive (Koestler, *et. al*., 1994). Water in any of its forms accelerates the decay due to the porous nature of the used historic materials (Viles, 2011). The salts and the pollutants present in water react with the materials and cause disintegration in most of the cases with their undesirable generated products (Hall and Hoff, 2002, Camuffo, 1998).

The heavy rain falls and humidity levels deteriorated historic structures and also accelerated the natural aging (Awan, 1993, 2008, Rehman, 2011). The massive masonry core of the buildings retain water and carried salts driven by rains for longer period, developing into salt crystallizations within or on the surface (Viles, 2011, Torres and Freitas, 2003). The water penetration in the historic structures can be seen as efflorescence, cracking, cavities and leakage with mold growths.

The importance of monuments of Haryana lies in the fact that it acts as witnessed of the majestic drama of the rise and fall of the Mughal, the Sikhs and the colonial rulers. The monuments of Haryana are the masterpiece of Mughal architecture with all its creative detail, beautiful surface renderings and unique construction techniques. These beautiful monuments reflect the glorious past of the area, which needs to be protected, preserved and passed on to future generations. The most of the monuments of Haryana are open historical structures. Permanent wetting of the external exposed surfaces for three to four months during the monsoon and improper rainwater disposal systems, increases penetration of water and moisture inside the masonry units make it more vulnerable to deterioration.

MATERIAL AND METHOD:

Study area

Haryana is situated between 27^0 39’ to 30^0 35’N latitude and 74^0 28’to 77^0 36’E longitude, covering an area of 44212 Km^2 i.e. 1.40 % of the total area of the country. The climate of the state is subtropical, semi-arid to sub-humid, continental and monsoon type with very hot summer and cold winter except during monsoon season when moist air of oceanic origin penetrates into the state. The south west monsoon sets in from last week of June and withdraws in end of September, contributed about 80 % of annual rainfall. Rest 20 % rainfall is received during non-monsoon period in the wake of western disturbances and thunder storms. The average rainfall of the state is 560 mm which varies from less than 300 mm in south-western parts to over 1000 mm in the hilly tracks of Shivalik hills.

The temperature varies between 30 and 48 °C in summer and between 5 and 25 °C during winter. Keeping in view of glorious history, architecture, strength of

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monuments of Haryana and present day natural and anthropogenic changes, six monuments of Haryana were taken for present study as summarized in table-1.

Table-1: General description of study area

S.No	Name of Site	Location of Site	Pollution Level	Masonry Units (Major)	Assigned Code for Study
1	Sheikh Chilli's Tomb, Thanesar, Kurukshetra	Urban	High	Bricks/Marble	SCTTK
2	Cantonment Church Tower, Karnal	Urban	Moderate	Brick/Lime Plaster	CCTK
3	Mughal Sarai gateway, Gharaunda, (Karnal)	Semi-Urban	High	Brick	MSGGK
4	Kabuli Bagh Mosque, Panipat	Urban	High	Brick/lime Plaster	KBMP
5	Khwaza Khizr's Tomb, Sonepat	Urban	Moderate	Brick/Kankar Stone	KKTS
6	Mound of Agroha, Agroha (Hissar)	Semi-Urban	Moderate	Brick	MAAH

Rain water sampling

The monsoon month's rainwater samples for three consecutive years from 2016 to 2018 were collected from each study area at different locations at the roof of the monument as the rainfall amount and frequency of rain during these months was maximum as compare to pre-monsoon and post-monsoon period. Rainwater collector consists of a polypropylene funnel with polypropylene bottle. The samples were filtered and only aqueous phase was analysed. Major anions (SO_4^{2-}, NO_3^-, Cl^-, HCO_3^- and F^-) and cations (Ca^{2+}, Mg^{2+}, NH_4^+ , K^+ and Na^+) along with pH and EC were analysed in laboratory. The instrument used for ionic analysis was Ion Chromatograph 930 compact IC Flex, for pH and EC, Digital pH meter (Systronics 361) and Digital conductivity meter (Systronics 306) were used, respectively. The concentration of bicarbonate was determined by titration method. The ionic concentrations of major cations and anions were obtained in mg/l which is converted in to µeq/l by using the formula: *µeq/l = (mg/l x Valence x1000)/ Wt*, while value of EC was obtained in µS/cm and reported as such.

RESULT AND DISCUSSION:

Mean concentrations of different ionic components in rainwater samples are shown in tab2, 3 and 4. The general order for anions are $SO_4^{2-} > NO_3^- > Cl^- > HCO_3^- > F^-$ while cations followed the order $Ca^{2+} > Mg^{2+} > NH_4^+ > K^+ \geq Na^+$. The

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reported average cations concentration of Ca^{2+}, Mg^{2+} and K^{+} varied in the range between 133.4-161.7 µeq/l, 63.9-71.9 µeq/l and 20.8-25.6 µeq/l respectively. The high concentration of Ca^{2+}, Mg^{2+} and K^{+} in rainwater attributed to high levels of total suspended particles and soil transported from neighboring state Rajasthan, stronger winds entraining large quantities of dry soil in summer and high washout rate release it during monsoon period.

The average concentrations of NH_4^+ during monsoon season recorded between 57.3 to 68.8 µeq/l resulted from ammonia generation by action of bacteria on nitrogen compound in the soil from industrial effluent. The average cations concentration of SO_4^{2-} and NO_3^- are varied in the range between 52.2-85.8 µeq/l and 49.4-65.5 µeq/l respectively. The high concentration of SO_4^{2-} and NO_3^- in Panipat and Gharaunda can be linked to the emission of the precursor gas from the cluster of large to small scale industries, coal fired power plants located in the vicinity of monuments. The major sources for chloride and fluoride ions within rain sample besides any natural source are could be due to plastic burning, brick kilns around the area and coal fired power plants as observed during field survey.

The average pH of rain water varied from 5.99 to 6.03 which may be due to the high loading of particulates in the atmosphere commonly found in these areas. Alkaline nature of rainwater was reported in northern part of India due to heavy loading of particulates rich in carbonates and bicarbonates of calcium, which neutralized the acidity (Kulshrestha, *et.al.*, 1996). The northwest part of India also exhibits higher pH values than rest of the country. This may be due to the incursion of sand/dust particles from the adjacent Thar Desert of Rajasthan (Mohan and Kumar, 1998). The suspended particulate matter rich in carbonates and bicarbonates of calcium buffered the acidity of rainwater generated by the conventional acids such as H_2SO_4 (Kulshrestha, et. al., 1996)**.** The NH_3 in the form of $NH4^+$ also acted as an important buffering agent next to Ca^{2+} salts.

Despite this, the acidity of rainwater could not be fully neutralized and rainwater shows the acidic tendency due to high concentration of SO_4^{2-} and NO_3^- released from the anthropogenic sources. This revealed the similarity to those reported at other Indian sites such as Rampur, Agra (Singh, et. al., 2001), Hyderabad (Kulshrestha, et. al., 2003) and Lucknow (Khare et. al., 2004). . In all these results, crustal Ca^{2+} was present in the highest concentration. The dust particles rich in calcium carbonate/bicarbonate acted as the major buffering agent for acidity generated by SO_4^{2-} and NO^{3-} in India. The pH of rainwater is generally affected by acidic (SO_4^{2-} and NO_3^-) and alkaline (Ca^{2+} and NH_4^+) components, which are dominated in the rainwater of study area.

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Table 2. Mean value of pH, EC and concentration of ionic components of rainwater samples (n=5) collected during monsoon season at Thanesar (Kurukshetra), Karnal and Gharaunda.

SCTTK	Cl^-	SO_4^{2-}	NO_3^-	F^-	NH_4^+	Na^+	K^+	Ca^{2+}	Mg^{2+}	HCO_3^-	pH	EC
2016												
Mean	26.5	75.1	47.5	2.5	63.0	20.0	20.6	142.5	70.1	9.4	6.0	61.7
S.D	2.3	3.79	2.84	0.71	4.81	1.25	1.76	4.19	4.25	0.36	0.02	2.21
2017												
Mean	30.1	77.5	51.2	2.6	62.6	20.3	20.4	144.6	67.1	9.5	6.0	59.4
S.D	1.83	6.83	2.82	0.52	4.23	1.98	2.48	6.42	2.34	0.48	0.02	3.62
2018												
Mean	30.0	81.6	49.6	2.8	62.6	20.2	21.4	155.8	69.8	9.8	6.0	65.7
S.D	1.5	5.8	1.6	0.5	3.7	2.5	3.0	7.2	4.3	0.8	0.0	8.5
CCTK												
2016												
Mean	26.7	66.7	52.2	3.1	78.0	20.4	23.7	147.9	72.1	9.4	6.0	71.6
S.D	2.5	7.5	4.3	1.0	14.0	2.4	4.2	9.1	10.6	0.3	0.0	14.4
2017												
Mean	31.1	75.5	63.2	2.7	65.2	21.5	22.0	144.0	68.9	10.0	6.0	62.7
S.D	2.4	4.9	22.4	0.3	4.2	2.2	8.1	34.3	6.5	0.8	0.0	2.0
2018												
Mean	27.2	85.4	53.2	3.1	63.3	20.6	27.0	170.8	70.9	9.8	6.0	68.3
S.D	2.2	4.4	8.1	0.5	5.0	1.6	5.8	19.0	5.6	0.9	0.0	13.1
MSGGK												
2016												
Mean	26.5	75.6	48.7	2.7	66.1	20.7	24.0	150.8	74.1	8.9	6.0	66.3
S.D	2.3	5.5	4.4	0.8	13.1	2.6	3.3	10.0	3.1	0.4	0.0	6.2
2017												
Mean	29.2	74.2	53.9	3.1	67.9	22.7	25.2	145.6	69.4	9.4	6.0	58.6
S.D	0.8	5.1	5.9	0.6	7.6	1.9	3.6	22.9	5.6	0.3	0.0	1.9
2018												
Mean	26.5	68.7	51.9	3.6	61.4	21.6	25.4	170.7	66.1	8.9	6.0	66.0
S.D	2.4	5.6	7.9	0.9	2.9	1.5	5.1	19.9	6.8	0.4	0.0	6.3

(Concentration in µeq/l and EC is in µS/cm)

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Table- 3. Mean value of pH, EC and concentration of ionic components of rainwater samples (n=5) collected during monsoon season at Panipat, Sonepat and Agroha (Hissar).

KMBP	Cl^-	SO_4^{2-}	NO_3^-	F^-	NH_4^+	Na^+	K^+	Ca^{2+}	Mg^{2+}	HCO_3^-	pH	EC
2016												
Mean	27.1	79.5	52.3	3.7	64.0	23.1	24.5	151.6	74.5	8.7	6.0	64.1
S.D	2.1	10.05	12.08	0.62	9.25	3.53	6.69	17.22	1.84	0.63	0.03	6.80
2017												
Mean	26.1	90.2	68.6	4.1	68.9	21.6	25.5	166.7	72.0	9.2	6.0	59.4
S.D	1.90	9.93	6.27	0.96	5.04	1.75	2.90	22.39	2.83	0.73	0.04	4.44
2018												
Mean	27.2	87.6	75.6	4.0	63.2	23.5	26.7	166.9	69.2	9.1	6.0	72.2
S.D	2.0	19.5	15.1	0.5	11.8	2.5	3.3	10.5	0.7	0.1	0.0	13.2
KKTS												
2016												
Mean	25.5	67.2	54.0	2.8	59.6	20.8	22.9	134.6	69.1	9.1	6.0	62.7
S.D	3.6	4.5	3.3	0.3	1.7	5.4	3.5	3.2	1.6	0.1	0.0	4.6
2017												
Mean	27.6	69.6	59.0	2.9	62.9	23.3	24.4	137.8	69.7	9.3	6.0	62.7
S.D	1.8	4.0	2.2	0.4	3.1	1.0	2.8	3.7	0.5	0.4	0.0	2.0
2018												
Mean	25.6	66.4	64.1	2.8	62.1	23.5	24.7	139.7	69.2	9.1	6.0	63.3
S.D	5.1	2.3	8.0	0.1	9.2	1.3	1.5	1.6	0.7	0.1	0.0	5.1
MAAH												
2016												
Mean	29.7	50.4	50.4	2.4	56.1	22.7	21.6	131.6	61.9	9.1	6.0	57.8
S.D	0.6	1.9	5.1	0.2	2.8	2.1	3.3	2.7	5.4	0.1	0.0	1.7
2017												
Mean	29.7	53.3	55.7	2.5	59.0	23.9	23.3	134.3	65.3	9.2	6.0	58.6
S.D	2.3	1.4	3.2	0.3	1.6	2.8	2.3	7.1	1.9	0.4	0.0	1.7
2018												
Mean	30.0	52.9	52.2	2.4	56.7	23.3	22.7	134.2	64.4	9.1	6.0	58.8
S.D	1.3	2.9	2.0	0.3	3.9	0.6	2.5	3.2	5.4	0.6	0.0	5.7

(Concentration in μeq/l and EC is in μS/cm)

Table- 4. Mean value of pH, EC and concentration ionic components of rainwater samples from 2016-2018

	Cl^-	SO_4^{2-}	NO_3^-	F^-	NH_4^+	Na^+	K^+	Ca^{2+}	Mg^{2+}	HCO_3^-	pH	EC
SCTTK	28.9	78.1	49.4	2.7	62.7	20.2	20.8	147.7	69.0	9.5	6.02	62.2
CCTK	28.3	75.9	56.2	3.0	68.8	20.8	24.2	154.2	70.7	9.7	6.03	67.5
MSGGK	27.4	72.8	51.5	3.1	65.1	21.7	24.9	155.7	69.9	9.1	6.00	63.6
KMBP	21.2	85.8	65.5	3.9	65.3	22.7	25.6	161.7	71.9	9.0	5.99	65.3
KKTS	26.2	67.7	59.0	2.8	61.5	22.5	24.0	137.4	69.3	9.2	6.00	62.3
MAAH	29.8	52.2	52.8	2.5	57.3	23.3	22.6	133.4	63.9	9.2	6.00	58.4

(Concentration in μeq/l and EC is in μS/cm)

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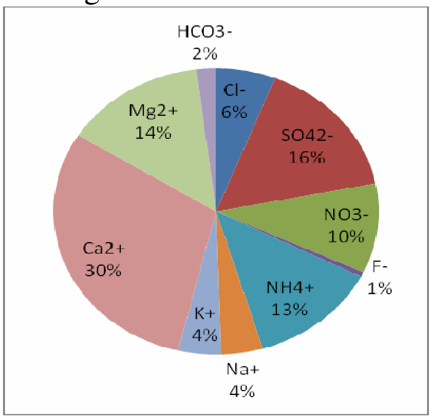
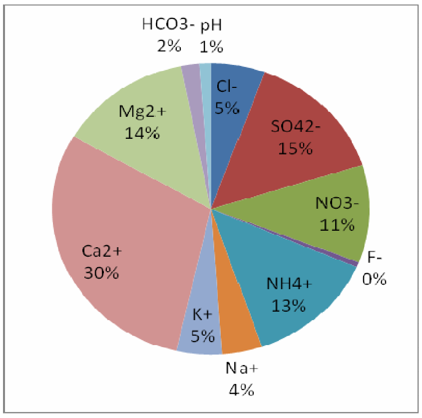
The percentage contributions of each ionic species to the total ionic concentration for different study area are shown in figure- 1. Ca^{2+} was the dominant cation of all the ions contributing 29 to 31 % to the total ionic concentrations. Next to Ca^{2+}, SO_4^{2-} was the dominant anion contributing 12 to 16 % to the total ionic concentration. The other important cations were Mg^{2+} and NH_4^+ contributing 14% and 12-13% to the total ionic concentration, respectively. The contribution of Na^+ and, K^+ to the total ionic was < 10%. Cl^- and NO_3^- contributed 4-7 % and 10-12 % to the total ionic concentration, respectively. The contribution of HCO_3^- was 2 % to the total ionic concentration and contribution of F^- was negligible to the total ionic concentration.



SCTTK



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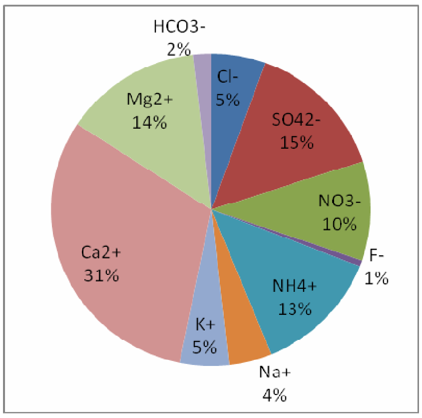
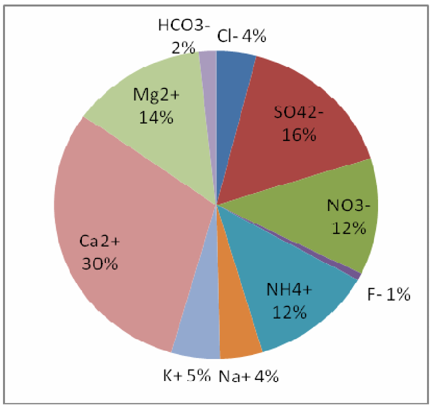


MSGGK



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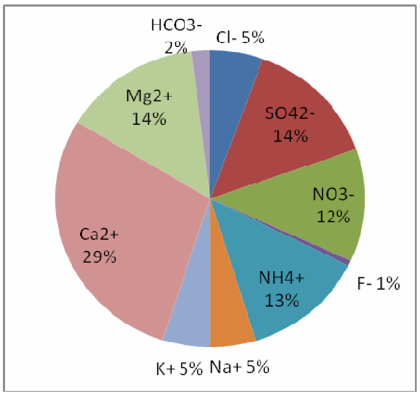
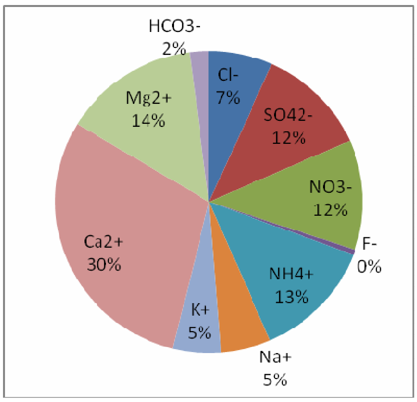


KKTS



MAAH

Figure -1. Each ion contribution at different study area

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SO_4^{2-}/NO_3^- Ratio

The results of SO_4^{2-}/NO_3^- ratio of rainwater samples of all study areas are given in table 5. The SO_4^{2-}/NO_3^- ratio is used to find the contribution of anthropogenic and relative contributions of mobile and stationary sources. The value of this ratio was varied between 0.99- 1.58 from Thanesar (Kurukshetra) to Agroha (Hissar). Approximately same value shows that the same types of factors are within the vicinity of the monuments. Relative contribution of SO_4^{2-} and NO_3^- towards the acidification was computed by using the ratio (SO_4^{2-}/ (SO_4^{2-} +NO_3^-) and (NO_3^-/(SO_4^{2-} + NO_3^-) respectively. The contribution of H_2SO_4 in rain samples of different sites was found between 50 - 61% with highest value of 61 % for Thanesar while other sites i.e. Karnal, Gharaunda, Panipat, Sonepat and Agroha have similar values in between 50-59 %. The contribution of HNO_3 was 39 - 50% with highest value of 50 % for Agroha.

Table -5. SO_4^{2-}/NO_3^- ratio of rainwater samples at different study area

	SO_4^{2-}	NO_3^-	SO_4^{2-} / NO_3^-	SO_4^{2-} / SO_4^{2-} + NO_3^-	NO_3^- / SO_4^{2-} + NO_3^-
SCTTK	78.1	49.4	1.58	0.61	0.39
CCTK	75.9	56.2	1.36	0.57	0.43
MSGGK	72.8	51.5	1.42	0.59	0.41
KMBP	85.8	65.5	1.33	0.57	0.43
KKTS	67.7	59.0	1.15	0.53	0.47
MAAH	52.2	52.8	0.99	0.50	0.50

The contribution of sulphate and nitrate is different due to difference in relative contribution of anthropogenic sources. Industries, vehicular emission and coal burnings in power plants located in the vicinity of monuments are the contributors of SO_x. While potential sources of NO_x in this region are vehicular emissions, industries, biomass burnings and agriculture field.

Marine contribution

The equivalent ratio of major ionic components like Ca^{2+}, Mg^{2+}, K^+, SO_4^{2-} and Cl^- are shown in table 6 with respect to sodium by considering the fact that sodium is completely from marine origin. Ratios of major sea salt components are found to be constant in the sea water. The non-sea salt or sea salt faction of any particular component (say X) is calculated by-

$$NSSF_X = [X_{rain}]-[\{Na_{rain}\}*\{X/Na\}_{sea\ water}]$$

$$SSFx = [Na_{rain}]\ [X/Na]_{seawater}$$

The % SSF and the NSSF of the any components (say X) is calculated by

$$\%SSF = 100\ (Na)\ (X/Na)\ sea\ /X$$

$$\%NSSF = 100-\%SSF$$

The enrichment factor (EF) = $(X/Na)_{rainwater}$ / $(X/Na)_{seawater}$

The ratio vale of Cl^- /Na^+ was found to be near to the sea water. The correlation value as shown in table 6 indicate strong relation between Cl^- and Na^+ which indicate influx of marine spray from Bay of Bengal during south west monsoon and soil dust in this region.

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Table- 6. Equivalent concentration ratios of ionic components at different study area.

	Cl-/Na+	Mg2+/Na+	K+/Na+	Ca2+/Na+	SO42-/Na+
SCTTK	1.43	3.42	1.03	7.31	3.87
SW	1.160	0.227	0.220	0.044	0.125
SSF (%)	81	7	21	1	3
NSSF (%)	19	93	79	99	97
EF	1.23	15.05	4.68	166.13	30.92
CCTK	1.36	3.40	1.17	7.42	3.65
SW	1.16	0.227	0.22	0.044	0.125
SSF (%)	85	7	19	1	3
NSSF (%)	15	93	81	99	97
EF	1.17	14.96	5.30	168.52	29.18
MSGGK	1.26	3.22	1.15	7.17	3.36
SW	1.16	0.227	0.22	0.044	0.125
SSF (%)	92	7	19	1	4
NSSF (%)	8	93	81	99	96
EF	1.09	14.19	5.21	163.06	26.85
KMBP	1.18	3.17	1.13	7.12	3.78
SW	1.16	0.227	0.22	0.044	0.125
SSF (%)	98	7	20	1	3
NSSF (%)	2	93	80	99	97
EF	1.02	13.95	5.12	161.90	30.23
KKTS	1.17	3.08	1.07	6.11	3.01
SW	1.16	0.227	0.22	0.044	0.125
SSF (%)	99	7	21	1	4
NSSF (%)	1	93	79	99	96
EF	1.01	13.58	4.85	138.77	24.07
MAAH	1.28	2.74	0.97	5.72	2.24
SW	1.16	0.227	0.22	0.044	0.125
SSF(%)	91	8	23	1	6
NSSF (%)	9	92	77	99	94
EF	1.10	12.08	4.40	130.08	17.92

CONCLUSION:

The present study reveals that the rainwater carries the impression of anthropogenic activities which affect the historical monuments in the Haryana State. The quality of rainwater depends upon the overall concentrations of anionic as well as the cationic species. The relative contribution of primary aerosols (Ca^{2+}, K^{+}, Mg^{2+}) and secondary aerosols (SO_4^{2-} and NO^{3-}) as well as NH_4^{+} in rainwater influences its pH.

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