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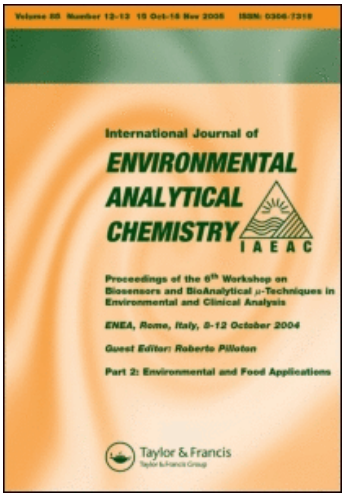

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Monitoring of Heavy Metals in Urban Snow as Indicator of Atmosphere Pollution

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MONITORING OF HEAVY METALS IN URBAN SNOW AS INDICATOR OF ATMOSPHERE POLLUTION

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Snow samples from six sites located in and around urban areas in Sivas city, Turkey, were collected in the winter of 2000. After ultrasonic leaching of the melted snow solutions, they were analysed for heavy metals (Mn, Zn, Pb, Cu, Ni and Cd) by atomic absorption spectrometry (AAS). The results of the analyses were used to determine major sources and magnitude of heavy metal pollution. Besides, it was compared with the heavy metal concentrations in the urban snow with those of background areas to see whether they could be used as an effective indicator of urban atmosphere pollution.

High concentrations of heavy metals were detected in snow samples collected in close proximity to emission sources such as motor-vehicle emissions, combustion of coal and fuel-oil, industrial area. The heavy metal levels in urban snow were several times higher than the background levels. After heavy metal pollution factors of each site were determined, their possible pollutant sources were detected. Our findings show that, data on the heavy metal concentrations in snow are a reliable guide to the degree of air pollution, and can be used as a simple and effective indicator of urban atmosphere pollution. The precision of method varied between 1.22 and 5.62% ($n = 54$) as average relative standard deviation % (RSD %) depended on the analyte and nature of the sample.

Keywords: Atmosphere pollution; Heavy metals; Snow; Ultrasonic leaching; AAS; Sivas-Turkey

INTRODUCTION

Heavy metals constitute a class of environmental pollutants, which require particular attention in the management of the environment due to their toxicity to man and the general ecosystem. Particularly, anthropogenic emissions have brought about an appreciable rise in the airborne heavy metal concentrations. Elevated levels of heavy metals have been observed in soil, vegetation, snow and ambient air in urban areas due to burning of fossil fuel, automobile exhaust and industrial emissions. Many detailed studies have been carried out in order to quantify man-made emissions[1–15]. The snow (chemistry) reflects the chemical composition of its parent aerosol, dry fallout and surface adsorbed material. Therefore, it is extensively used to monitor and characterize atmospheric depositions in the high latitude areas[16–18]. Murozumi *et al.*[19] proved that lead pollution in the atmosphere has spread right around the world by

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analysing the geochemistry of polar snow. Labarre *et al.*[20] and Grandstaf and Myer[21] have shown that lead aerosols are introduced into the atmosphere by the combustion of lead-containing fuels in the internal combustion engines of automobiles, resulting in lead pollution of snow.

Environmental pollutants are possible hazard to human health, plants and other constituents[22, 23]. The increase in the air pollution may result in massive deaths, and acid rains which destroy the historical and cultural pieces, plant and aquatic life and other various materials[23–24]. It is known that heavy metal pollution such as Pb, Ni and Cd is common in urban areas rather than rural areas due to the presence of industrial resources[25]. In general, toxic heavy metals such as Pb, Cd, Ni, Zn, Mn, Cu does not exist in snow. Even trace amounts of these heavy metals indicate possible contamination.

In this study, the melted snow solutions were leached under ultrasonic effect for high recoveries of elements from atmospheric particulate in the samples. It has been reported that ultrasonic leaching method (ULM) gives high recoveries of organics from sediment[26], fly ash[27], biological materials[28], and elements from atmospheric particulate[8, 29] and sediments[30, 31] in a much shorter time than is required for other extraction procedures.

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The essential characteristics of atmosphere pollution in an area of heavy snowfall were investigated in detail. It was aimed to monitor heavy metals in snow as an indicator of urban atmosphere pollution in Sivas city, Turkey, and this kind of study wasn't realized in the city before. Besides, it is discussed whether the concentrations of Pb, Mn, Zn, Ni, Cu and Cd in snow are suitable for use as an index of atmosphere pollution. In this paper, the Pb, Mn, Zn, Ni, Cu and Cd contents of the snow samples collected from six sites in and around urban areas of Sivas city, Turkey, during the period of January–March 2000, have been determined by AAS.

Heavy metal pollution is expressed as pollution factor (PF) which is an important parameter of anthropogenic pollution[4, 8]. PF can be described as the ratio of the determined pollution to base level or background pollution. The pollution of site 6 which was thought to be unaffected by anthropogenic sources has been chosen as the base level.

It will be possible to take necessary precautions in the city by determining of anthropogenic heavy metal pollution and pollutant sources according to the heavy metal pollution results. A simple and effective method for assessing atmosphere pollution in urban areas is presented.

EXPERIMENTAL

Sampling sites

The City of Sivas has the terrestrial climate in which the winters are cold and snowy, the summers are usually drought and hot, having a height of 1250 m from the sea level. During the period under consideration total snowfall is measured for January, February and March as 75.2, 45.4, 27.0 mm respectively, by National Meteorology Office, Turkey. Sivas is a small industry and agriculture city with the population of about 225000 in central Anatolia, Turkey. The main characteristics of sites selected for sampling in Sivas are the following (see Fig. 1). Site 1 is open to the effect of the

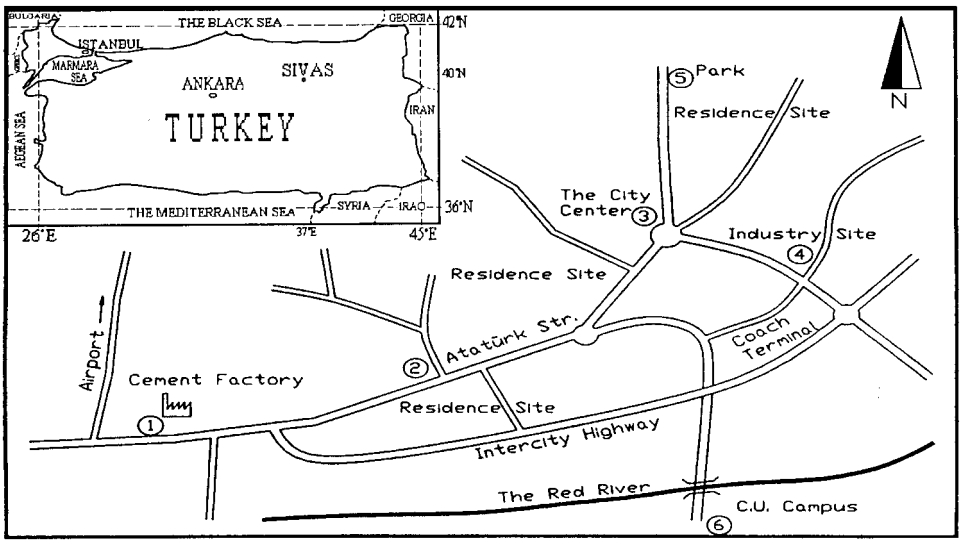

FIGURE 1 Location of the sampling sites for the Sivas city.

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cement factory emissions; site 2 is a residential area with a medium level traffic density; site 3 is the city center exposed to higher vehicular traffic and with dense population; site 4 is an area open to emissions of the automotive industry and some other industrial production sites, and having medium level vehicular traffic; site 5 is also a residential area; site 6 is a non-residential area out of the city atmosphere, chosen as the background level.

Sample collection and preparation for the analysis

Snow samples were collected, three samples (2.0 ± 0.5 kg) from each site (total 18 samples per month), between January–March 2000 for 30 days intervals. There was about 0.7–1.0 m of snow at the sites, in urban areas. A cylinder, 1 m long and 10 cm internal diameter, made of polyvinyl chloride, was used for snow sampling. The snow samples were brought back to the laboratory in deionised polyethylene bottles (3 L). The pH of the melted snow solution was measured immediately after melting. To prevent chemical precipitation forming in the samples, the pH was adjusted to ≤ 1.0 by adding conc. HNO_3 (Merck). After the samples were evaporated to 90 ml final volume (to 45 ml for site 6) by sub-boiling evaporation, 10 ml conc. HNO_3 was added for each sample (5 ml for site 6). Then, the samples were leached in ultrasonic bath with 50–60 kHz nominal frequency for 20 min in order to get high recoveries of elements from atmospheric particulate in the samples. After the leached samples were filtered by suction through a Whatman-42, the total volume was made up to 100 ml (to 50 ml for site 6) using 0.1 M HNO_3 and kept in plastic bottles (100 ml) for AAS analyses. The solutions obtained from the site 6 in January 2000 were used for heavy metal analysis as blank.

Element determinations

The samples were analysed for Zn, Mn, Pb, Cu, Ni and Cd using a UNICAM Model 929 AAS functioning with air/acetylene burner. Each result exhibits the average of four readings. The operating parameters for working elements were set up as recommended by the manufacturer. The concentrations were obtained directly from calibration graphs after correction of the absorbance for the signal from an appropriate reagent blank.

The dynamic range of the heavy metal concentrations required in carrying out both the concentration and dilution of the sample before the analysis. The higher concentrations were attained by sub-boiling evaporation of the samples in the Teflon beakers. Where ever necessary, the samples were diluted with 0.1 M HNO_3. In site 5 concentrations of Pb and Cd and in site 6 concentrations of Pb, Cd, Cu and Ni were found to be at the limit values.

The data subjected to a statistical analysis were produced to examine the relationships between the metal concentrations. Student's *t*-tests were employed to estimate the significance of values[32].

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RESULTS AND DISCUSSION

Heavy metal pollution

The present work provides data on the some heavy metal concentrations in the snow samples of Sivas city, Turkey. The melted snow solutions that were leached under ultrasonic effect have given $12 \pm 3\%$ high recoveries for all elements as compared to non-leached melted snow solutions. The mean concentrations of Ni, Cu, Mn, Zn, Pb and Cd in snow samples and pH of melted snow samples at different locations in Sivas are given as a mean of the three replicates in Table I.

Table I indicates that pH values have increased from January to March for all sites except site 6. While site 1 has the highest pH, site 6 has the lowest. This can be explained that the site 1 is affected by the cement factory emissions. Site 6, which is far away from city's atmosphere showed the lowest pH. In general, pH exhibited parallel behavior with heavy metal concentrations, it slightly increased with the increasing metal pollution. However, pH of the samples varied between 6.39–7.28 (average $\Delta pH = 0.50$), except for sites 1 and 6, and this was not thought to lead the loss of the heavy metals by precipitation at lower pH.

Table I shows that the concentrations of each element were dramatically higher in the inner-city area than background area (site 6). In addition, heavy metal pollution in snow increased from January to March for all sites in the Sivas city. This phenomenon may be the results of the accumulation and mixing of the various pollutants. Elik and Akçay have shown that the mean heavy metal concentrations are more variable at increasing distance from the center of the city and significant[7]. Within the urban area, heavy metal concentrations in the snow are affected by many factors; for example, winds created by buildings and anthropogenic pollutants, which may be brought in from other areas. It is important to select a variety of contrasting sampling sites if the concentration of heavy metal in snow is to be used as an indicator of atmosphere pollution.

As can be seen in Table I, the average heavy metal concentrations of city in the snow samples were in the order Mn > Zn > Pb > Cu > Ni > Cd. Sites 3 and 4 have higher

TABLE I Mean concentrations of heavy metals in snow samples and pH of melted snow samples in the Sivas city in 2000

			μg metal/L ($\bar{x} \pm st/\sqrt{N}, N = 3, P = 0.05$)					
Sites	*Months*	*pH*	*Zn*	*Pb*	*Mn*	*Cu*	*Ni*	*Cd*
1	January	7.60	277.1 ± 7.8	89.0 ± 7.9	299.2 ± 10.9	41.0 ± 6.2	48.56 ± 6.7	0.26 ± 0.05
	February	8.20	336.3 ± 6.9	97.3 ± 8.9	370.4 ± 6.7	66.6 ± 7.4	62.3 ± 3.2	0.28 ± 0.04
	March	9.02	428.5 ± 16.7	145.0 ± 6.2	565.7 ± 7.9	90.4 ± 9.2	80.4 ± 9.7	0.36 ± 0.04
2	January	6.50	247.0 ± 8.7	111.2 ± 7.4	241.1 ± 8.9	50.5 ± 5.5	50.0 ± 6.9	0.33 ± 0.05
	February	6.85	306.3 ± 14.7	154.6 ± 6.7	297.3 ± 10.4	63.5 ± 6.9	64.7 ± 4.4	0.37 ± 0.02
	March	7.03	386.7 ± 10.6	203.3 ± 10.2	414.6 ± 12.4	84.7 ± 10.2	75.1 ± 8.4	0.48 ± 0.06
3	January	6.67	195.0 ± 10.4	134.1 ± 5.7	262.0 ± 7.7	42.6 ± 3.2	50.2 ± 2.7	0.32 ± 0.05
	February	6.71	245.2 ± 15.2	192.4 ± 10.2	342.5 ± 4.7	59.1 ± 6.2	64.0 ± 4.5	0.64 ± 0.07
	March	7.28	371.4 ± 8.8	293.2 ± 10.9	534.1 ± 6.9	84.3 ± 5.2	80.0 ± 5.7	0.82 ± 0.08
4	January	6.45	234.4 ± 6.9	130.0 ± 4.2	237.5 ± 6.9	38.0 ± 7.9	44.4 ± 5.7	0.44 ± 0.04
	February	6.74	297.1 ± 6.3	183.7 ± 7.9	352.9 ± 3.7	64.1 ± 8.9	58.5 ± 4.5	0.77 ± 0.05
	March	6.94	402.5 ± 12.7	245.1 ± 8.9	603.0 ± 10.4	87.0 ± 10.2	80.1 ± 8.4	0.98 ± 0.07
5	January	6.39	109.4 ± 8.0	25.3 ± 5.0	115.0 ± 5.5	28.8 ± 4.2	21.2 ± 2.7	0.18 ± 0.03
	February	6.69	149.6 ± 6.1	44.4 ± 4.2	156.4 ± 8.9	39.8 ± 2.2	29.0 ± 3.7	0.20 ± 0.03
	March	6.80	189.2 ± 10.0	72.5 ± 7.9	190.3 ± 7.4	56.2 ± 5.4	40.3 ± 5.2	0.26 ± 0.03
6	January	6.22	20.7 ± 3.0	10.0 ± 5.0	20.4 ± 5.2	8.1 ± 5.2	10.1 ± 3.0	0.10 ± 0.03
	February	6.10	21.1 ± 5.7	10.2 ± 4.4	22.9 ± 7.4	10.2 ± 4.2	11.3 ± 3.7	0.10 ± 0.02
	March	6.17	21.8 ± 1.5	11.1 ± 5.7	23.1 ± 4.7	10.9 ± 3.0	12.0 ± 1.7	0.12 ± 0.04
General average levels			250.1	128.2	296.3	54.7	53.6	0.41

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vehicular traffic. The lead levels are the lowest at site 6 (13.1 μg/L) and the highest by a factor of 18–27 at site 3 (293.2 μg/L), site 4 (245.1 μg/L) and site 2 (203.3 μg/L). Similar decreasing trends have also been observed for concentrations of Zn, Mn, Cu, Ni and Cd in snow. In general, most of the sites in Sivas showed a significant correlation between airborne Pb and vehicular (petrol fuelled) traffic density indicating that automobile exhaust emission could be the prominent source of atmospheric Pb in the city[7], because of the usage of leaded petrol on the automobiles in Turkey. The Pb concent of gasoline is between 150–400 mg/L at Turkey. Higher levels of Zn and Cd in high vehicular traffic indicate that fragmentation of car tyres is a likely source of these metals[33, 34]. Ni and Cu comes from corrosion of metallic parts of cars[35]. The main origin of Mn is from geological structure of middle Anatolia of Turkey and traffic[13].

Concentrations of Pb, Cd, Zn, Mn, Ni and Cu in snow as an indicator of city atmosphere pollution were measured in industrial areas of the city as compared to residential areas. The concentrations of Pb, Zn, Mn and Cd in industrial areas were found to be 245.1 μg/L, 402.5 μg/L, 603.0 μg/L and 0.98 μg/L, respectively, while it were 72.5 μg/L, 189.2 μg/L, 190.3 μg/L and 0.26 μg/L in residential areas in Sivas. The cement factory is a major source of Mn, Zn, Cu and Ni (see Table I).

Each value in Table I is the mean of three replicate measurements and RSDs were calculated from pooled data for the method. In the precision test, the average RSD% of snow samples for all metals are in the range of 2.07–5.62% ($n = 54$) for method. The statistical analysis (student's t test)[32] based on these data showed that snow samples collected from sites located in the city and its surroundings are under strong anthropogenic pollution, since results were significantly different at 0.05 probability (P), according to the background level site (site 6).

Smelting and machining of nonferrous materials are responsible for enhanced concentrations of metals in the atmosphere of Sivas. We will try to distinguish between pollution, caused by municipal, coal combustion furnace, industries and traffic in the Sivas city.

Background levels

The concentrations of pollutants in the atmosphere depend on a variety of mechanisms, as well as on the uniqueness of each area, so that the assessment of background levels is problematic. There have been reports on atmospheric precipitation sampling procedures, and chemical properties and chemical composition have been studied in attempts to determine the composition of background aerosol[4, 36, 37]. As the concentrations of heavy metals in snow vary according to different meteorological situations, it is insufficient to show that heavy metal concentrations increase with time in the yearly layers of snow, for this could result from a number of different factors, some of which are unrelated to general increases of heavy metals in the atmosphere. Heavy metal concentrations in snow can be more reliably related to atmospheric concentrations if they are compared with their background level concentrations in snow from an urban area. Murozumi *et al*., analysed the concentrations of heavy metals in snow from Greenland and showed that the concentration of Lead in snow is today $\sim 0.2\,\mu g/L$ vs. only $0.001\,\mu g/L$ 3000 years ago[19].

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Table II shows a comparison of the mean values of heavy metal concentrations in snow collected from unpolluted areas on Greenland[38], from Nebraska in the USA[39], from Sapporo in the Japan[4] and from the Sivas (this study) in the Turkey. At the Sivas, the heavy metal concentrations in the snow higher than the mean values at the Nebraska, Greenland and Sapporo, but site 6 in Sivas was thought to be unaffected by anthropogenic sources. These concentrations can therefore be regarded as background values for snow in Sivas. Natural reference levels have also been established for the polar areas[38].

Because the concentrations of heavy metals in atmosphere depend on a variety of mechanisms, it has always been difficult to assess heavy metal pollution. The pollution factor (PF) was chosen as a general indicator of anthropogenic inputs to the heavy metal budget of the snow samples. The limitations of this approach, caused by variable composition of the crustal components, are well known[40]. Nonetheless, in cases when PFs differ by orders of magnitude for samples collected in this same general area, they can be treated as a reasonably valid measure of the anthropogenic pollution. Any pollution over background levels in the considered snow material can be called as anthropogenic pollution in the city atmosphere.

The PF as a simple and effective method for assessing of city atmosphere pollution is defined as the ratio of heavy metal contents in urban snow to the heavy metal contents

TABLE II Comparison of heavy metal concentrations in snow of unpolluted areas

	Element, μg metal/L						
	Pb	*Zn*	*Mn*	*Cu*	*Cd*	*Ni*	*References*
Greenland	0.5	1.05	0.25	0.85	0.64	—	Weiss *et al*., 1975
Nebraska	7.8	12.0	4.0	4.0	0.63	—	Struempler, 1976
Sapporo	2.3	3.4	2.5	0.6	0.05	—	Sakai *et al*., 1988
Sivas	10.2 ± 4.4	21.1 ± 5.7	22.9 ± 7.4	10.2 ± 4.2	0.10 ± 0.02	11.3 ± 3.7	This study, 2000

in unpolluted snow (background level). Values for snow from site 6 were taken as background levels for Sivas; at this site, the heavy metal concentrations were at their minimum value.

Pollutant sources

Table III shows a comparison of the average heavy metal PFs according to sites. The PF values obtained in our study show that Mn and Zn concentrations were about 20.61 and 14.27 times greater than the background levels at site 1, Pb, Mn, Zn and Cd were about 22.54, 22.33, 13.40 and 8.16 times greater at site 4, and Pb, Cd, Cu and Ni were about 22.59, 6.83, 6.46 and 5.80 times greater at site 3, in March, respectively.

The PFs of Mn, Zn and Pb were greater than those highly enriched in snow samples collected at the site 1 located *ca*. 100 m from the cement plant. The Pb is highly enriched in snow samples taken from sites 3, 4 and 2. The different level of Pb enrichment at these three sites is paralleled by the particulate material load, and reflects the relative volume of vehicular traffic in these three locations. The high Pb enrichment can be attributed to Pb from the automobile emissions[41] and accumulation of this element in the road dust[42]. The Mn concent was higher at sites 1, 3 and 4. Barrie and Vet have pointed out that Mn is significantly enriched in the snowpack relative to the crustal rock composition[43]. So a high Mn concentration may result from soil or street dust carried by automobiles from rural areas. High levels of Zn are present in snow samples collected downtown. The proximity of the cement plant may be responsible for high levels of Zn at the site 1, while its enrichment in the downtown snow samples can be explained by the presence of Zn from tire wear in the resuspended road way dust[42]. The Cd can come from a variety of local sources. Although its concentrations in the snow are relatively low, the PFs indicate a strong anthropogenic input. Low concentrations of Cu and Ni in snow samples can be expected on the basis of its

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TABLE III Average heavy metal PFs according to sites.

Sites	*Months*	*Pb*	*Zn*	*Mn*	*Cu*	*Ni*	*Cd*
1	January	8.92	13.85	14.95	5.12	4.80	2.61
	February	9.74	16.00	16.09	6.60	5.17	2.82
	March	13.18	19.44	24.56	8.17	6.67	3.00
2	January	11.10	12.35	12.11	6.25	5.00	3.30
	February	15.40	14.57	12.91	6.30	5.33	3.71
	March	18.45	17.54	18.00	7.66	6.27	4.06
3	January	13.43	9.75	13.14	5.25	5.03	3.18
	February	19.22	11.66	14.93	5.90	5.32	6.54
	March	26.59	16.86	23.20	7.56	6.67	6.86
4	January	13.00	11.72	11.85	4.75	4.41	4.42
	February	18.30	14.14	15.30	6.40	4.84	7.76
	March	22.24	18.25	26.23	7.89	6.68	8.16
5	January	2.50	5.45	5.75	3.50	2.10	1.80
	February	4.40	7.10	6.78	3.90	2.42	2.07
	March	6.59	8.59	8.29	5.12	3.33	2.17
General average		13.53	13.15	14.93	6.02	4.93	3.71

low emission rates. However, small PFs for Cu and Ni are out of step with the other heavy metals (Mn, Zn and Pb). Oil combustion, automobile emissions and residential coal combustion can be the main source of Cu and Ni.

It is clear that average PFs arisen from anthropogenic resources show a regular increase from January to March for all metals at each site. The average heavy metal PFs of Sivas city are given in Table III. As it can be seen, the average heavy metal PFs are in the order Mn (14.93) > Pb (13.53) > Zn (13.15) > Cu (6.02) > Ni (4.93) > Cd (3.71), (general average for all months and sites). In accordance with previous results, the PF values of sites 1, 3 and 4 are the highest and site 5 is the lowest for all metals. It is considered that the PFs of heavy metals at each site are suitable for use as an index of atmosphere pollution.

It appears that the city is under heavy Mn, Pb and Zn pollution while Cu, Ni and Cd pollution are lower.

The average concentrations of Pb, Zn and Ni in the snow samples collected from Sivas – Turkey are somewhat higher than levels reported for snow samples collected in inner urban area from Sapporo – Japan [4], but are comparable for Mn, Cu and Cd.

These studies do not indicate the individual sources of heavy metal pollution of an environment. Central to all major questions concerning assessment of environmental pollution using trace and minor elements is the availability of a set of natural background values. A background value for any element may undergo extreme variations between different regions and even within a particular region, as a result of factors such as source-rock geology and weathering conditions.

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CONCLUSION

The aim of this contribution was to define the essential characteristics of atmospheric pollution and the source of heavy metal pollution in an area of heavy snow. This study demonstrated that snow chemistry in the Sivas city atmosphere successfully reflects the magnitude of the local atmospheric deposition. We examined the question of whether the concentrations of Mn, Zn, Pb, Ni, Cu and Cd in snow were suitable for use as an index of atmosphere pollution. We found the measurement of heavy metal concentrations in snow, which is important in monitoring environmental pollution, to be a simple reproducible and effective method for assessing atmosphere pollution in urban areas. Besides, the melted snow solutions that were leached under ultrasonic effect have given $12 \pm 3\%$ high recoveries for elements as compared to non-leached melted snow solutions. The heavy metal concentrations in the snow clearly increased from January to March. In the cities, the heavy metal pollution in snow was influenced more by the traffic and industrialization than by population size. The highest heavy metal concentrations were found in the sites (sites 1 and 4) with industrial emissions and in the sites (sites 3 and 2) with heavy traffic. The lowest levels for the heavy metals in snow were found in the sites 6 and 5. The mean concentrations of the elements studied were in the order Mn > Zn > Pb > Cu, Ni > Cd.

Results found for the snow samples showed that snow samples contained significant levels of the metals studied compared with the background values. The PFs were fairly consistent with heavy metal pollution originating from anthropogenic sources. The PF can be used as simple and effective indicators of urban atmosphere pollution.

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