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Chromated Cooling Tower Drift and the Terrestrial Environment: A Review*

By F. G. Taylor, Jr.†

Abstract: *Numerous investigations have been conducted at Department of Energy (DOE) facilities in Paducah, Ky., and Oak Ridge, Tenn., to determine the environmental effects of cooling tower drift and the effects of a specific drift toxicant (hexavalent chromium) on biological systems. The results of these investigations provided the first quantitative evidence of the transport of chromium in cooling tower drift to vegetation in the surrounding area, illustrated that most of the drift (>75%) fell within DOE properties (1 km downwind), and provided evidence for potential adverse biological effects in plants. Additional studies investigated the transfer of drift constituents along food chains and the horizontal and vertical movement of drift chemicals in soils by moisture flow. Experiments simulating drift provided estimates of fractional interception and retention times of drift on plants with diverse foliage (grasses, pine, broadleaf plants). This review provides condensed summaries of several ecological studies; although the studies are site specific, the results have generic application in monitoring design, sample collection, and assessment of drift effects at other sites.*

In implementing the National Environmental Policy Act of 1969 (NEPA), the Atomic Safety and Licensing Board (ASLB) issued a decision calling for the retrofitting of cooling towers at the Indian Point power plant on the Hudson River. The use of cooling towers was viewed as a means of meeting thermal discharge requirements while reducing aquatic impacts related to the entrainment and impingement of juvenile fish. When this decision was made, the potential for environmental impact deviated, in part, from an aquatic to a terrestrial problem. Water entrained in the air flow (drift) coming from the cooling towers would contain elements (salts) found in the recirculating cooling water system. Thus the drift from the cooling towers would represent a pathway for the transfer of

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increased levels of trace contaminants contained in water treatment chemicals (corrosion inhibitors, biocides, algacides, etc.) to terrestrial environments.

At Department of Energy (DOE) uranium enrichment facilities (gaseous diffusion plants) in Oak Ridge, Tenn., Paducah, Ky., and Portsmouth, Ohio, the heat of compression (up to 2000 MW) is dissipated by mechanical-draft cooling towers that use a chromate, zinc–phosphate corrosion inhibitor (20 ppm CrO_4^{2-}) in the recirculating cooling water systems. Hexavalent chromium (Cr^{6+}) has been identified as a potential toxicant and carcinogen.[1] The towers have been in operation for more than two decades and therefore represent an opportunity for identifying impacts from a known toxicant (Cr^{6+}), while determining the distribution and quantifying the levels of the toxicant in an ecological system.

A research activity was initiated to inventory drift-derived chromium along the axis of predominant winds at the Oak Ridge Gaseous Diffusion Plant (ORGDP). The results provided the first quantitative evidence for the transport of chromium in drift from a cooling tower to vegetation and also provided evidence for potential adverse biological effects in test plants.[2] Additional studies conducted at Oak Ridge and Paducah were aimed at identifying the transfer of drift chemicals along food chains[3] and the horizontal and vertical movement of drift chemicals in soils by means of moisture flow.[4]

An anticipated increase in enrichment capacity through the Cascade Improvement Program (CIP) and the Cascade Uprating Program (CUP) at the enrichment plants has resulted in a greater demand for cooling capability by 1983. New cooling towers have been built and new cells have been added to the existing cooling towers to meet these demands. Operation of the facilities at uprated levels will enhance the significance of ecological studies, since more drift will be associated with the increased cooling capacity.

The purpose of this article is to provide condensed summaries in a single document of several ecological studies conducted at the DOE facilities. Most of the research has been presented in detail in topical reports or publications, which are appropriately cited. Results of some subprojects are presented here for the first time.

On the basis of modeling predictions, the greatest environmental concentrations of drift chemicals (chromium and zinc at DOE facilities) would be expected to occur adjacent to the cooling tower and to decrease by atmospheric dispersion as a function of distance from the tower. Interception of drift by vegetation and soil, expressed as elemental concentrations, should correspond to predictions of drift dispersion across the landscape. Ratios of drift-derived chromium and zinc deposits to baseline concentrations in vegetation would illustrate the quantity of these trace elements which is transferred to terrestrial environments in the vicinity of the cooling towers. Analyses of the metal content of vegetation growing along horizontal gradients from the cooling tower should characterize the distribution of drift (and the chromium therein) in vegetation and identify the areas of greatest impact. Harvesting plots were located along radial transects in the direction of predominant winds at each DOE facility, and samples were collected at varying distances from the towers.

While this research has been site specific, the results have provided guidance applicable to generic studies in monitoring design, sample collection, and assessment of drift effects in ecosystems. The results of the research have proven valuable to environmental protection managers in upgrading monitoring protocols at their respective DOE facilities and have contributed to the evaluation and preparation of environmental impact assessments.

VEGETATION STUDIES

Oak Ridge Gaseous Diffusion Plant

Vegetation typical of various plant communities of the DOE reservation at Oak Ridge, Tenn., were collected from permanent plot locations and analyzed for chromium and zinc.[2] Grass samples consisted of fescue grass (*Festuca arundinacea* Schreb.), a common pasture species, and broom sedge (*Andropogon virginicus* L.), a dominant grass of old fields. Concentrations of chromium, expressed in parts per million (micrograms per gram), were plotted as a function of distance from the base of the cooling towers to 1.8 km (Fig. 1). Maximum concentrations [±1 standard error (SE)] observed 15 m from the tower ranged from 339 ± 117 ppm to 644 ± 94 ppm for grasses and litter, respectively. Concentrations decreased exponentially with distance, reaching near background levels (0.4 and 2.7 ppm for grass and litter, respectively) at 1830 m. The land around the tower (to nearly 450 m from the tower) is predominantly grassland with dense cover and heavy litter accumulations. These factors may contribute to the seemingly increased concentrations observed at 300 m, since both components afford effective trapping sites for drift. Forbs are not present adjacent to the tower because grass is the dominant

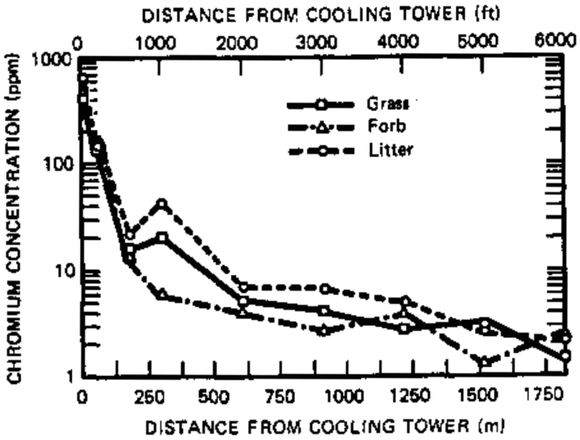

Fig. 1 Chromium concentrations in vegetation illustrating the transfer of increased quantities of chromium by cooling tower drift to the landscape. Background concentrations (parts per million ± 1 standard error) are: grass, 0.40 ± 0.03; forb, 0.65 ± 0.05; and litter, 2.65 ± 0.74.

vegetation maintained there. Forbs first occur 60 m east of the tower; at that location the concentrations of chromium in forbs are similar to those in grass and litter (157 ± 62 ppm). Empirical data suggest that transfer of chromium by drift is detectable above background to at least 1.8 km but that the major deposition occurs within the first 900 m (Fig. 1). The ratios of drift-derived chromium to background ranged from several orders of magnitude for grass and litter adjacent to the tower to four times greater than background at 1.8 km.

Trees were separated into morphologically different forms (deciduous and coniferous) for determination of foliar concentrations. With the exception of sycamore samples (*Platanus occidentalis* L.) collected 60 and 180 m from the towers, the category of deciduous trees consisted of a single taxon (dogwood); the category of coniferous trees consisted of loblolly pine (*Pinus taeda* L.) and junipers, which were collected at 180 m. Trees are relatively sparse within the environs of the production facility but become dominant features of the landscape beyond 300 m. Chromium concentrations in tree foliage showed exponential decreases similar to those observed for grass and litter, ranging from 27 to 1 ppm for deciduous broadleaf trees and from 7 to 0.5 ppm for conifers at 180 and 1830 m, respectively. Major concentrations, indicated by increased levels above background, occurred within 900 m. Chromium levels in deciduous species were consistently higher (by a factor of 2) than those in coniferous species. This may be attributed to different morphological characteristics of the taxa. Leaves of broadleaf species have a wide, flat plane, which presents a more accessible surface for the interception of drift than the cylindrical needles of pines.

Zinc concentrations in foliage follow exponential patterns similar to those of chromium but are of minor significance. The concentrations detected 15 m from the tower average 61 ± 5.3 ppm and 243 ± 31.8 ppm for grass and litter, respectively. Beyond 200 m, increased quantities of this trace element could not be differentiated from background levels. At 200 m, litter accumulated zinc by a factor of 2 above grass. Zinc concentrations in deciduous broadleaf trees and conifers were near background levels of 14 and 18 ppm at 200 m.

The most commonly observed symptom associated with excess zinc in plants is similar to the effect of iron chlorosis (i.e., yellowing of the leaves).[5] Because the landscape of the ORGDP is underlain by Chickamauga limestone, vegetation subjected to increased quantities of zinc from drift would not be expected to show symptoms of excess zinc, since additions of calcium carbonate (the major constituent of limestone) often reduce or overcome symptoms of zinc toxicity.[6]

Cross sections of trunks of mature trees were collected and segmented into growth increments corresponding to the year of maturation and then analyzed for trace element concentrations in an attempt to relate concentrations with operational data. Results show uniform quantities of chromium or zinc in all ages of wood (xylem) during the past two decades of cooling tower operation, averaging 0.27 ± 0.03 ppm for chromium and 2.37 ± 0.27 ppm for zinc. Concentration ratios of bark to xylem suggest a 20- and 12-fold increase in chromium and zinc, respectively. The higher concentrations in bark suggest surface contamination rather than uptake by roots as the mode of chromium and zinc incorporation by trees. Because zinc occurs naturally in relatively high concentrations in parent soils and plants, routine analyses for this element were eliminated from further studies.

Paducah Gaseous Diffusion Plant

The results of the vegetation survey at the Paducah Gaseous Diffusion Plant are similar to the findings of the study conducted at the ORGDP.[2] Concentrations in foliage at Paducah were consistently lower at all distances than the levels found at Oak Ridge.

There was uniformity of drift contamination in fescue grass among the transects in the direction of

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predominant wind dispersion at the Paducah site, with a rapid dilution of the containment in the environs.[4] Maximum concentrations within 15 m of the tower basin ranged from 251 ± 19 ppm in foliage to 1813 ± 56 ppm in litter. At the perimeter fence (≅100 m), chromium concentrations in foliage (37.0 ± 4.4 ppm) and litter (141.1 ± 11.7 ppm) were still several times greater than background levels (0.52 ± 0.1 ppm and 2.9 ± 0.4 ppm for foliage and litter, respectfully). Drift concentrations of chromium remained significantly greater than controls at the perimeter of the buffer zone (300 m) between the fence and the wildlife management area, reaching background levels at 1.5 km.

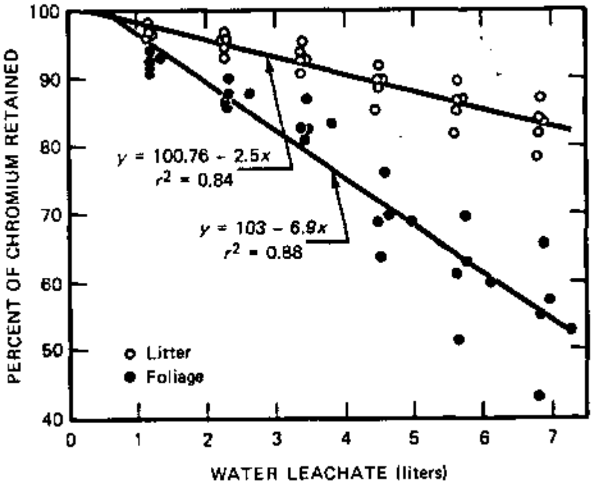

Fig. 2 Retention of drift-derived chromium on fescue foliage and litter following application of water to simulate throughfall equivalent to from 25 to 152 mm of rain (1.27 liters to 0.05 m² plot = 25 mm). (The statistic r_2 is the coefficient of determination.)

Drift Leachability

The two- to fourfold difference in chromium concentration noted between foliage and litter in a previous study[2] suggested that there were more exchange sites associated with the litter than with the foliage. It was hypothesized that the differences in concentration between the foliage and litter were related to the chemical properties of chromium rather than the physical trapping of the chemical residue. Drift deposited as hexavalent chromium on foliage should remain relatively water soluble, whereas the deposition on litter would likely be reduced to the less soluble trivalent species by humic substances and subsequently become bound to the decaying mat.

In a laboratory study, foliage and litter were leached to simulate throughfall from 25 to 152 mm of rain.[4,7] The content of drift in the leachate indicated that the rate of chromium loss from grass foliage was two to three times greater than the loss from litter (Fig. 2). Approximately 7 to 9% of the initial drift contaminant was removed from foliage with each throughfall simulant in comparison to only 3% from the litter. These data support the hypothesis that the difference in concentration between foliage and litter is likely related to a differential solubility of chromium in two oxidation states (Cr^{3+} and Cr^{6+}) and subsequent field losses due to weathering.

Following the leaching experiment, fescue foliage was examined, using a scanning electron microscope to determine the sites of drift contamination and the size of drift particles.[8] Both upper and lower surfaces of leaves were examined from samples collected at 60 and 300 m. Drift particles observed were deposited either as particulates or droplets which subsequently dried leaving a mineral residue. Particle density on the upper surfaces of leaves collected at 60 m was much less than that on the lower surfaces, and at 300 m particle density on both surfaces was less than that on the corresponding surfaces at 60 m. Recirculating cooling water (RCW) completely evaporated, depositing residue similar in size (6 to 40 μm in diameter) and configuration to the deposits observed on the foliage (5 to 50 μm in diameter).

To test whether the foliar deposits were actually drift-derived, energy dispersive X-ray analyses were made on the drop residue and compared by similar analyses to spectral characteristics of foliar deposits. The electron-induced X-ray fluorescence analyses showed that particles on the foliage contained elements common to the chemistry of the cooling water drop (Na, Mg, Si, S, Cl, K, Ca, and Fe). Each of these elements are contained in the RCW following concentration by several cooling cycles or in the makeup water. Analyses of washed and unwashed leaves from control areas did not produce comparable emissions spectra.

All particles examined did not contain the full spectrum of elements found in the cooling water drop. This suggests that the concentration of a specific element in individual drift droplets may not be equivalent to the concentration maintained in the recirculating water. This is an important finding, since a common assumption used in the calculation of cooling tower drift emissions is that drift droplets contain the same mineral concentration as the circu-

Table 1 Initial Interception of Simulated Drift by Vegetation*

	m_i, μCi/m² ± 1 SE	C_i, μCi/g ± 1 SE	a_i, C_i/m_i	W_l, g/m² ± 1 SE	F, a_iW_l	LAI, m²/m²
Fescue grass	89.6 ± 4.7	0.1831 ± 0.04	0.0020	118.69 ± 20.2	0.2374	2.30
Loblolly pine	89.6 ± 4.7	0.2239 ± 0.05	0.0025	181.99 ± 23.1	0.4550	2.85
Yellow poplar	89.6 ± 4.7	0.8877 ± 0.15	0.0099	72.57 ± 9.9	0.7184	4.00

*m_i = Initial deposition.
C_i = Initial interception per gram of foliage.
a_i = Contamination factor.
W_l = Plant density.
F = Fraction of initial deposition intercepted.
LAI = Leaf area index.
SE = Standard error.

lating water. The low density of deposits on the upper surfaces of the leaves suggests a field loss due to weathering. The chromium maintained in the cooling water for corrosion inhibition could not be detected by the X-ray analyses, since the concentration (9 ppm) is below the limit of detection (0.1 to 1%) by the energy dispersive technique.

Interception and Retention of Simulated Cooling Tower Drift (100 to 1300 μm in Diameter) by Vegetation

Predictions of drift deposition (average annual) from present models assume 100% interception by the substrate (soil surface). The potential for effects in plants should be proportional to the quantity of drift intercepted, retained, and available for uptake through the foliage. Variations in initial interception between plant genera are related in part to diverse leaf morphologies. Because the contamination of foliage by cooling tower drift is effected partially by droplet sedimentation, methods for estimating initial interception and retention are assumed to be similar to techniques used in evaluating the fate of simulated fallout in plant species.[9]

To determine the fraction of drift initially intercepted by foliage, a portable drift simulator was designed and fabricated to generate a spectrum of droplets representative of the droplet diameters expected in the drift at the discharge stacks of cooling towers, using current state-of-the-art drift eliminator designs. Radiolabeled sodium chromate ($Na_2{}^{51}CrO_4$) was added to the liquid to simulate the chromate in the recirculating cooling water of the ORGDP cooling towers.[7,8]

Fescue grass and loblolly pine and yellow poplar seedlings growing in pots were exposed once to the drift simulant. Immediately following contamination, plants were randomly selected, clipped, and bagged to determine the initial reception of the drift simulant. Subsequent collections were made at weekly intervals to determine the retention of drift on foliage.

Following the notation of Miller,[10] the initial concentration of the drift simulant on foliage may be expressed by a foliage contamination factor, a_i, such that

$$a_i = C_i/m_i \qquad (1)$$

where C_i is the quantity in microcuries of radionuclide initially intercepted per gram of dry weight of foliage and m_i is the quantity in microcuries of drift simulant deposited per square meter of soil surface area. The fraction, F, of drift that is initially intercepted by foliage is given by

$$F = a_iW_l \qquad (2)$$

where W_l is the biomass (dry weight) of foliage in grams per square meter of soil surface.

Species with high ratios of leaf surface area to soil surface area have greater interception efficiencies than species with low ratios of leaf surface area to soil surface area. [These ratios are referred to as the leaf area index (LAI).] For example, the yellow poplar and loblolly pine seedlings in this study, with LAIs between 4 and 2.85, intercepted 72 and 64% of the deposition flux, respectively, whereas fescue grass, with an LAI of 2.3, intercepted 24% (Table 1). The higher contamination found in the leaves of the yellow poplar (Fig. 3) is likely a function of the orientation of its foliage perpendicular to the deposition path in addition to the greater surface area of its foliage. A study by Chamberlain[11] showed that the fraction of contaminant initially intercepted by herbage is related to an

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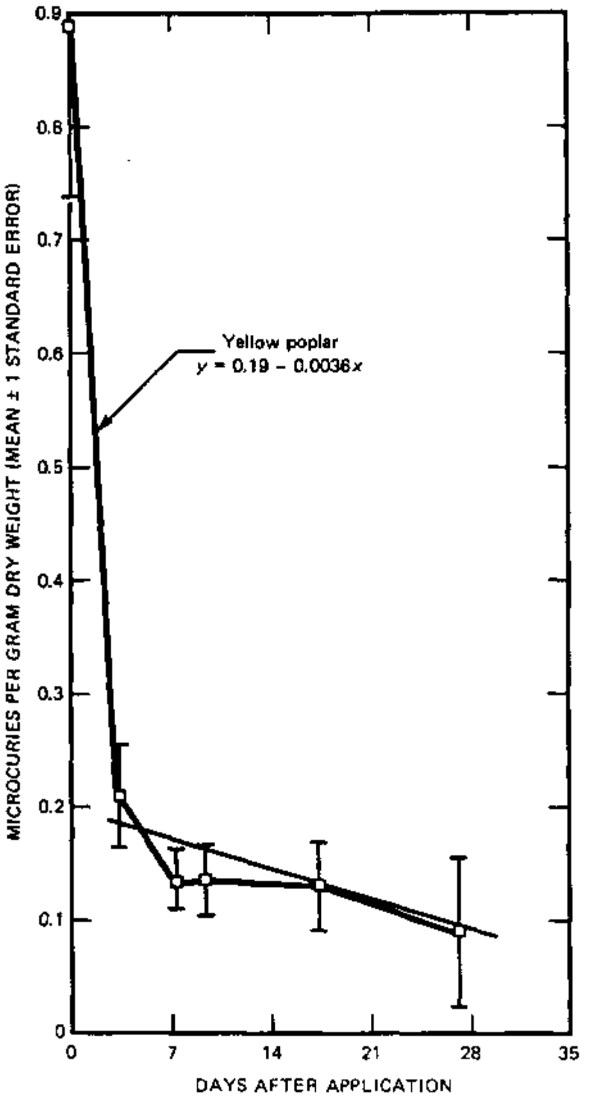

Fig. 3 Retention of ^{51}Cr on vegetation up to 9 weeks after application. Each data point represents the mean (number of samples = 4) at each sample period. Bars denote ±1 standard error (SE).

uptake coefficient which is dependent on biomass density, with experimental values of uptake coefficient ranging from 2.3 to 3.3 m^2/kg among grasses. In the present study, uptake coefficients were computed and found to be 2.27 and 3.33 m^2/kg for fescue grass and loblolly pines, respectively; these values are comparable to Chamberlain's results. On the other hand, the uptake coefficient for yellow poplar was 17.49. Among yellow poplars, the ratios of leaf surface area to biomass were two to three times greater than for fescue grass and loblolly pines, suggesting that the area of the receptor was independent of biomass in determining the fraction of initial interception. Surface characteristics, such as pubescence or roughness, seem to be insignificant to initial interception in this study, since the surface texture of the foliage among the three genera is similar. The retention of the drift simulant by the three species resulted in a two-component curve (Figs. 3 and 4), which was resolved into a short-time or initial-loss phase (0 to 3 days) and a long-time component (3 to 63 days).

Most of the initial contaminant (60 to 75%) was lost by all species during the first 3 days following contamination. Since the chromate tag was water soluble and since there was no rainfall before the harvesting 3 days after application, the initial quantities lost (~75%) were likely in the coalescing dew drops which fell from the foliage by gravity flow. Loss of the remaining simulant (~25%) over the long-time component (3 to 63 days) showed no correlation with the occurrence of rainfall or with the quantity of precipitation. Chamberlain[11] suggested that leaching of radioactive aerosols from foliage by rain was not the major mechanism of field loss. It has been demonstrated experimentally that plants are capable of releasing trace metals into the atmosphere.[12] Such releases, or field losses, are due primarily to cuticle weathering, resulting from surface abrasion and leaf bending induced by various stresses. The trace contaminants released are likely contained in salt particles, wax rodlets, or cuticular and other surface fragments.[13] It is hypothesized in this study that a significant fraction of the remaining simulant (25%) removed over the long-time component (3 to 63 days) was released from the plants in this manner. The rates at which these materials (surface fragments) are removed can be enhanced by rain, dew, and wind. Linear regression analyses were performed on ^{51}Cr concentrations through the time period of 3 to 63 days after application to estimate retention times of the remaining fraction (Figs. 3 and 4). Retention half-times of the remaining fractions were 4 weeks for loblolly pines and yellow poplars and 6 weeks for fescue grass. Less than 1% was estimated to remain on loblolly pine and yellow poplar seedlings at 7 weeks (Fig. 3) compared to the same quantity of fescue grass at 11 weeks (Fig. 4). The ratios of the slopes of the regression curves suggest that loss rates are the same for loblolly pines and fescue grass, whereas the loss rate for yellow poplar is three to four times greater than that for the pines or fescue grass.

These results are relevant to the assessment of drift effects in vegetation, where drift deposition to the

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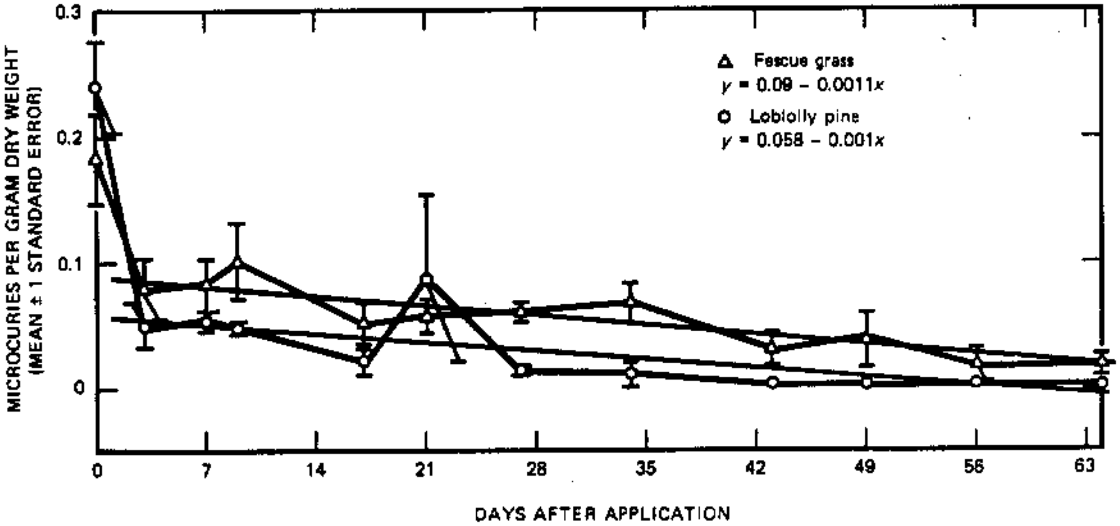

Fig. 4 Retention of ^{51}Cr on vegetation up to 9 weeks after application. Each data point represents the mean (number of samples = 4) at each sample period. Bars denote ±1 standard error (SE).

environs from operating and proposed plants has been estimated by dispersion model techniques. The estimates of initial interception probably represent worst-case examples, since they are derived from a droplet distribution characteristic of discharges at the stack rather than at the point of impingement and represent estimates for individual plants without influence of a canopy. Consequently, an assessment of potential drift effects in vegetation incorporating these results should provide conservative estimates.

DRIFT EFFECTS

Responses of Tobacco to Chromium

The effects of chromium from cooling tower drift on biota are unknown. Tobacco, *Nicotiana tabacum*, was exposed to drift from ORGDP cooling towers to assess the effect of aerosols containing chromium on plant growth.[14] Tobacco was selected because it is reported[15] to be sensitive to elevated levels of chromium (CrO_4^{2-}). Potted plants were placed downwind at distances of 15, 200, 600, and 1400 m from the cooling towers. Control plants were located far from the drift on Chestnut Ridge. Plants were harvested at 1-week intervals from each location and, in addition to chromium concentration, the total plant biomass, leaf biomass, leaf area, and general vigor were determined. Leaf size proved to be the most sensitive parameter for assessing effects on growth.

All plants accumulated chromium above the background levels within 1 week, at which time the concentration in plants 15 m from the cooling towers was approximately 30 times background concentration. These plants attained a maximum foliar concentration of 237 ± 18 ppm after 5 weeks. Lower than maximum concentrations of chromium accumulated on plants at 200, 600, and 1400 m. After the fifth week, there was a decrease in chromium concentration related to a 40% reduction in the chromate in the cooling tower makeup water. The decrease in the chromium level in the makeup water was reflected by reduced concentrations of chromium in tobacco plants at each distance, which further illustrates the sensitivity of tobacco plants as receptors of airborne contaminants from industrial sources.

The effect of increased quantities of chromium is illustrated by the 75% reduction in leaf size of plants at 15 and 200 m distance compared with controls and those located at distances of 600 and 1400 m (Fig. 5). The difference is statistically significant ($P^* < 0.01$), and these data constitute the first reported effects on plant growth from a chemical in cooling tower drift. The effects were manifested at chromium concentrations of <10 ppm in foliage. This observation is consistent with the report that 5 ppm was toxic when the chromium was supplied to tobacco in nutrient culture.[5] Similar effects would not be expected with

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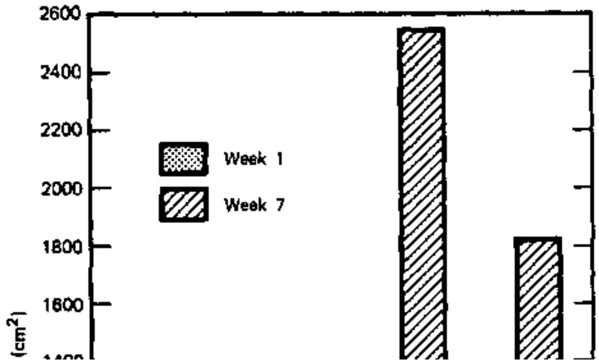
*Probability.



controls and the aerosol-treatment groups, whereas leaf weights in the soil-treatment group were significantly lower ($P < 0.01$) than those in the controls or the aerosol-treatment group. Chromium concentrations in the roots of the soil-treatment group were significantly greater than chromium concentrations in the roots of other groups. Concentration factors for roots (5.1 to 14.3) of plants grown in soils treated with the cooling water were two orders of magnitude greater than concentration factors for leaves (0.05 to 0.2), indicating that the chromium accumulated at the site

Table 2 Comparison of Distribution Patterns of Chromium in Cotton Rats (*Sigmodon hispidus* Say and Ord) Exposed to Cooling Tower Drift and in Control Animals

Organ or tissue	Freeze-dry weight, g	Concentrations (μg/g ± 1 SE)	
		Control	ORGDP
Heart	0.08	0.105 ± 0.013	0.124 ± 0.016
Liver	0.89	0.046 ± 0.016	0.160 ± 0.039
Kidney	0.19	0.087 ± 0.014	0.124 ± 0.040
Spleen	0.02	0.493 ± 0.123	0.713 ± 0.084
Lung	0.10	0.289 ± 0.072	0.292 ± 0.063
Bone	5.02	0.160 ± 0.008	0.460 ± 0.024*
Muscle	13.82	0.234 ± 0.043	0.288 ± 0.029
Gastrointestinal tract	0.43	1.046 ± 0.277	1.006 ± 0.183
Pelt	8.35	0.092 ± 0.007	1.056 ± 0.133*
Hair	0.98	0.395 ± 0.021	4.397 ± 0.555*
Residual†	1.67‡	0.200 ± 0.032	0.311 ± 0.025

*$P < 0.01$ significant difference.

†Blood, reproductive organs, brain, salivary glands, and thyroid.

‡Converted from mean ratio of freeze-dry weight to fresh weight of all other soft tissues.

by feeding 20 adult animals fescue grass labeled with isotopic chromium in a chemical form ($Na_2{}^{51}CrO_4$) similar to the form maintained in the recirculating cooling waters. Each animal was fed grass containing 3.75 μCi of ^{51}Cr and was measured radiometrically for retention during the 2-week period following feeding. Initial whole-body radioactivity (A_0) was 0.0073 μCi/g ± 0.001 SE. The elimination of ^{51}Cr by the cotton rats resulted in a two-component curve which was resolved into a short component representing gut clearance and a long component illustrating loss of the assimilated fraction. The retention data were fitted to an exponential model[17] described by the equation

$$A_t = A_0\,(a_1 e^{-\lambda_1 t} + a_2 e^{-\lambda_2 t}) \tag{3}$$

where A_t represents whole-body activity at time t, A_0 is the initial whole-body activity, and a_2 and a_1 describe the assimilated and nonassimilated radionuclide eliminated at rates λ_2 and λ_1, respectively.

The percent assimilated, as determined from the intercept of the long component, is 0.79 or 0.06×10^{-3} μCi/g, where $a_2 = 1 - a_1$ or $a_2 = 1 - 0.9921$. Within 3 days after feeding, more than 99% of the initial whole-body radioactivity was eliminated through gut clearance ($\lambda_1 = 0.94$/day), whereas the remaining fraction (<1%), which represents loss by body tissues of the assimilated radionuclide, was eliminated at a much slower rate ($\lambda_2 = -0.001$/day). By 16 days after feeding, approximately 0.7%, or 113 dpm*/g, of the initial whole-body radioactivity remained.

The low assimilation (0.8%) and rapid initial loss of hexavalent chromium suggest the nonessential nature of the element in cotton rats. The retention data also confirm the lack of any significant bioaccumulation, as shown in analyses of stable chromium (Table 2), and further suggest the reduced probability of a toxic effect through ingestion of drift-contaminated vegetation or inhalation of drift-contaminated air. The estimated fraction absorbed by man (<0.005) through ingestion[18] is similar to the fraction assimilated by the native rate (0.008). Similarly, the biological half-times of chromium assimilated by man[18] and cotton rats are 616 and 693 days, respectively. The magnitude of the halftimes suggests that chromium derived from a chromate has a high potential for biological interactions. However, the fraction assimilated is very low, which reduces the likelihood of toxic effects. The data suggest that extrapolations from small mammals to man are reasonable for chromium.

Drift-Derived Chromium in Arthropods

Arthropods were trapped in an area adjacent to the ORGDP cooling towers (100 m) and in a control area to determine the effects of drift-derived chromium on

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*dpm = disintegrations per minute.

invertebrates. Both sample areas consisted of a 6- by 6-m plot in a fescue grass area. Pitfall traps were located at 1-m intervals (49 per plot) and opened for a total of 490 trap-nights. The arthropods caught in the traps were identified and analyzed for chromium content. Results indicated that there were no differences in the kinds or numbers of arthropods collected, but there were differences in the chromium concentrations within taxa and between plots, with 190 invertebrates (Coleoptera, Orthroptera, and Araneae) being collected in the drift-contaminated area compared to 170 in the control plot. Mean chromium concentration in ground beetles collected next to the tower was 9.05 ± 1.05 ppm in contrast to 0.49 ± 0.02 ppm in controls, while concentration in crickets ranged from 37.63 ± 3.6 ppm for those collected near the towers to 0.81 ± 0.04 ppm in controls. Even though there is elevated chromium concentration in the two orders represented (Orthroptera and Coleoptera), it is hypothesized that the elevated chromium is not assimilated by the organism, but represents the chromium in the gut contents from feeding on drift-contaminated foliage or insects as in the case of the ground beetles.

Effects of Drift-Derived Chromium on Root and Litter Respiration

Fescue roots were collected in the spring and summer of 1976, and litter was collected during the summer to measure respiration. Samples were taken from plots located ~50 m from the K-31 cooling tower. Control samples from 0.05-m^2 plots were collected at a location remote from the towers. Carbon dioxide evolution was measured using an infrared gas analyzer.

The roots collected during the spring showed a slight diel pattern in CO_2 evolution, with increases ranging from 14.5 and 17.1 mg CO_2/(h • g) during the day in drift-contaminated plants and controls, respectively, to 22.0 and 23.3 mg CO_2/(h • g) during the late evening. Similarly, CO_2 efflux rates from roots collected in the summer ranged from minima of 16.1 and 21.1 mg CO_2/(h • g) to maxima of 28.9 and 32.2 mg CO_2/(h • g) between treatment and control plants. Peaks occurred in the early morning and early evening (Fig. 6). The CO_2 efflux rates in litter collected in summer were higher than the CO_2 values recorded for the roots collected in the spring and summer. Carbon dioxide evolution varied from minima of 23.4 to 27.9 mg CO_2/(h • g) between drift-contaminated plants and controls to maxima of 34.1 and 32.7 mg CO_2/(h • g) between the two groups.

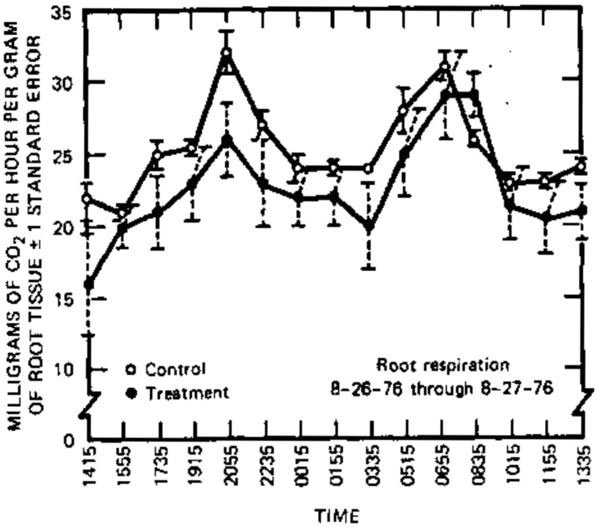

Fig. 6 Carbon dioxide evolution from fescue grass roots grown in drift-contaminated soils and in control soils. Each data point represents mean CO_2 evolution from four replicate samples.

During both seasons the roots and litter collected at 50 m from the cooling tower showed inhibition, although not statistically significant, of respiration (up to 20%) when compared to controls. It is not known if these responses are the result of increased chromium or zinc from drift deposition, decreased microbial activity in the litter as the result of drift toxicants, or micrometeorological effects (i.e., moisture, temperature) produced by cooling tower operation. The data suggest that process measurements may be useful parameters for assessing drift effects in the absence of visual symptoms.

SOILS AND COOLING TOWER DRIFT

Accumulations of Chromium at the ORGDP Site

As indicated in the interception and retention study, from 25 to 75% of the deposition flux to receptors may be deposited directly on the soil surface or continued field losses from foliage may increase soil burdens of drift components. Availability of chromium to plants through uptake from the soil is pH dependent.[19] That portion considered essentially available to plants was determined, with the use of extraction techniques, not to exceed 5% of the total.[20] In nutrient culture experiments, excess quantities of certain chromates[21,22] produce effects ranging from chlorosis, growth inhibition, interference with uptake, and translocation of essential elements to death. Considering the long history of chromate–zinc treatment to the circulating waters of the cooling towers of the ORGDP, the quantity in the soil of the surrounding area was determined to assess possible accumulation over years of continuous deposition.[2]

Analyses of soils at ORGDP within the area of drift deposition indicate only slight increases in chromium above background levels. Total analyses of chromium in soils remote from drift (background) averaged 50 ± 5 ppm. Increased quantities, vis., within the top 6 cm, are evident to a distance of 180 m from the cooling towers, ranging from 10 times background at 15 m to 3 times background at 60 m. Beyond 180 m, the concentrations of chromium are similar to background levels. Chromium concentrations in soils decrease exponentially with increasing distance similar to patterns observed in vegetation. Concentrations considered available to plants, based on extraction with 1*N* ammonium acetate, varied from 1 to 6% of the total. The fraction considered mobile is similar for both the top centimeter and the subsequent 5-cm increment. The percent extractable or mobile is consistent with previous studies. Available chromium to distances of 180 m ranges from 27 to 3 ppm. Toxicity symptoms would not be expected in vegetation except at very close distances, where highly sensitive indicator species, such as tobacco, might develop symptoms of excess quantities.

The apparent absence of any significant accumulation in soils beyond 200 m after 17 years of drift releases from cooling towers suggests that chromium may attain an equilibrium concentration dependent on production level of the facility. Equilibrium distribution coefficients (K_d) were calculated for several concentrations of chromium and zinc in an ammonium acetate extractant adjusted to various pH levels to determine the result of constant input to the soils from drift. The term K_d is defined as the fraction of the element adsorbed per gram of soil divided by the fraction in solution per milliliter. Low K_d values indicate that the soil has little affinity for the element, whereas a high K_d would indicate that the element had been adsorbed. Likewise, a low coefficient would indicate that the element was free to move from the soil system. The K_d for chromate covering a range of expected pH levels in soil is extremely low, ranging from 1 to 6. Distribution coefficients for chromate added to the ammonium acetate extractant at four pH levels show that adsorption to soil is not influenced by changes in acidity. Results are similar to K_d values calculated for chromate added to distilled water. The values indicate that little absorption of hexavalent chromium occurs in the soil; instead, chromium would be highly mobile and would be readily transported from the terrestrial system to surface streams or ponds. However, an accumulation of chromium in soils would be expected, provided that the potential for reduction from hexavalent to trivalent forms does indeed exist. The fraction of chromium extractable from soil is probably in the Cr^{6+} form. The slight accumulations (total analysis) to a distance of 180 m likely represents the fraction of chromate drift reduced to the chromic species (Cr^{3+}) after more than 20 years of tower operation. Once reduced, the chromic oxide would be of little biological concern owing to its inert nature.[19]

Movement of Drift-Derived Chromium in Soil Water

Liquid effluents from the Paducah Gaseous Diffusion Plant flow into the Ohio River via Little Bayou and Big Bayou creeks. Little Bayou Creek is surrounded by government property (Department of Energy and Tennessee Valley Authority) along its entire length, whereas Big Bayou Creek passes through privately owned farmland. The National Pollutant Discharge Elimination System (NPDES) permit specifies allowable concentrations for pollutants in plant discharges. To meet the discharge limit of 0.05 ppm hexavalent chromium, blowdown from the four recirculating cooling water systems (~1.8 m^3/min) is delivered to the Plant Liquid Effluent Pollution Control Facility. This facility was designed to decrease the concentration of total chromium in Big Bayou and Little Bayou creeks by reducing the hexavalent chromium to the trivalent state and subsequently precipitating it in a clarifier using lime. The clarified water then passes to a 98×10^3 m^3 lagoon for equalization and further settling before it is discharged to Big Bayou Creek. When the Paducah Gaseous Diffusion Plant is fully uprated (1981), with a blowdown rate of 4.7 m^3/min, the lagoon will provide a 14-day retention time.

The performance of the facility prior to the addition of new cooling towers in the fall of 1977 indicated that the facility was operating at design specifications with chromium concentrations in Little Bayou and Big Bayou creeks within the NPDES limit of 0.05 ppm. However, even with all blowdown diverted to the Plant Liquid Effluent Pollution Control Facility, there still remained a potential for chromium concentrations in nearby Little Bayou Creek to exceed NPDES limits either by direct deposition of drift water or from the transport of drift-derived chromium by soil moisture flow; for example, the soils of the area of the Calloway and Henry series, developed from loess deposits and characterized by (impermeable) fragipan from 10 to 70 cm below the surface. This edaphic feature could provide a significant pathway for the contamination of Little Bayou Creek by channelization of drift-contaminated soil water along the fragipan.

In order to test this hypothesis, soil water samples were collected along a distance gradient from the cooling tower to Little Bayou Creek at 1300 m. Samples were collected at the hardpan (~60 cm deep) and one-half the distance from the hardpan to the surface (~30 cm) 24 and 48 h following a rainstorm.[4]

The analytical results suggest that the chromium concentration in soil water (shallow wells) at 60 and 200 m from the tower is greater than the concentration at the hardpan. However, linear regression analysis between concentration and depth indicated that there was no correlation. Subsequently, sample data were combined (disregarding depth) and analyzed by distance, using a one-way analysis of variance. The results indicated that there was a significant difference (5% level) between the mean concentration at 60 m and that at other distances, but failed to distinguish the differences between those distances. The data were further analyzed according to Duncan's multiple-range test by logarithmic transformation to stabilize the variance. The results (Table 3) identify the differences in chromium concentration in soil water between 60 and 1260 m from the cooling tower. The concentration at 60 m (0.112 ppb) is significantly different ($P < 0.05$) from the concentrations at all other distances, while the concentrations at 200, 950, and 1260 m are different from that at the 410-m distance. The concentration at 410 m (0.029 ppb) was the lowest along the gradient, but not statistically different from the concentration at the most distant plot (1260 m).

Composite soil samples were taken from each well at the two depths and analyzed for texture, pH, and organic matter content. Chromium concentration in litter, ranging from 123.8 ± 7.8 ppm at 60 m to 1.2 ± 0.1 ppm at 1260 m, was indicative of drift deposition along the distance gradient to the Little Bayou Creek. Surprisingly, the total amount of chromium in the soil indicated that there was very little accumulation even at close-in distances. Among the soil properties determined, only the organic matter content offers any plausible explanation for the lack of accumulation. In the presence of organic matter, hexavalent chromium is easily reduced and subsequently bound to organic particles as trivalent chromium. The kinetically inert trivalent chromium is the most stable oxidation state of chromium and therefore would accumulate with chronic deposition. The low organic matter content (0.5 to 1.0%) of the Paducah soils (Calloway and Henry series) has a low potential for chemical reduction of hexavalent chromium to the less soluble trivalent species. Drift-derived chromium in the soils is likely continuously removed through weathering (rain and runoff). At 60 m total soil chromium (69.3 ± 2.3 ppm) was 15% greater than controls (60.6 ± 1.9 ppm) at 1260 m.

Table 3 Duncan's Multiple-Range Test of Chromium Concentration in Soil Water Along a Distance Gradient

Distance, m	No. of samples	Mean concentration of log transformed data	Groupings*
60	15	−2.415900	A
950	18	−3.089200	B
1260	18	−3.185600	B, C
200	16	−3.493200	B, C
410	14	−3.602700	C

*Means with same letter are not significantly different ($P < 0.05$).

At 60 m from the cooling tower, there is a higher chromium concentration (0.112 ppb) in soil water than at 1260 m (0.05 ppb) near Little Bayou Creek. Even though the increase in concentration at 60 m is statistically significant, it is several orders of magnitude lower than the NPDES limit of 0.05 ppm. It should be noted that the concentrations are within or below the range reported for spring water on the Walker Branch watershed at Oak Ridge.[23] It is concluded from this study that the transport of drift-derived chromium in soil water is not a significant pathway for introducing chromium into the stream.

CONCLUSIONS

The studies summarized here were designed to answer specific questions concerning the impacts of chromated drift from mechanical-draft cooling towers at DOE facilities and have provided pioneering evidence of the transfer of chemicals in drift to the terrestrial environment. The general impressions from the investigations indicate that many potential environmental impacts either have not materialized or have been overlooked, since the studies reflect tower operation during a relatively short time period (5 years) and the towers have been operational for nearly three decades. Similar conclusions have been drawn concerning saline drift from natural-draft cooling towers.[24-27] In cases where adverse effects are demonstrated, it must be stressed that these occur from 50 to 200 m from the tower basin.

During the time spanning these investigations, additional problem areas to be researched have been recognized. For example, a major concern is the direct depositon or runoff of chromated cooling water to surface streams and, consequently, the possible noncompliance with water standards. The infrequent addition of exotic chemicals (i.e., sodium pentachlorophenate, chromated copper arsenate, and acid copper chromate) to the recirculation cooling water for wood preservation poses a potential for ecological and health effects. Quantification of these compounds in drift following treatment (every 3 to 5 years) needs to be determined at some site.

Another important inference from these studies is that drift effects can be minimized by proper selection of sites for the towers and the use of reliable and well-maintained equipment during tower operation. The results of the research summarized are site specific but have provided guidance applicable to generic studies in monitoring design, sample collection, and assessment of drift effects in ecosystems. The results of the research have proven valuable to environmental protection managers in upgrading monitoring protocols at their respective DOE facilities and have contributed to the evaluation and assessment of environmental impacts related to plant operation.

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