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Assessment of the Influences of Groundwater Colloids on the Migration of Technetium-99 at the Paducah Gaseous Diffusion Plant Site in Paducah, Kentucky

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Environmental Sciences Division
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ASSESSMENT OF THE INFLUENCES OF GROUNDWATER COLLOIDS
ON THE MIGRATION OF TECHNETIUM-99 AT
THE PADUCAH GASEOUS DIFFUSION PLANT SITE IN PADUCAH, KENTUCKY

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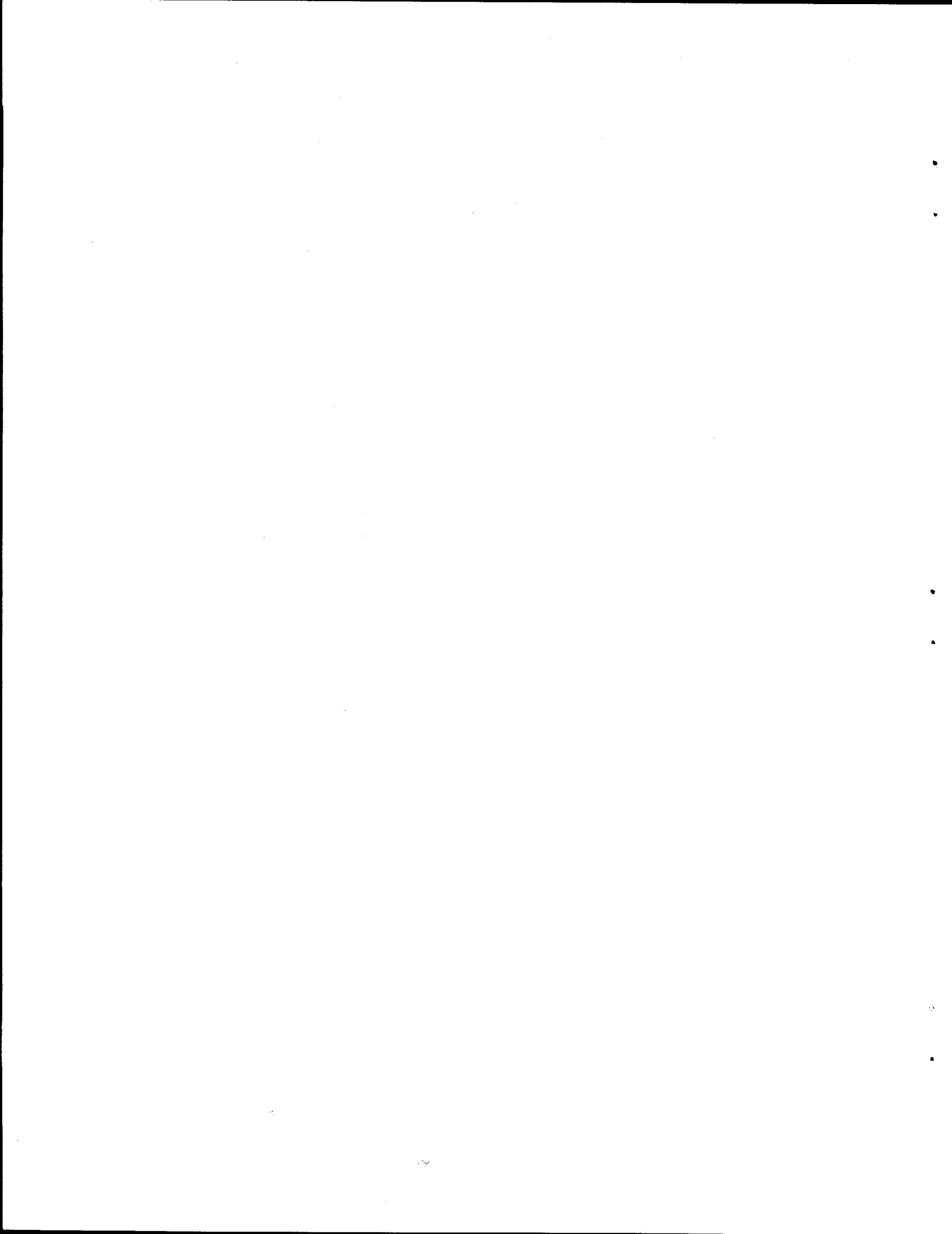
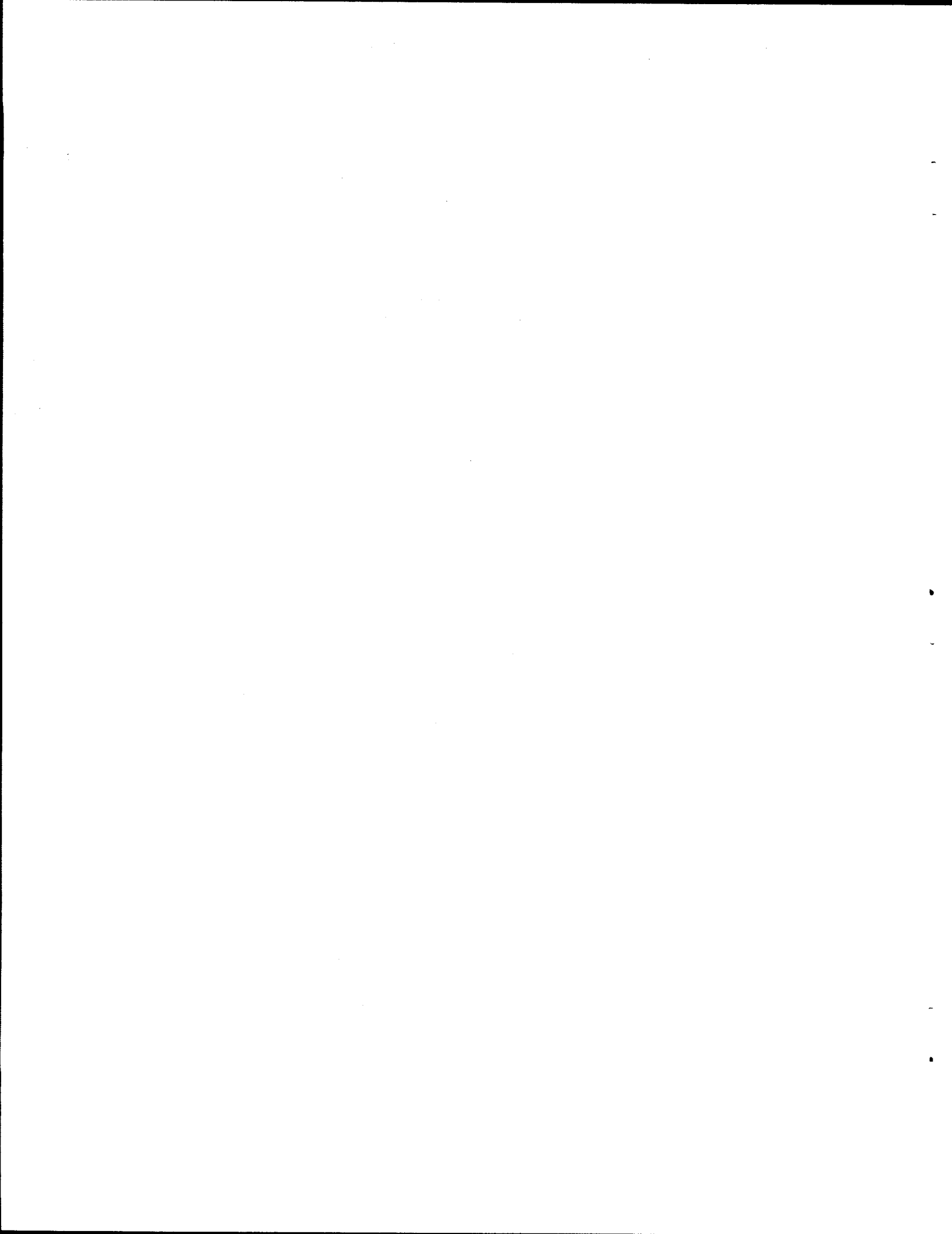


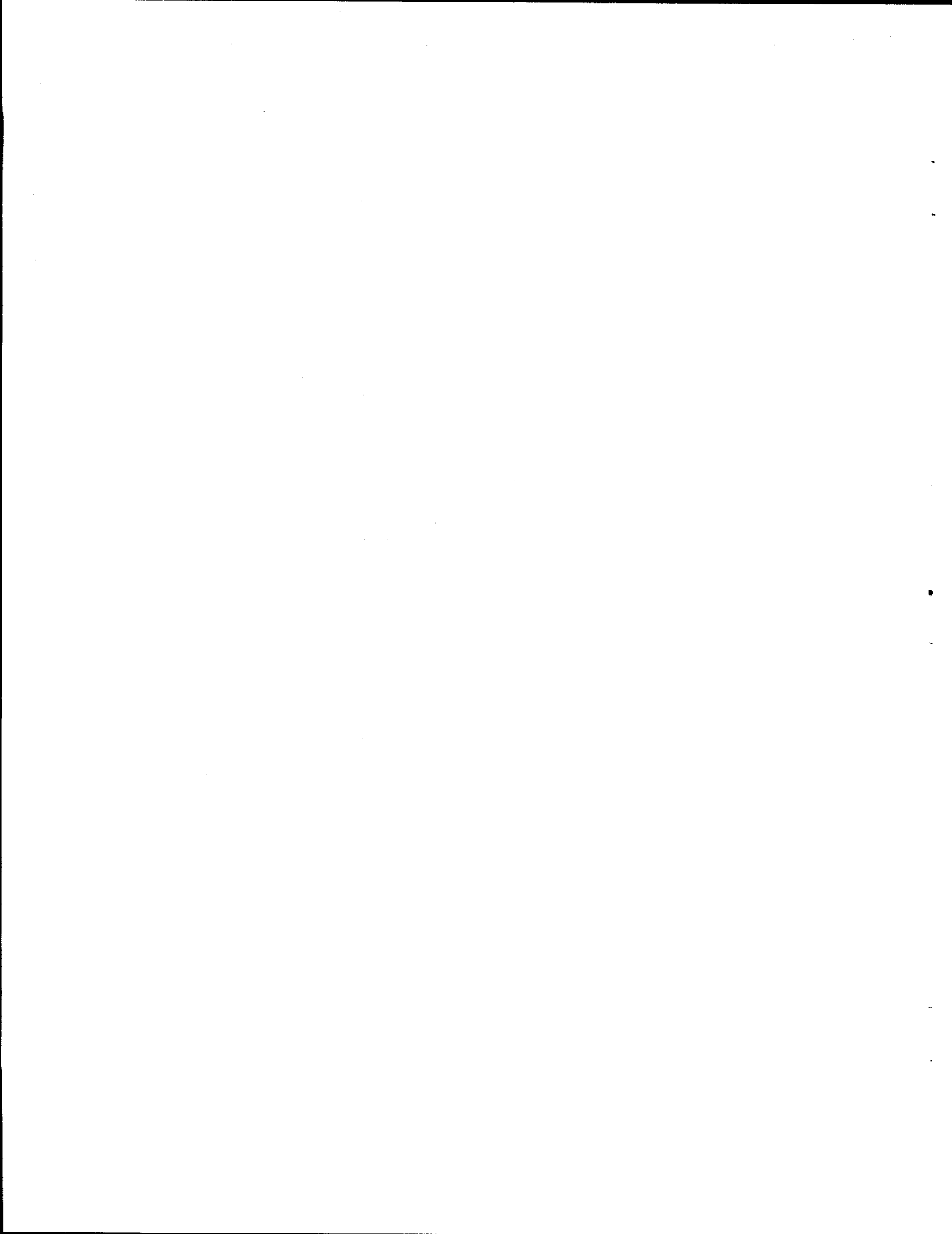
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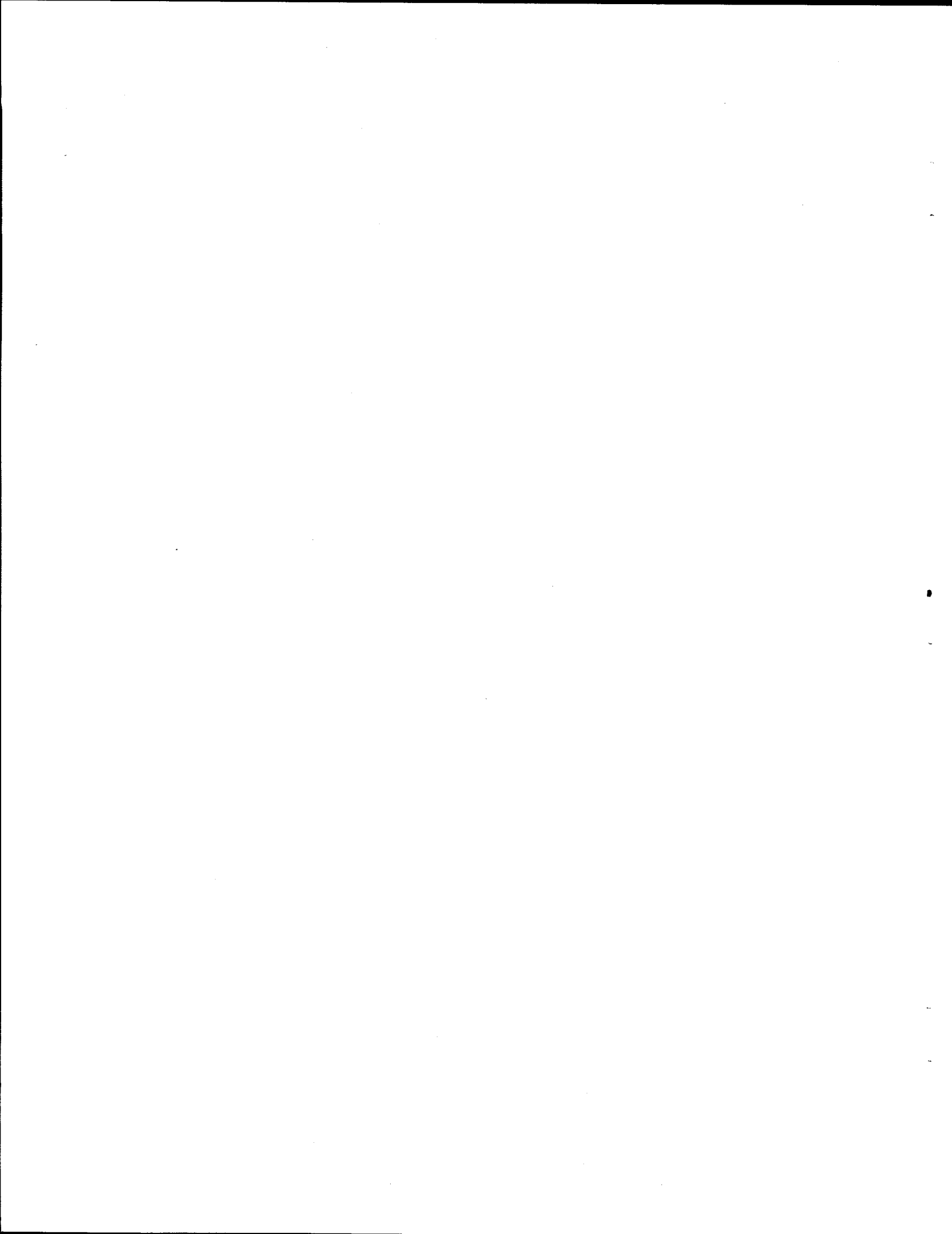
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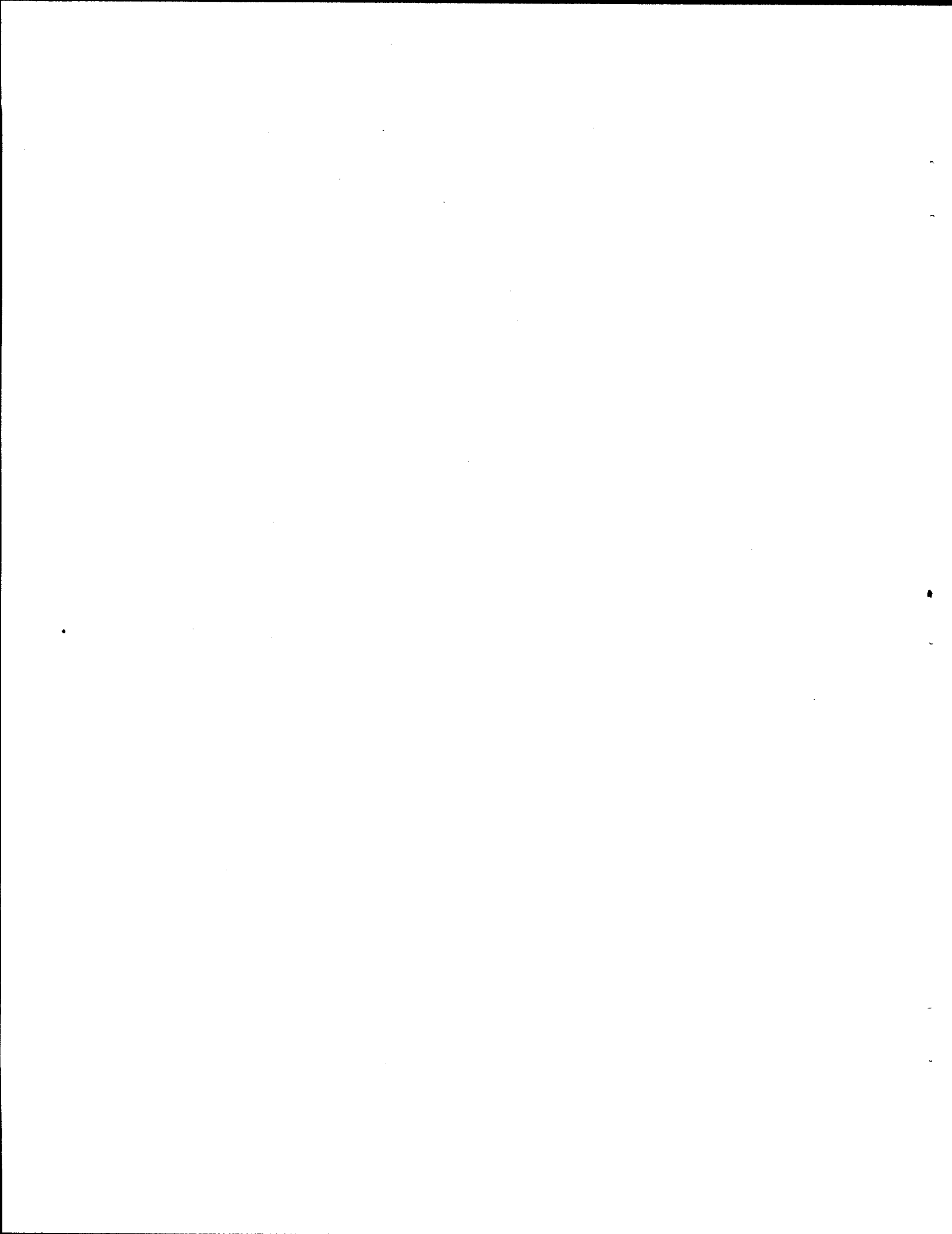
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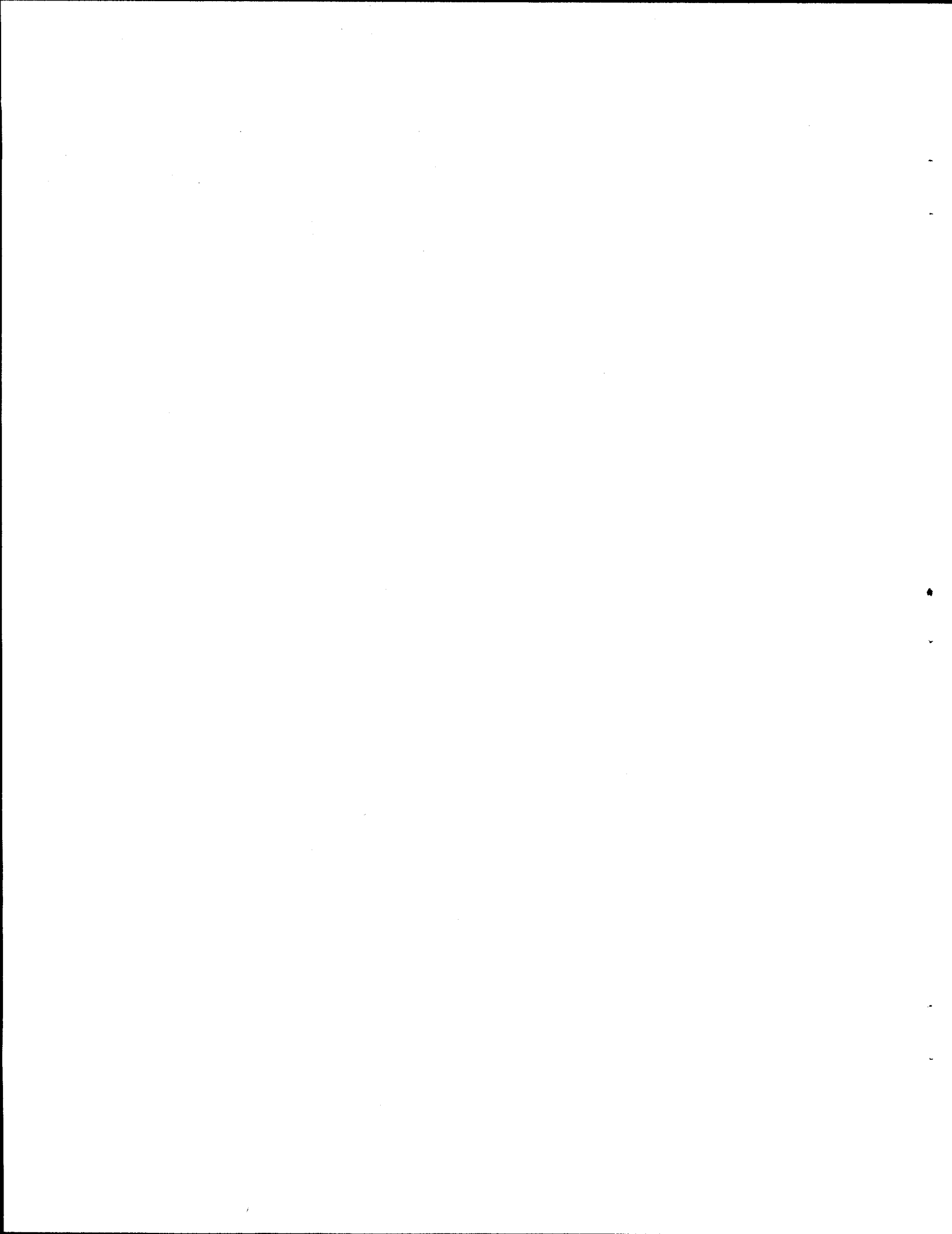
ABBREVIATIONS

DO	dissolved oxygen
DTPA	diethylenetriaminepentaacetic acid
EDTA	ethylenediaminetetraacetic acid
HS	humic substances
MW	monitoring wells
NOM	natural organic matter
PGDP	Paducah Gaseous Diffusion Plant
⁹⁹ Tc	technetium-99
TCE	trichloroethylene



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ABSTRACT

This short report summarizes the influences of groundwater colloids on the migration/transport of ^{99}Tc at the Paducah Gaseous Diffusion Plant (PGDP) site in Paducah, Kentucky. Limited data suggest that inorganic colloidal materials (e.g., aluminosilicate clay minerals) may not play a significant role in the retention and transport of Tc. Studies by size fractionation reveal that both Tc and natural organic matter (NOM) are largely present in the <3K fraction (equivalent to ~ 1 nm in diameter). Therefore, the role of NOM on Tc retention and transport is not conclusive on the basis of this study. However, a literature review suggests that Tc is very likely associated with the groundwater organics. The presence of the organic matter could have increased the solubility and cotransport of Tc at the PGDP site. It is recommended, therefore, that further studies, applying such techniques as gel chromatography, size exclusion, and spectroscopy, may be useful in order to determine the association of organic matter with Tc. If Tc is associated with groundwater organics, the appropriate protocols for removal of organic matter associated with Tc may be developed.

It is also pointed out that, because time and resources were limited, this study is not comprehensive with respect to the role of mobile organic and inorganic colloidal materials on Tc transport in subsurface soils. The redox conditions (DO) of groundwaters reported may not represent the true groundwater conditions, which could have influenced the association and dissociation of Tc with groundwater colloidal materials. Because Tc concentrations in the groundwater (on the order of nCi/L) at the PGDP site is much lower than the solubility of reduced Tc (IV) (on the order of $\sim 10^{-8}$ mol/L or parts per billion), regardless of the redox conditions, Tc will stay in solution phase as Tc(IV) or Tc(VII). The mechanisms of adsorption/association vs precipitation must be understood under reduced and low Tc conditions so that strategic plans for remediation of Tc contaminated soils and groundwaters can be developed.

1. INTRODUCTION

Data on radioactivity in groundwater monitoring wells at the Paducah Gaseous Diffusion Plant (PGDP) have raised concern over the possibility that some of the observed transport of ^{99}Tc may involve the cotransport of Tc sorbed onto mobile inorganic colloids or on natural organic matter (NOM), which can complex and potentially cotransport Tc.

The goal of groundwater monitoring is to determine the concentration of contaminant that is mobile in the aquifer. Is the contaminant present as a dissolved solute, adsorbed on a mobile colloid, or complexed with NOM in the groundwater? It is important to recognize which of these alternate transport processes is responsible for Tc migration. An assessment of groundwater quality and a decision on the appropriate action at the site need to be based on the correct description of the mechanisms of Tc transport in the aquifer. Chemical and physical descriptions of the transport of Tc as a solute are fundamentally different from that of Tc sorbed to colloids or to NOM; in the latter cases, the transport of Tc is controlled by the behavior of the colloids or the NOM macromolecules - not the chemistry of Tc itself. Failure to recognize the appropriate transport mechanism can lead to erroneous predictions concerning the extent of transport and distribution of contaminants within the aquifer. Furthermore, appropriate descriptions of Tc transport mechanisms may reveal novel opportunities for remediation. If Tc is transported mostly by colloids, for example, Tc may be confined primarily to the more transmissive zones of the aquifer, thereby making pump-and-treat remediation or source control options more attractive.

The primary objective of this study was to assess if Tc is associated with mobile organic and inorganic colloidal materials. If Tc is associated with colloids, additional tasks may be considered to determine the scope of the problem at the PGDP site and to develop appropriate protocols for remediation of colloid-bound Tc.

2. APPROACH

2.1 The Monitoring Wells

Three monitoring wells (MWs), namely MW 66, MW 201, and MW 202, that may represent Tc plumes at Paducah (Figs. 1 and 2) were selected. (*Note: The initial plan to sample four monitoring wells was not accomplished because several unexpected interruptions occurred during field work*). The MW 66 was an old well installed in 1986, whereas MW 201 and MW 202 were relatively new, having been installed in 1991. Table 1 shows some physical data about these monitoring wells. All the wells were equipped with Well Wizard bladder pumps and packers.

2.2 Groundwater Sampling

Groundwater samples were taken after at least 2 h of initial purging to discard the stagnant water in the well bore and to obtain water samples that were representative of the formation water. The time required for purging was determined when several key physicochemical parameters of the recovered water leveled off. These parameters include pH, dissolved oxygen (DO), turbidity, specific conductivity, and temperature. The turbidity was measured by a portable turbidity meter, whereas pH, DO, specific conductivity, and temperature were monitored by a flow-through Hydrolab sensor. It should be pointed out, however, that all the wells showed relatively high DO concentrations [from ~3 to 5 parts per million (ppm)], probably because of the puff-and-huff (or pumping and sucking) action of the bladder pump so that the recovered water was in fact coming into contact with air (or oxygen). Therefore, the measured DO does not represent the actual DO of the groundwater.

MW 201 showed an unusually high turbidity, probably due to insufficient well development. An attempt to decrease the flow rate even made it worse (turbidity substantially increased as the flow rate decreased). Therefore, the final flow rate was maintained at 1200 mL/min, and the turbidity was 145 ± 10 .

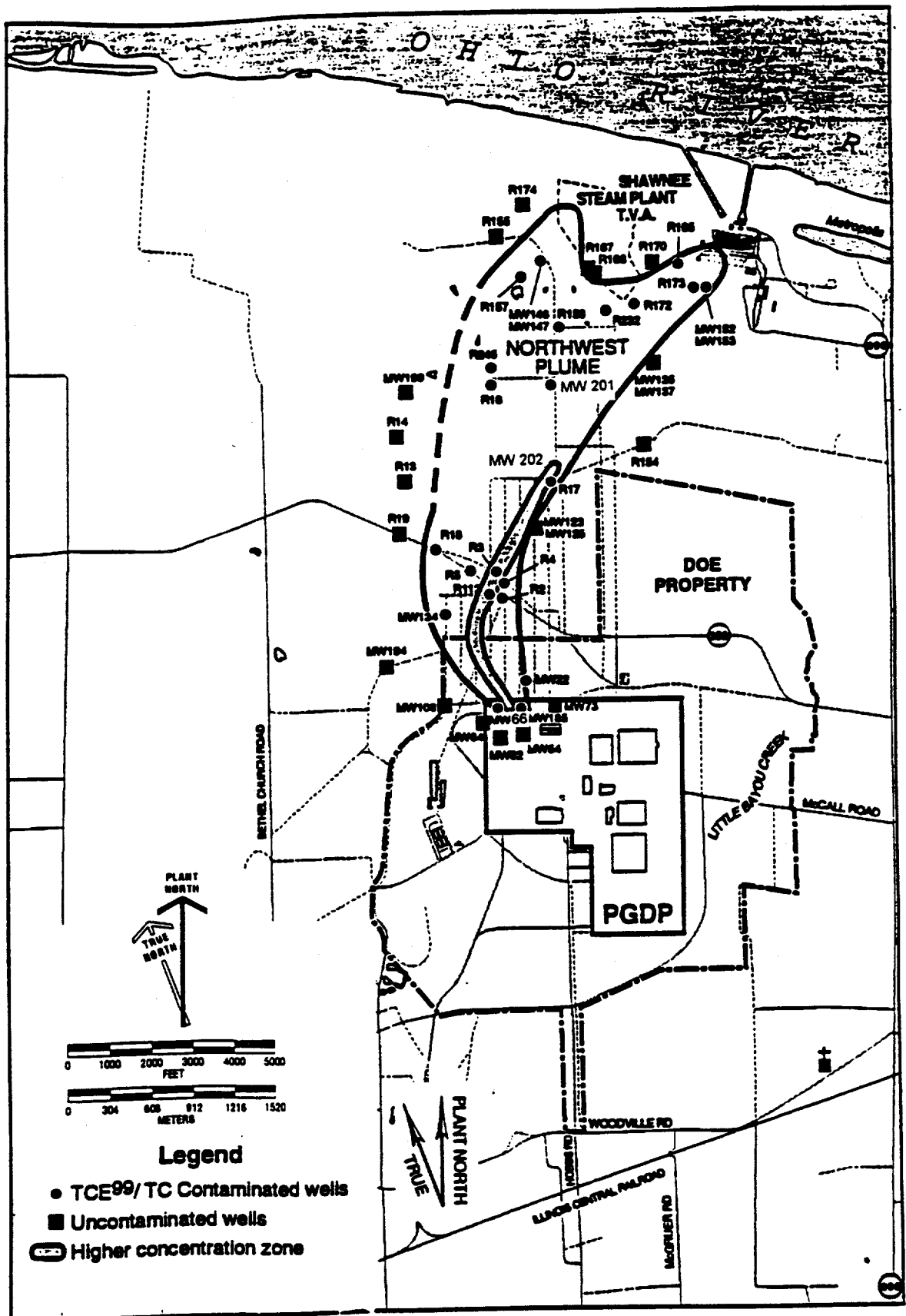


Fig. 1. The location of the northwest ⁹⁹Tc and trichloroethylene (TCE) plume at the Paducah Gaseous Diffusion Plant site in Paducah, Kentucky (from Clausen and Richards 1994).

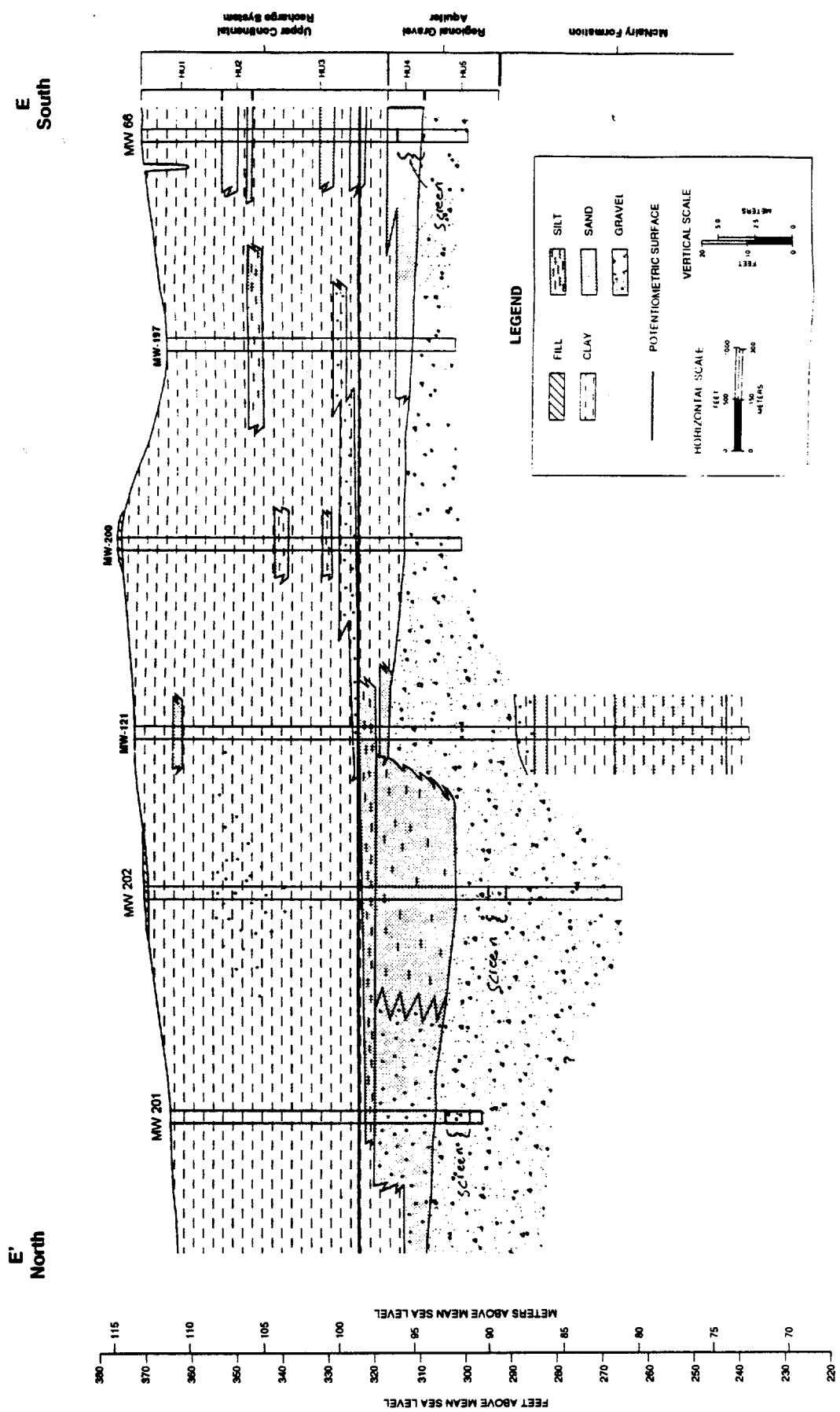


Fig. 2. Geological cross section of E - E' (from Clausen et al. 1993).

Table 1. Some physical data of the monitoring wells (MWs)

	MW 66	MW 201	MW 202
Elevation (ft)	371.1	366.3	372.6
Water table (ft)	43.1	41.4	46.5
Top of screen (ft)	55.2	62.5	80.1
Bottom of screen (ft)	60.2	67.5	86.1
Top of sand pack (ft)	53	56.5	72
Bottom of sand pack (ft)	64	70	85
Casing diameter (in.)	2	2	2
Casing material	Stainless steel	Stainless steel	Stainless steel

2.3 Size Fractionation

In order to determine if natural organic and inorganic colloidal materials in groundwater play a role in Tc mobility and transport, a series of size fractionations were performed by using Amicon hollow-fiber filters. The permeates from a series of filters at nominal molecular weight cutoffs of 3,000 and 100,000 dalton [corresponding to particles ~1 and 5 nm in diameter) and a 100-nm filter were collected and preserved by acidification for Tc, gross β , gross α , and organic carbon analysis. Raw water was also collected for Tc, gross β , gross α , organic carbon, cation, and anion analysis. Total suspended solids in the groundwater were also determined by gravimetrically collecting on a 0.45- μ m membrane filter. It should be noted, however, that on-line filtration did not occur; rather, filtration was performed after the groundwater was collected in a flask. This is because a positive pressure could not be maintained during the filtration again because of the problems of the bladder pumps. As indicated earlier, the redox of the collected water did not represent the actual groundwater.

2.4 Analysis

Tc, gross β , and gross α were analyzed at Paducah. Tc was determined by methyl ethyl ketone extraction followed by liquid scintillation counting. Gross β was determined by a direct evaporation of 200 mL of water sample and counted on a gas-flow proportional counter. Total nonpurgeable organic carbon was determined by a Shimadzu TOC-5000 Analyzer (Shimadzu Co., Japan). Because all three groundwater samples contained very low organic carbon concentrations (< 0.5 ppm), these water samples were also concentrated about 10-fold on a Speed Vac and reanalyzed for total organic carbon concentrations to reduce the analysis error.

3. RESULTS

3.1 Concentration Distributions of ^{99}Tc

The concentration distributions of Tc in three different size fractions are illustrated in Fig. 3. First of all, MW 66 contained the highest Tc concentration, ~1-2 orders of magnitude higher than the Tc concentrations of MW 201 and MW 202, because MW 66 is closest to the PGDP site (or the source of contamination). For the same reason, MW 66 showed a higher activity relative to that of MW 201 and 202 (Table 2).

By examining the Tc concentration distributions in the three size fractions, we noticed that the Tc concentration in the <3K fraction (~1700 pCi/L) was lower than that of the <0.1- μm and <100K fractions (~2250 pCi/L) for MW 66, whereas there are no significant fractionations in the distribution of Tc in MW 201 and MW 202 within the experimental error.

However, it is also noted that Tc concentration in the raw water (unfiltered) of MW 66, which is lower than the concentrations of <0.1- μm and <100K size fractions, seems odd. As a precaution, Tc concentrations were reanalyzed in those fractions. The same results were obtained, which indicated that the observation was not because of an analysis error. There are no satisfactory explanations for this observation, although this may be attributed to coprecipitation/sedimentation or adsorption of Tc with amorphous Si or its associated carbonates, or Ca-carbonate during storage (or before analysis), given the relatively high concentrations of Si, Ca, and alkalinity in the groundwater (Table 3). Of course, since only one data point is available, any further speculation may not be very helpful.

Nevertheless, given the experimental and analytical errors and limited numbers of monitoring wells analyzed, we conclude that Tc may not be significantly associated with groundwater colloids, at least for those of >3K organic and inorganic materials. Note that for MW 201, although Tc in the size fraction of <100K (but >3K) was not determined, our conclusions shall not change.

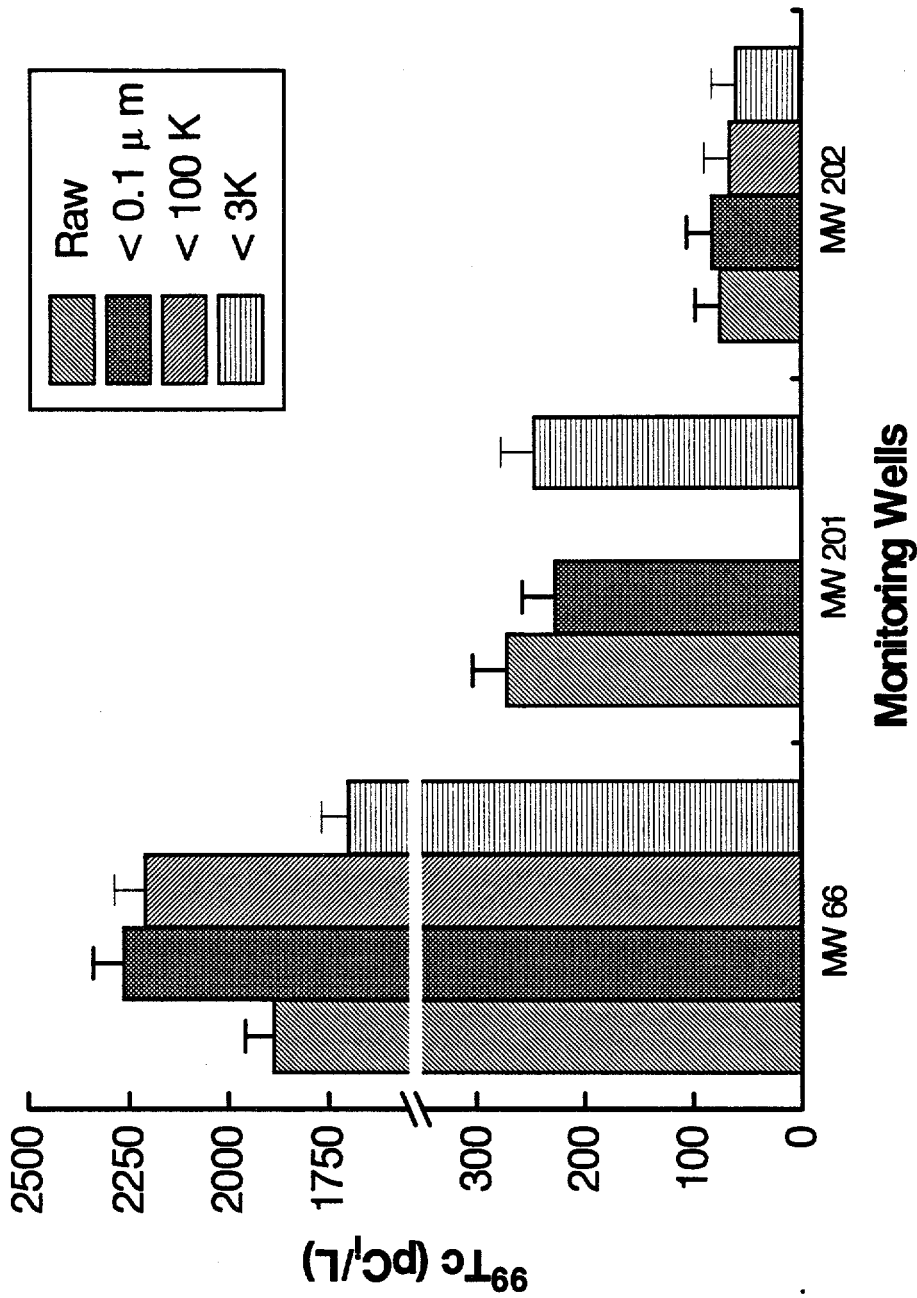


Fig. 3. Distributions of ^{99}Tc in different size fractions of three groundwater samples at the Paducah Gaseous Diffusion Plant site in Paducah, Kentucky.

Table 2. Gross α and β in different size fractions of groundwaters sampled between December 13 and December 16, 1993, in Paducah, Kentucky

Wells	Filter size	Gross α (pCi/L)	Gross β (pCi/L)
MW 66	Raw	64.3 \pm 7.1	1319 \pm 26
	0.1 μ m	70.0 \pm 7.0	1130 \pm 23
	100K	67.2 \pm 6.7	1488 \pm 30
	3K	70.4 \pm 7.0	1397 \pm 28
MW 201	Raw	13.8 \pm 3.3	300 \pm 15
	0.1 μ m	6.9 \pm 1.8	169 \pm 10
	3K	10.2 \pm 2.4	219 \pm 11
MW 202	Raw	4.1 \pm 1.3	59 \pm 5
	0.1 μ m	3.0 \pm 1.0	58 \pm 5
	100K	-0.7 \pm 0.3	52 \pm 5
	3K	0.9 \pm 0.3	52 \pm 5

Table 3. Some physical and chemical properties of the unfiltered groundwaters

Element	MW 66	MW 201	MW 202
Alkalinity (mg/L)	84	99	75
Cl ⁻ (mg/L)	30	36	23
NO ₃ ⁻ (as N) (mg/L)	5	2.6	2.1
SO ₄ ²⁻ (mg/L)	11	16	12
Si (mg/L)	15	23	22
Al (mg/L)	0.13	9.13	0.67
Fe (mg/L)	0.15	40.6	1.19
Ba (mg/L)	0.17	0.33	0.15
Ca (mg/L)	30.9	24.4	18.1
Mg (mg/L)	7.74	9.79	7.7
Na (mg/L)	18.4	33.7	24.7
K (mg/L)	< 2	2.91	< 2
Mn (mg/L)	0.02	1.16	0.06
pH	6.1	6.1	6.1
DO ^a (mg/L)	3.8	3.96	4.32
Turbidity (NTU ^b)	2.0	360	17
Temp. (°F)	58	57	55

^a DO = dissolved oxygen

^b NTU = nephelometric turbidity unit

3.2 Organic Carbon Distributions

The concentration distributions of organic carbon in the three size fractions are plotted in Fig. 4. In general, groundwater samples in the three monitoring wells contained the nonpurgeable organic carbon between 0.2 and 0.5 mg/L, as commonly observed for many groundwaters. Within the experimental error, no significant differences are observed for organic carbon distributions in the three size fractions. This indicates that the organic matter in these groundwaters consists of primarily low molecular weight compounds.

The association of organic matter and Tc and the role of organic matter in Tc transport are, therefore, nonconclusive, because both organic carbon and Tc are largely present in the <3K fraction (Figs. 3 and 4). Other experimental techniques and further studies must be performed in order to assess the role of groundwater organic matter in Tc migration and transport.

However, a review of literature data does suggest that low molecular weight organic matter (or ligands) may form complexes with Tc, especially the reduced forms of Tc and, therefore, increase the solubility of Tc (Wildung et al. 1986). Pilkington (1990) studied the solubility of hydrated TcO_2 and found that pH had little effect on the measured solubility of Tc over the range of 1 to 12.5. On the other hand, the presence of small amounts of organic compounds increased the measured solubility of Tc by about a factor of 10. Wolfrum and Bunzl (1986) reported that sorption of Tc by humic substances (HS) increased when the concentration of $CaCl_2$ or the amount of dissolved oxygen was decreased. Two mechanisms of Tc sorption were suggested by those authors: (1) complexation with organic matter fractions, and (2) reduction to less soluble species. The evidence of complexation between HS and Tc can be illustrated by the fact that reduced Tc species (HS-Tc complexes or precipitates) are not resolubilized by complexing agents such as ethylenediaminetetraacetic acid (EDTA) or diethylenetriaminepentaacetic acid (DTPA), which are known to form stable Tc complexes (Stalmans et al. 1986). This also indicates that Tc forms even more stable complexes with HS than with those of EDTA and DTPA. Similarly, Henrot et al. (1989) studied the sorption and sequential extraction of Tc on soils. They found that a large fraction of the Tc sorbed was

associated with soil organic matter, and the predominant mechanism of Tc sorption is due to complexation between Tc and functional groups (e.g., COOH) of soil organic matter. They also reported that Tc was predominantly associated with the low molecular weight fractions of the organic matter, presumably because of their abundance of carboxyl and hydroxyl functional groups.

Therefore, given the above fact, it might be reasonable to conclude that the observed Tc in the three monitoring wells may be associated with organic materials present in the groundwaters (<3K). Further studies that apply such techniques as gel chromatography, size exclusion, and sequential adsorption and extraction are recommended in order to elucidate if the groundwater organics are associated with Tc at the PGDP site.

3.3 Total Suspended Solid in the Groundwaters

Fig. 5 shows the total suspended solid (<0.45 μm) concentrations in the three monitoring wells. MW 201 showed an unusually high suspended solid concentration, which was attributed to insufficient well development. Nevertheless, even with such a high solid concentration, Tc did not appear to be associated with those colloidal materials [presumably a mixture of aluminosilicate minerals and iron (hydr)oxides]. Tc concentrations in the three fractions studied (raw, <0.1 μm , and <3K for MW 201) were about the same within the experimental error. This observation suggests that aluminosilicate clay minerals or mixed colloids of clay and iron (hydr)oxides do not play a significant role in Tc retention and transport in the subsurface soil environment. Clay minerals and iron/aluminum-(hydr)oxides coated with organic matter in natural groundwater are known to be negatively charged (Gu et al. 1994), which may explain their low ability to retain Tc (as TcO_4^-). Similarly, Henrot et al. (1989) reported that Bt horizon soil with low organic matter showed a very low sorption potential for Tc. Routson et al. (1977) studied the sorption of Tc (as TcO_4^-) on two extremely weathered soils and found that Tc was very poorly sorbed by the soils.

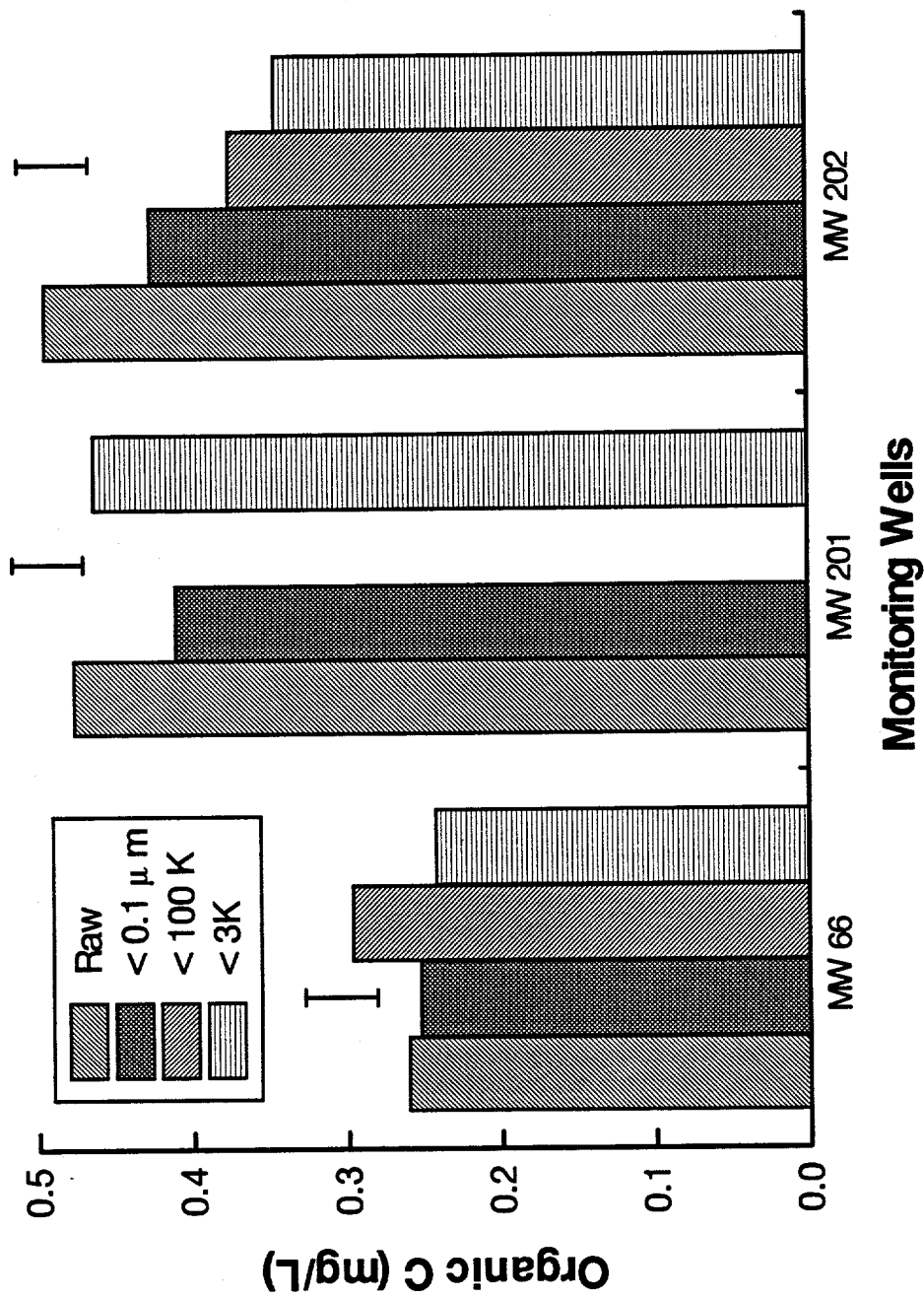


Fig. 4. Distributions of organic carbon in different size fractions of three groundwater samples at the Paducah Gaseous Diffusion Plant site in Paducah, Kentucky.

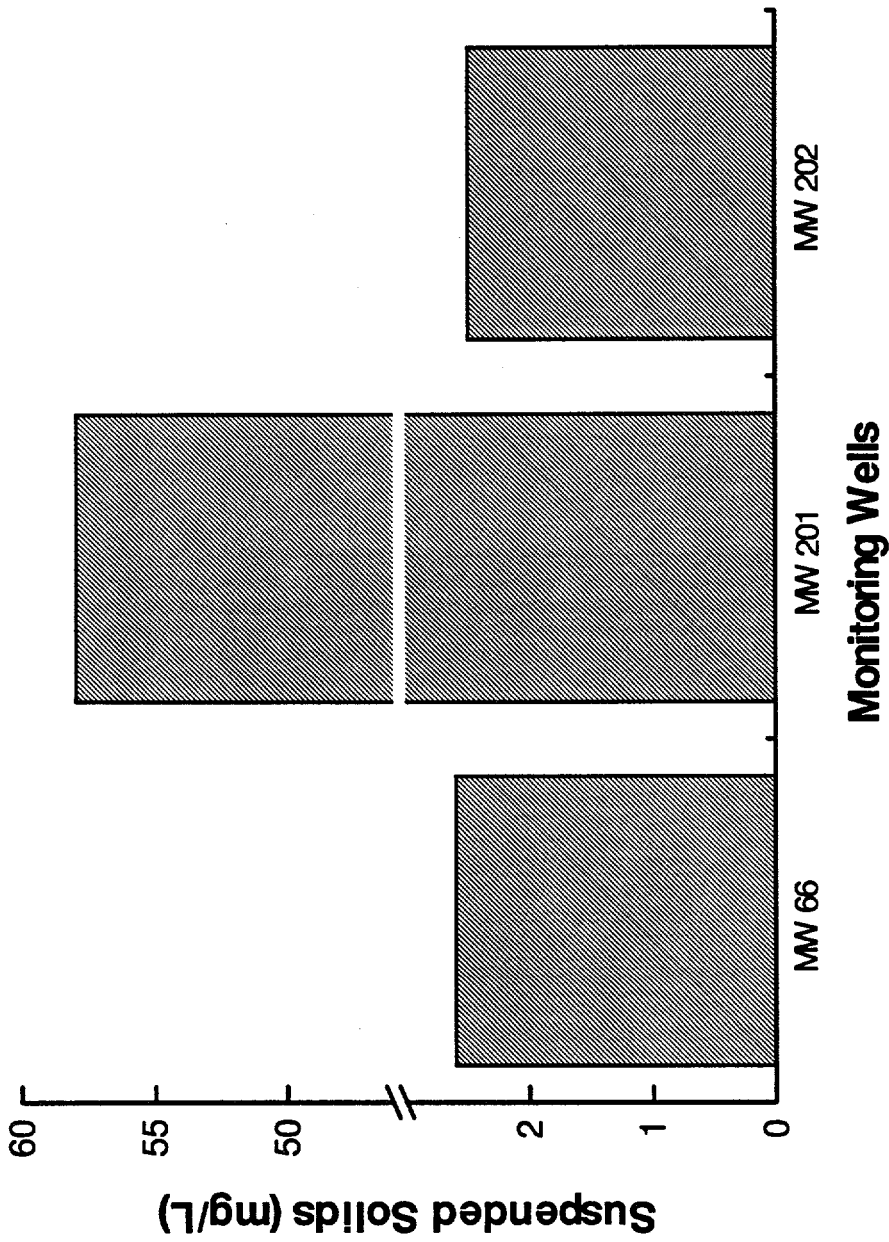


Fig. 5. Total suspended solid concentrations in three groundwater samples from the Paducah Gaseous Diffusion Plant site in Paducah, Kentucky.

4. SUMMARY AND RECOMMENDATIONS

This short report summarizes the influences of groundwater colloids on the migration/transport of Tc at the Paducah Gaseous Diffusion Plant site in Paducah, Kentucky. Limited data suggest that inorganic colloidal materials (e.g., aluminosilicate clay minerals) may not play a significant role in the retention and transport of Tc. Studies by size fractionation reveal that both Tc and NOM are largely present in the <3K fraction (equivalent to ~1 nm in diameter). Therefore, the role of NOM on Tc retention and transport is not conclusive on the basis of this study. However, a literature review suggests that Tc is very likely associated with the groundwater organics. The presence of the organic matter could have increased the solubility and cotransport of Tc at the PGDP site. It is recommended, therefore, that further studies, applying such techniques as gel chromatography, size exclusion, and sequential adsorption and extraction, may be useful in order to determine the association of organic matter with Tc. If Tc is associated with groundwater organics, the appropriate protocols for removal of organic matter associated with Tc may be developed.

Again, because time and resources were limited, only three monitoring wells were analyzed in this study. Therefore, this study is not comprehensive with respect to the role of mobile organic and inorganic colloidal materials on Tc transport in subsurface soils. The redox conditions (DO) of groundwaters and the high turbidity of MW 201 reported here may not represent the true groundwater conditions, as has been noted earlier. The actual DO concentration may be lower than what we observed, and this may have influenced the association and dissociation of Tc with groundwater colloidal materials because Tc is very sensitive to redox conditions (Gu and Schulz 1991). To date, no studies have actually examined the association and/or adsorption of reduced Tc(IV) on minerals and organic colloids. Most studies were performed at high DO conditions (Tc as TcO_4^-), or studies were performed at high Tc concentrations so that TcO_2 precipitation [as Tc(IV)] dominates under reduced conditions. In reality, we encounter much lower Tc concentrations in the groundwater (on the order of

nCi/L) such as at the PGDP site. This concentration level is much lower than the solubility of reduced Tc(IV), which is on the order of $\sim 10^{-8}$ mol/L or parts per billion (Meyer et al. 1986, 1987; Pilkington 1990). In other words, regardless of the redox conditions, Tc will stay in solution phase as Tc(IV) or Tc(VII). As a matter of fact, the presence of groundwater organics may further enhance the solubility of Tc(IV).

Therefore, creating a reducing environment is unlikely to cause the precipitation and removal of Tc in the groundwaters at the PGDP site (given the low concentrations of Tc and its complexation with NOM). However, the reduced Tc(IV) may exhibit a higher potential (or affinity) for being associated with minerals, iron (hydr)oxides, and soil organics than that of TcO_4^- (VII) (yet to be evaluated). The mechanisms of adsorption/association vs precipitation must be understood under reduced and low Tc conditions so that strategic plans for remediation of Tc contaminated soils and groundwaters can be developed.

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