International Journal of Science, Strategic Management and Technology
Volume **02** Issue **03** March-**2026** | ISSN: **3108-1762 (Online)** | Impact Factor: 3.8
An International, Peer-Reviewed, Open Access Scholarly Journal Indexed in recognized academic databases

Transport and Fate of PFAS and Micro/Nanoplastics in Groundwater

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https://doi.org/10.55041/ijsmt.v2i3.356

Cite this Article: Hembram, S. (2026). Transport and Fate of PFAS and Micro/Nanoplastics in Groundwater. International Journal of Science, Strategic Management and Technology, *02*(03). https://doi.org/10.55041/ijsmt.v2i3.356

Abstract

Landfills and wastewater treatment plants have become major sources of new types of pollution, particularly microplastics and chemicals called PFAS. These PFAS chemicals are especially worrying because they're found everywhere in our environment, they don't break down naturally, and they can harm living things. That's why health experts are really concerned about them. Both landfills and wastewater facilities are releasing lots of these new pollutants, including tiny plastic particles and PFAS chemicals, into the environment. What makes this worse is that these substances are completely artificial - they don't exist in nature at all.

PFAS is actually a huge group of different chemicals that manufacturers use to make things like non-stick pans, waterproof clothing, paper products, and firefighting foam. Companies love using these chemicals because they're great at repelling water and oil, they can handle extreme temperatures, and they reduce friction. After decades of making and using these chemicals, they've ended up scattered throughout our environment. When PFAS gets into groundwater, it behaves pretty much like other dissolved pollutants - it moves around through processes like water flow, spreading out, sticking to soil particles, and getting diluted.

Researchers are only beginning to grasp just how harmful PFAS can be to both people and nature. Certain kinds, especially PFOA and PFOS, are really troublesome since they move through water systems so easily, stick around forever without breaking down on their own, and keep accumulating in plants and animals as time goes on.

KEYWORDS: groundwater pollutant., Microplastics, per- and polyfluoroalkyl substances, Diffusion, zwitterion.

1. Introduction

Underground water serves as a crucial freshwater supply for no less than two billion people around the globe, and people rely on it for everything from drinking water to farming, household needs, and manufacturing activities. (Re, 2019). Sadly, underground water sources can get polluted by all sorts of new contaminants that come from human activities. (Lap worth et al., 2012), such as per– and poly-fluoroalkyl substances (PFASs) and pharmaceuticals (Sui et al., 2015). This could stop people from being able to use groundwater in helpful ways and might mean expensive and difficult cleanup work has to be done. (Siegel, 2014). Groundwater gets polluted mainly from farm chemicals washing away (Abdalla and Khalil, 2018), recycled sewage and organic waste (Lapworth et al., 2012), and factories dumping stuff (Xu et al., 2021). Since plastic production really started booming back in the 1950s, we've become absolutely obsessed with these materials at a crazy pace. We went from making just 1.5 million tons of plastic each year back in the 1950s to churning out more than 370 million tons by 2019 (Kumar, Verma et al. 2021). What's really troubling is that about 79% of all the plastic we make ends up in landfills or scattered throughout our environment, while only a tiny 9% actually gets recycled properly (Geyer et al. 2017). Because of this, plastic waste and tiny plastic particles are now everywhere you look in nature. These microscopic plastic bits keep showing up in rivers, lakes, and streams, and researchers have even found them contaminating our underground water supplies (Hoellein et al., 2017; Lenaker et al., 2019; Panno et al., 2019; Xu, Ou et al., 2022). PFAS make up a big group of chemicals that companies use to make things like non-stick pans, fabrics, paper goods, certain firefighting foams, and tons of other stuff we use every day. What makes these chemicals so popular is that they're really good at keeping oil and water away, they can handle really hot or cold temperatures without breaking down, and they help reduce friction between surfaces. The thing about PFAS is that they come in all different sizes and shapes at the molecular level, with various structures and parts that do different jobs. Because we've been making and using these chemicals for so many years, they've ended up spreading throughout our environment.

2. Physical Properties

The way PFAS behaves in the environment - like what form it takes and how it spreads around - depends on both the physical and chemical makeup of these compounds and the specific conditions of the environment they're in.

2.1 Physical State/Appearance

At room temperature, you'll typically find most PFAS as solid materials that look crystalline or powdery. But here's the thing - the ones with shorter chains, like the acid versions of PFCA and PFSA, along with FTS and FTOH that have 4 to 6 carbon atoms in their tail, actually stay liquid when they're at room temperature.

2.2 Density

When liquid PFAS compounds are denser than water, they can sink down through groundwater or surface water as what scientists call DNAPL - basically a heavy liquid that doesn't mix with water. Take 4:2 FTOH as an example - it's liquid at room temperature, weighs about 1.59 grams per cubic centimeter, and dissolves in water at around 974 milligrams per liter. If you spilled pure 4:2 FTOH into the environment, it would act a lot like carbon tetrachloride, which has similar density and dissolving properties. But here's the thing - when 4:2 FTOH actually dissolves in water, the mixture doesn't get heavy enough to form its own separate layer. Interestingly, researchers have noticed that when PFOA and PFOS are mixed with water at really high concentrations, you can actually see separate liquid layers floating on top.

2.3 Melting/Boiling Points

These characteristics decide if a particular pure PFAS chemical will be liquid, solid, or gas at normal environmental temperatures. You'll find that this information can differ between different sources. While we have predicted melting and boiling points for most PFAS chemicals, we don't have actual measured values for many of them. The predicted numbers are helpful for getting a sense of what physical state these PFAS chemicals might be in, but we really don't know how accurate these predictions are - that's something that needs more research. What we do know from the available information is that PFAS chemicals tend to have higher melting and boiling points when their fluorinated chains get longer. Take PFBA for instance - it melts at -17.5°C, but perfluorotetradecanoic acid (PFTeDA) melts somewhere between 130 and 135°C.

2.4 Solubility

Solubility is determined by looking at how much of a substance can dissolve in a liquid under specific temperature and pressure conditions. It's usually expressed in units like milligrams per liter or moles per liter. Right now, we have actual lab measurements for the water solubility of many well-known PFAS chemicals, but we're still missing data for the ones that haven't been studied as much. It's important to keep this data gap in mind when you're working with PFAS solubility information. Most of the numbers you'll see in research are actually predictions or computer models rather than real experimental results, and even the numbers used to create those models might be educated guesses.

2.5 Partitioning to Fluid-Fluid Interfaces

PFAAs have a dual nature - they're made up of water-loving heads attached to water-repelling tails, which makes them act just like regular surfactants. What this means is that they naturally gather at the boundaries between different fluids, like where air meets water or where oil meets water. When they line up along these boundaries, they position themselves so their water- repelling tails stick out into the air while their water-loving heads stay in the water.

3 Chemical Properties

3.1 Carbon-Fluorine (C-F) Bond Properties

The characteristics of PFAS mainly come from what makes the carbon-fluorine bond so special.

Table 1 Fluorine characteristics

Fluorine Characteristic	**Description**	**Effect**	**Resulting Property of PFAS**
High electronegativity	Tendency to attract shared electrons in a bond	Strong C-F bond	Thermal stability
		Polar bond with partial negative charge toward F	Chemical stability (low reactivity)
			Strong acidity (low pKa)1
Low polarizability	Electron cloud density not easily impacted by the elec	Weak intermolecular interactions	Hydrophobic and lipophobic surfactant properties2
		Low surface energy	
Small size3	Atomic radius of covalently bonded fluorine is 0.72 Å	Shields carbon	Chemical stability (low reactivity)

1. When paired with an acid functional group such as a carboxylic or sulfonic acid.
2. When paired with a functional group that is hydrophilic (for example, a carboxylate)
3. Smallest of the halogen atoms. Å = angstrom

Fluorine has some pretty special characteristics - it's highly electronegative and really small, which creates an incredibly strong bond with carbon that's actually the toughest covalent bond you'll find in organic chemistry. Since fluorine doesn't get polarized easily, it doesn't interact much with other molecules through things like Van der Waals forces or hydrogen bonds. These distinctive traits are what make many PFAS compounds so good at repelling both water and oil, which is why they work great for stain-resistant products and as surfactants. They're also incredibly stable when exposed to heat or chemicals. That said, not every PFAS compound has all of these features - surface activity, for instance, isn't something you'll see across the board.

3.2 Functional Group Properties

PFAS contain various functional groups like carboxylates, sulfonates, sulfates, phosphates, amines, and several others. Whether these groups are dissociated or not plays a big role in how PFAS behave and move around in the environment. Basically, whether a compound has an electrical charge affects its physical and chemical characteristics, which then determines what happens to it in nature.

Because PFAAs have really low acid dissociation constants, you'll almost always find them as negatively charged anions in the environment - it's extremely rare to see them any other way. Some PFAS compounds can actually break apart into either positive or negative ions when they're in water, depending on the pH levels. Take PFOA, for example - when it dissolves in water across most pH ranges, it splits into a perfluorooctanoate anion and a hydrogen ion. The part of ionic PFAS that contains the fluoroalkyl chain can end up being negatively charged, positively charged, or even both at the same time as a zwitterion.

PFAS can be grouped into four different categories depending on their functional groups.

- Anionic means it has acidic parts like carboxylic acids, sulfonic acids, sulfates, and phosphates that can give off a hydrogen ion, which creates a negatively charged particle called an anion.
- Cationic substances have basic functional groups like amines that can pick up a hydrogen ion to become positively charged, or they might already have a permanent positive charge like you'd see with quaternary ammonium groups.
- A zwitterionic compound has two or more functional groups where at least one can become negatively charged and another can become positively charged.
- Nonionic substances don't break apart into charged particles - alcohols are a good example of this type of

compound. Given how other positively and negatively charged surfactants behave, we'd expect positively charged PFAS to move through the environment differently than negatively charged PFAS.

3.3 Thermal Stability

The ability of a chemical to stay stable when heated up, known as thermal stability, helps us figure out how long that chemical might stick around in our environment. Chemicals like PFOA and PFOS, which belong to a group called PFAAs, are incredibly tough - they don't break down easily when exposed to heat or other chemical processes, and they resist both degradation and oxidation. What makes these PFAAs so remarkably stable when heated comes mainly from the super-strong bonds between carbon and fluorine atoms in their fluoroalkyl chains, though how stable they are also depends on what specific functional group is attached to that fluoroalkyl part. Among fluorinated surfactants, PFCAs and PFSAs are the champions when it comes to handling heat. Different studies give varying numbers for the temperatures needed to break down PFAS, but it looks like you might need temperatures over 1,000 degrees Celsius to destroy PFAS that's in soil. Earlier research showed that even at 700 degrees Celsius, only a limited amount of PFOS, PFOA, and PFHxA actually got completely broken down - we're talking about 72% or less.

3.4 Chemical Stability

Just like how we look at thermal stability, understanding how chemically stable a molecule is helps us figure out how long it will stick around in the environment. PFCAs and PFSAs have proven to be really persistent out there in nature. PFCAs can resist breaking down from oxidation in normal environmental conditions, though scientists have managed to transform them when using strong oxidizing agents under really intense pressure. In the fluorinated chain part of these alkyl acids, you've got the strong bond between carbon and fluorine, plus the way fluorine atoms shield the carbon, and the pull effects from fluorine being so electronegative - all of this adds up to make these molecules really chemically stable. Think about it this way: there are these electron-rich molecules called nucleophiles that would normally be drawn to the slightly positive carbon atoms. But if these nucleophiles could actually get close enough to bond with the carbon, the reaction that follows might kick out a fluorine atom and replace it with the nucleophile, which could then make the whole molecule more susceptible to breaking down.

4 Fate and Transport Processes

4.1 Partitioning

PFAS move around, get transported, and change form across different types of environments. The PFAS chemicals we usually find in nature have a special structure - they've got a carbon-fluorine "tail" part and a regular "head" part that contains a polar group. You can see this setup in Figure 2 with PFOS and PFOA as examples. The tail part repels both water and fats, while the head part attracts water.

Several key things control how PFAS behave in the environment, including their tendency to avoid water and fats, electrical attractions between charged particles, and how they act at boundaries between different materials. That water-and-fat-avoiding behavior makes them stick to organic matter in soil. The electrical interactions depend on whether the polar head group is charged or not.

Generally speaking, PFAS with longer tails stick around more and move more slowly through soil compared to those with shorter tails. This means short-chain versions of PFSAs and PFCAs don't get held back as much as the long-chain ones. Also, PFSAs typically stick to surfaces better than PFCAs when they have the same tail length, and the branched versions don't stick as well as the straight-chain ones.

4.2 Transport

Since most PFAS chemicals don't break down naturally through biological processes or environmental conditions - aside from when precursor compounds transform - the way they physically move through the environment becomes really important for understanding how they spread and where people might come into contact with them.

4.2.1 Advection, Dispersion, Diffusion

The movement of PFAS chemicals through different environments gets heavily affected by things like advection, dispersion, and diffusion. When we talk about advection, we're basically talking about how these compounds get carried along by flowing fluids like water or air - and this is actually what drives PFAS movement in most situations. The thing is, advection just moves the chemicals around without actually making them less concentrated as they travel.

When air currents and water flow speeds change on a smaller scale, they end up scattering contaminants in various directions. This leads to quick vertical mixing of PFAS and helps them jump between different environments - like going from surface water into sediment or falling from the air onto soil.

Diffusion is what happens when molecules move around because of concentration differences - you see this in both air and water. When water or air gets turbulent and creates mixing, scientists also call this eddy diffusion. In groundwater, people usually don't worry much about diffusion because it happens really slowly compared to advection. But here's the catch - when contaminants spread through diffusion into materials that don't let much through, like clay, bedrock, or concrete, it can actually make PFAS stick around in groundwater for much longer periods.

4.2.2 Deposition

Although most PFAS don't easily turn into gas, some of them can still travel through the air when they're released from factories and industrial plants, like through smokestacks. When these chemicals get into the air, sunlight can break them down and they can be carried by wind currents. Eventually, they settle back down to earth and build up in dirt and water sources where we can actually measure them. This settling process happens in two main ways - either the chemicals just fall out of the air on their own along with tiny particles, or they get washed out when it rains or snows. The dry way happens when PFAS stick to small droplets or particles floating in the air and naturally fall down through gravity or other natural processes. The wet way is basically when rain or snow picks up these contaminated particles as it falls and brings them down to the ground.

4.2.3 Leaching

When it rains or when areas get irrigated, PFAS chemicals found in soil that isn't completely saturated tend to get washed downward as water dissolves these pollutants that are stuck to soil particles. This washing process can move PFAS from the surface soil down into groundwater and nearby water sources, which is especially concerning since these chemicals often end up on the surface through spills or falling from the atmosphere in the first place. This same leaching process can also affect how plants take up PFAS, particularly around landfills that don't have proper systems to control contaminated water runoff. How much leaching actually happens depends on things like the soil's pH level and how much organic matter it contains, as well as the specific characteristics of the PFAS chemicals themselves, such as their electrical charge and the length of their molecular chains.

4.2.4 Surfactant Properties and Micelle Formation

PFAS act like surfactants because they have parts that repel water and parts that attract it, making their movement through the environment complicated and poorly understood. Many PFAS are designed to naturally gather at the boundary between air and water, with their fluorine-containing tails pointing toward the air and their water-loving heads staying in the water. This tendency affects how they travel through the air in tiny droplets and settle out, and it means PFAS tends to build up at water surfaces. This same preference for air-water boundaries probably matters when PFAS moves through soil above the water table, since there's lots of contact between air and water in those spaces. When PFOS and PFOA stick to these air-water boundaries, it can slow down how fast they move through water.

5. PFAS Transformation

PFAA formation can happen when certain polyfluorinated substances called precursors go through natural biological processes or chemical changes in the environment. But once PFAAs are formed, they're pretty much stuck that way - they don't break down under normal environmental conditions. The precursor PFAS are different from the fully fluorinated PFAAs because they have carbon-hydrogen and carbon-oxygen bonds that can actually react and change through various biological and chemical processes, eventually turning into final products. Most of what we know about how these precursor PFAS transform comes from laboratory studies done under controlled conditions, but several real-world field studies have shown that these precursors play a significant role at different locations, like the research done by Weber and colleagues in 2017 and Dassuncao and team that same year.

5.1 Abiotic Transformation

they can create Non-living processes can change chemical precursors into other compounds when they're exposed to normal environmental conditions. These processes include breaking down with water, breaking down with light, and reacting with oxygen. When some precursors break down with water and then get processed by living organisms, PFCAs and PFSAs. Take PFOS, for instance - it comes from a compound called perfluorooctane sulfonyl fluoride when it breaks down this way, as Martin and his team found in 2010. Similarly, PFOA and other PFCAs form when fluorotelomer-based precursors break down with water, according to Washington and Jenkins' 2015 research.

While scientists haven't seen PFAS compounds break down directly from sunlight, some precursors do break down indirectly from light exposure. This happens especially with fluorotelomer alcohols in the atmosphere, which can lead to PFCA deposits falling back to earth. Armitage, MacLeod, and Cousins documented this in 2009, and Yarwood's team did similar work in 2007. There's another group of compounds called perfluoroalkane sulfonamides that can break down in the atmosphere through oxidation reactions. When this happens, they produce PFCAs at rates that might be ten times higher than what you get from fluorotelomer alcohols, based on Martin's 2006 research. Even shorter-chain compounds like PFBS can form when hydroxyl radicals react with sulfonamido derivatives through oxidation, as D'Eon and colleagues showed in 2006.

Sometimes these non-living transformation processes don't immediately create any PFAA compounds, but they might still lead to PFAA formation eventually, according to Martin's 2010 findings.

5.2 Biotic Transformation

Although PFOA, PFOS, and similar PFAAs can't be broken down by microbes, researchers have found that many of their precursor compounds can be transformed by biological processes, much like the non-biological changes we've talked about. What we know from research shows there are many ways these transformations happen when oxygen is present, they occur pretty quickly, and it looks like all these polyfluorinated precursors could potentially be converted into PFAAs through these oxygen- dependent biological processes. There hasn't been as much research done on what happens when oxygen isn't available. Scientists have seen that FTOHs can change without oxygen, but they seem to turn into stable polyfluorinated acids instead of becoming PFCAs or PFSAs. Researchers also noticed that PFOA and PFOS lost some of their fluorine atoms when ammonium was processed without oxygen in conditions where iron was being reduced.

6. CONCLUSION:

The movement of PFAS chemicals through groundwater happens through the same basic processes we see with other contaminants - they get carried along by water flow, spread out through dispersion, stick to soil particles, and get diluted. How fast these chemicals move depends mainly on how quickly the groundwater flows and how much they stick to the surrounding soil and rock materials.

The chemistry of the groundwater itself and what the underground materials are made of play a big role in determining where PFAS ends up and how it moves around. When groundwater is more acidic, has more calcium, or contains more dissolved minerals, PFAS tends to stick better to the soil, which can slow down how fast it spreads and reduce how much stays dissolved in the water. Having more organic matter in the soil or mineral surfaces with positive charges also helps trap many types of PFAS, making them move more slowly through the ground.

The specific makeup of different PFAS chemicals affects how they behave underground too. These chemicals can have carbon chains that are short or long, straight or branched, or even formed in rings, and all of this influences how well they stick to soil. Generally, PFAS with longer, straight chains are more likely to get held up by sticking to soil particles. These chemicals also have a polar "head" that makes them interact with electrical charges.

What makes PFAS different from many other groundwater pollutants is that most of them don't break down naturally - they just stick around indefinitely. Some related chemicals called precursors might partially break down, but they just turn into the more stable PFAS that then persist without changing further. This means these chemicals can actually build up over time instead of disappearing like other contaminants might.

Even though most PFAS act like soaps or detergents, the amounts we typically find in the environment are usually too low for them to form the structures that soaps make when concentrated. Still, even at these lower levels, PFAS molecules might clump together in small groups.

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