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Perspective

Outside the Safe Operating Space of a New Planetary Boundary for Per- and Polyfluoroalkyl Substances (PFAS)

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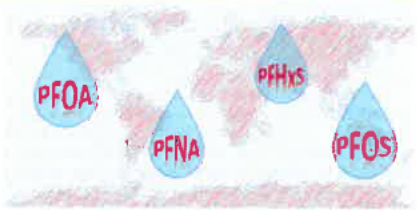
ABSTRACT: It is hypothesized that environmental contamination by per- and polyfluoroalkyl substances (PFAS) defines a separate planetary boundary and that this boundary has been exceeded. This hypothesis is tested by comparing the levels of four selected perfluoroalkyl acids (PFAAs) (i.e., perfluorooctanesulfonic acid (PFOS), perfluorooctanoic acid (PFOA), perfluorohexanesulfonic acid (PFHxS), and perfluorononanoic acid (PFNA)) in various global environmental media (i.e., rainwater, soils, and surface waters) with recently proposed guideline levels. On the basis of the four PFAAs considered, it is concluded that (1) levels of PFOA and PFOS in rainwater often greatly exceed US Environmental Protection Agency (EPA) Lifetime Drinking Water Health Advisory levels and the sum of the aforementioned four PFAAs (Σ4 PFAS) in rainwater is often above Danish drinking water limit values also based on Σ4 PFAS; (2) levels of PFOS in rainwater are often above Environmental Quality Standard for Inland European Union Surface Water; and (3) atmospheric deposition also leads to global soils being ubiquitously contaminated and to be often above proposed Dutch guideline values. It is, therefore, concluded that the global spread of these four PFAAs in the atmosphere has led to the planetary boundary for chemical pollution being exceeded. Levels of PFAAs in atmospheric deposition are especially poorly reversible because of the high persistence of PFAAs and their ability to continuously cycle in the hydrosphere, including on sea spray aerosols emitted from the oceans. Because of the poor reversibility of environmental exposure to PFAS and their associated effects, it is vitally important that PFAS uses and emissions are rapidly restricted.



KEYWORDS: *PFAS, planetary boundary, chemical pollution, environmental exposure*

INTRODUCTION

A recent review article in *Science*[1] highlighted the global threat posed by plastic pollution. These concerns were based on the high environmental persistence of plastics, the related "poor reversibility" and a range of potential effects. Other researchers, including ourselves,[2,3] have pointed out similar concerns related to highly persistent nonpolymeric substances, but these concerns are not equally obvious to the public compared to the concerns with plastics. The relatively high public concern regarding plastics is possibly driven by the visibility of plastic waste compared to nonpolymeric substances.[4] Clearly, both plastic pollution and pollution by highly persistent nonpolymeric substances lead to similar global problems. Persistence is generally seen as a less immediate hazardous property than toxicity, but it actually is the key factor that lets pollution problems spiral out of control.[2] This is because persistence enables chemicals to spread out over large distances, causes long-term, even life-long exposure, and leads to higher and higher levels in the environment as long as emissions continue. These increasing levels will with high probability sooner or later lead to adverse effects. Importantly, microplastic is under consideration for restriction in the EU because of the extreme persistence of plastics and the irreversibility of the exposure caused by plastic particles in the environment.[5]

Recently a group of scientists flagged the concerns regarding the inability of scientific analyses to keep pace with the amount of chemicals produced and released into the environment,[6] which limits the ability to discover new environmental threats in time. Others have similarly pointed out the need for precautionary chemicals managements; a notable example is the report, "Late Lessons from Early Warnings",[7] where many historical examples of global contamination problems are provided, often associated with persistent chemicals.

A well-known class of pollutants, the per- and polyfluoroalkyl substances (PFAS), have also recently featured in a review in *Science*.[8] The vast majority of PFAS are highly persistent (based on the EU REACH definition whereby a substance is persistent if it is persistent itself or has persistent degradation products[9]), and this has been seen as basis for managing them as a chemical class.[3] While the review article in *Science* pointed out the ubiquity and high persistence of PFAS, it did not point out the current widespread and poorly reversible risks associated even with low-level PFAS exposures. It is hypothesized here that due to the global spread of PFAS, the irreversibility of exposure to PFAS, and the associated biological effects, a new planetary boundary for PFAS has been exceeded.

Unfortunately, although there are many thousands of substances defined as PFAS in use (PFAS include any substance with at least one $-CF_2-$ or $-CF_3$ moiety in its structure[10]), the current understanding of biological impacts is based primarily on studies of four perfluoroalkyl acids (PFAAs), namely, perfluorooctanesulfonic acid (PFOS), perfluorooctanoic acid (PFOA), perfluorohexanesulfonic acid (PFHxS), and perfluorononanoic acid (PFNA). Whereas all PFAS can be grouped into a class on the basis of their high persistence,[3] it is not possible to group many of them according to biological risk because of a paucity of data on exposure and effects for most PFAS.[11] Therefore, because of data gaps, the analysis presented here is based only on the four PFAAs mentioned above. In the following, we provide four pieces of evidence to support the claim that, even considering only these four PFAAs, the new planetary boundary for PFAS has been exceeded.

In the planetary boundary concept, an attempt is made to estimate the boundaries for "a safe operating space for humanity with respect to the functioning of the Earth System".[12,13] Chemical pollution was one of the original nine anthropogenic impacts for which planetary boundaries were postulated because it can influence Earth System functioning: "(i) through a global, ubiquitous impact on the physiological development and demography of humans and other organisms with ultimate impacts on ecosystem functioning and structure and (ii) by acting as a slow variable that affects other planetary boundaries."[12,13] The "chemical pollution" boundary was renamed as the "novel entities" (NEs) boundary by Steffen et al.,[14] where NEs are defined as "new substances, new forms of existing substances and modified life forms", including "chemicals and other new types of engineered materials or organisms not previously known to the Earth system as well as naturally occurring elements (for example, heavy metals) mobilized by anthropogenic activities". Several groups of scientists[6,15,16] have pointed out the challenges in quantifying the planetary boundary for NEs, and recently it was proposed to instead use various control variables to determine if the boundary is exceeded.[6] It is, in our opinion, an insurmountable task to quantify the boundary for all NEs because (1) there are critical data gaps for a large proportion of existing NEs, (2) NEs of various types and mixtures of NEs are continuously being generated and released to the environment, and (3) there are multiple possible effects (not only toxic effects) that individual NEs or groups/mixtures of NEs can cause. Several of the existing planetary boundaries are related to the release of NEs. For example, the boundaries for "stratospheric ozone depletion" and "climate change" address the release of ozone depleting substances and gases with global warming potential, respectively. Therefore, rather than being a single planetary boundary, the boundary for NEs can be thought of as a placeholder for multiple planetary boundaries for NEs that may emerge. It is argued here that PFAS define a new planetary boundary for NEs.

We argue that if drinking water health advisories and other guidelines designed to protect human health are exceeded due to the global environmental spread of PFAS, then there is a real danger of global health effects (e.g., affecting human physiology) occurring and that it can be argued that the planetary boundary for PFAS is exceeded. We do not deem it necessary to demonstrate the prevalence of global human health effects due to PFAS exposure to prove our hypothesis, and we hope that such widespread effects in the human population are never observed.

THE US EPA LIFETIME DRINKING WATER HEALTH ADVISORIES FOR PFOS AND PFOA ARE OFTEN LOWER THAN THEIR RESPECTIVE LEVELS IN RAINWATER AND THE DANISH DRINKING WATER LIMIT VALUE FOR Σ4 PFAS IS ALSO OFTEN LOWER THAN THE LEVEL OF Σ4 PFAS IN RAINWATER

In June 2022, the US Environmental Protection agency (EPA) announced the release of health advisories for four PFAS, including interim updated nonregulatory lifetime drinking water health advisories for PFOA and PFOS of 4 pg/L and 20 pg/L, respectively.[17] The US EPA health advisories identify the concentration of chemicals in drinking water at or below which adverse health effects are not anticipated to occur and, in divergence with previous advisories, are based on human epidemiology studies in populations exposed to these chemicals. The most sensitive noncancer effect and the basis for the risk assessment behind the interim updated health advisories for PFOA and PFOS is suppression of vaccine response (decreased serum antibody concentrations) in children. The US EPA's previous nonregulatory lifetime drinking water health advisories were 70 ng/L for the sum of concentrations of PFOS and PFOA. In 2020, the European Food Safety Authority (EFSA) published their Opinion on the risks to human health arising from the presence of PFAS in food[18] and proposed a group tolerable weekly intake (TWI) of 4.4 ng/kg body weight for the sum of PFOA, PFNA, PFHxS, and PFOS. On the basis of the available studies in animals and humans, effects on the immune system were considered the most critical for the basis of the risk assessment.[18] In June 2021, on the basis of the TWI in the EFSA Opinion, the Danish Environmental Protection Agency tightened their drinking water limit values and announced that drinking water must not contain more than 2 ng/L of Σ4 PFAAs.[19]

PFAS drinking water guidelines have progressively decreased over the last 22 years.[20] For example, in the US the PFOA drinking water guideline for West Virginia was 150 000 ng/L,[20] which is higher by a factor of 37.5 million than the recently announced US EPA drinking water lifetime advisory for PFOA of 4 pg/L. As a result of this decrease, international drinking water guidelines for PFAS are now close to, or even lower than, levels in precipitation. Humans residing in industrialized areas of the world do not often drink rainwater in modern life, but it should nevertheless be a reasonable expectation that the environment is clean enough that rainwater and mountain stream water fed by precipitation is safe to drink. Furthermore, in some parts of the world, notably in some arid and tropical regions, rainwater remains an important source of drinking water.[21]

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continue to remain in the environment due to their high persistence and will continually cycle in the hydrosphere.

The analysis presented here has purposefully referred to the most stringent PFAS guideline values on an international basis, which are not representative of international guideline values for PFAS. There is, for example, a large disagreement internationally, and even between individual states in the US,[20] regarding drinking water guidelines for PFAS. The various guidelines were developed by different scientists at different time points and the risk assessments are often based on varying end points. A clear and disturbing temporal trend emerges, however, with more recent guidelines being several orders of magnitude lower than older guidelines.[20] Guidelines in the US and Europe have been driven downward recently as a result of emerging evidence for the suppression of vaccine response in children.[57] We make no attempt to determine which of the many guidelines (see compilation[58]) is based on the strongest empirical evidence on effects because such a judgment is outside of our expertise. The point that we want to make is that the most stringent risk-based health advisories are often well below environmental levels, and this should be of concern and a reason for taking stringent measures.

Although PFAS are globally present in all environmental media and locations, there are still some few areas of the planet where the environmental levels of PFAS remain relatively low. However, even in these remote and sparsely populated regions, such as Antarctica and the Tibetan plateau, the most stringent PFAS guidelines are exceeded (Figure 1). These areas cannot support large populations and are not available for settlements where major parts of the population could move. In most other areas, PFAS guideline values are exceeded and this implies potential public health impacts: higher incidences (notably in large populations, i.e., many cases) of PFAS-related effects, such as reduced immune response, but also high additional costs for healthcare and, where possible, remediation.[59] Moreover, in many cases, PFAS-related impacts occur in combination with other environmental issues, such as water scarcity or pollution by other contaminants.

Finally, we conclude that PFAS define a new planetary boundary that has been exceeded, based on PFAS levels in environmental media being ubiquitously above guideline levels. Irrespective of whether or not one agrees with our conclusion that the planetary boundary for PFAS is exceeded, it is nevertheless highly problematic that everywhere on Earth where humans reside recently proposed health advisories cannot be achieved without large investment in advanced cleanup technology. Indeed, although PFOS and PFOA were phased out by one of the major manufacturers (3M) 20 years ago, it will take decades before levels in land-based water and precipitation approach low picogram per liter levels. Moreover, the problems associated with PFOS, PFOA, or Σ4 PFAAs are likely to be only the tip of the iceberg given that there are many thousands of PFAS in the class and the risks associated with most of them are unknown.[60] In view of the impacts of humanity's chemical footprint on planetary health, it is of great importance to avoid further escalation of the problem of large-scale and long-term environmental and human exposure to PFAS by rapidly restricting uses of PFAS wherever possible.[61] Furthermore, as has been stated by ourselves[3] and others[7] before, society should not continually repeat the same mistakes with other persistent chemicals.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.2c02765.

Raw data and references used to create Figure 1 (XLSX)

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Notes

The authors declare no competing financial interest.

Biography

Ian Cousins is Professor in Environmental Organic Chemistry at the Department of Environmental Science at Stockholm University in Sweden. He is originally from the UK but has been living in Sweden and working at Stockholm University for more than 20 years. Prof. Cousins has a B.Sc. in Chemistry from the University of York and a Ph.D. in Environmental Science from Lancaster University. He is well-known for his research on the sources, transport and fate, and exposure pathways of PFAS. In recent years, Prof. Cousins has written a series of policy-related articles driven by his concern about the continued use of PFAS. This Perspective is his latest contribution. Prof. Cousins coordinates the PERFORCE3 project, which is a Europe-wide multipartner doctoral research training programme in the field of PFAS and funded by the European Union's Horizon 2020 research and innovation programme under its Marie Sklodowska-Curie Actions (Grant Agreement No. 860665). He is also Associate Editor of the American Chemistry Society journals Environmental Science and Technology and Environmental Au.

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