Groundwater phosphorus concentrations: global trends and links with agricultural and oil and gas activities

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Abstract

Although observations show that anthropogenic phosphorus (P) can reach groundwater supplies, there has been no comprehensive evaluation of P in groundwater at the global scale and P contamination from sources such as agriculture and oil and gas activities are poorly understood. We compile and analyze 161,321 groundwater P measurements in 12 different countries to determine the extent of P contamination at the global scale. We find that all 12 countries report groundwater samples with concentrations >0.1 mg P l\textsuperscript{-1}, a concentration at which the risk for eutrophication of surface waters is high. In Canada and the United States (US), we perform an analysis of 1529 dissolved oxygen (DO) concentrations to determine the degree of association between DO and groundwater P measurements. For P concentrations <0.1 mg P l\textsuperscript{-1}, we find a strong inverse relationship between DO and P. However, for P concentrations >0.1 mg P l\textsuperscript{-1}, we find a weak inverse correlation, which suggests anthropogenic sources are responsible for elevated P concentrations in groundwater. To identify anthropogenic sources of P, we conduct an analysis on land use data and the 24,146 P concentrations in Canada and the US. Although we find that 12% (2899) of all P concentrations are >0.1 mg P l\textsuperscript{-1}, 33% of P concentrations from P monitoring sites located on pastureland (managed grassland) report concentrations >0.1 mg P l\textsuperscript{-1}. In Alberta and Ontario, we analyze P measurements with respect to their proximity to oil and gas wells and find the relationship to be inconclusive. Overall, we find a positive correlation with agriculture and elevated groundwater P concentrations, but additional data are needed to identify a relationship between oil and gas wells and elevated groundwater P concentrations. Characterizing the role of agriculture and oil and gas wells on groundwater P contamination can help regulators develop effective strategies to protect water quality and ecosystem health.

1. Introduction

Excess phosphorus (P) in surface water systems can lead to ecological effects such as eutrophication, which is one of the most prevalent causes of water quality impairment, and eventual death of aquatic ecosystems [1–3]. To sustain health of lakes, rivers, estuaries, and other surface water systems, government agencies are actively working to monitor and control anthropogenic sources of P. The current extent of P contamination in groundwater worldwide and the full range of pathways through which this contamination is occurring are unknown. Historically, it has been assumed that groundwater concentrations of P are negligible due to high rates of adsorption of mobile P, typically orthophosphate, to the soil and sediment matrix [4]. However, recent studies indicate that P concentrations in groundwater may not be negligible and that characterizing the extent of P contamination in groundwater is important due to interactions between groundwater and surface water [3–15].

Although there have been site-specific studies on local or ‘point’ sources of P in groundwater, such as wastewater releases and residential underground septic tank systems [5, 12–14, 16–19], there have
been limited studies conducted on nonpoint sources, such as agriculture. However, agricultural activities account for more than 60% of anthropogenic P additions to the environment through the use of commercial fertilizers as well as manure from livestock [20]. Globally, 38% of anthropogenic P loads to freshwater ecosystems are contributed by agriculture [21]. Therefore, we evaluate P concentrations with respect to land cover/use types, encompassing all P sources from wastewater to agriculture to evaluate their relative effects.

A potential pathway through which anthropogenic P, including those from fertilizer and fluids injected and produced from oil and gas production, may enter groundwater is oil and gas wells, particularly those that are unplugged and leaky (SM: Oil and gas: contamination sources and leakage pathways). Numerous studies have studied researched leakage of hydrocarbons and water through oil and gas wells from deep formations to shallow groundwater aquifers and the atmosphere [22, 23]. Studies so far have found that the risk of upward migration of brines and produced water from oil and gas production is relatively small, especially compared to surface spills [24–28]. Moreover, groundwater samples are typically collected within 200 m of the surface [29], which is shallow relative to depths of most oil and gas activities, which generally occur at depths greater than 1000 m [30] (figure S1 available online at stacks.iop.org/ERL/17/014014/mmmedia). Based on available studies detailing oil and gas well leakage and the abundance of oil and gas wells in Canada and the United States (US) [31], oil and gas wells may potentially act as a pathway through which surface P enters the subsurface. To obtain indications of the broad potential effects of oil and gas wells on P concentrations in groundwater as a source and/or leakage pathway (SM: Oil and gas: contamination sources and leakage pathways), we analyze oil and gas well distributions and available concentrations of P in groundwater.

In this paper, we: (a) compile available global groundwater P data, (b) analyze the extent of elevated P concentrations in groundwater and the spatial distribution of available data globally, (c) examine the relationship between dissolved oxygen (DO) and P concentrations in groundwater, (d) identify spatial relationships between land use/land cover types and elevated total P concentrations in groundwater in Canada and the US, and (e) compare oil and gas well locations to elevated groundwater P concentrations in regions with high densities of oil and gas wells and P data (British Columbia, Ontario and Alberta, Canada). These results can be used to guide agricultural and energy policy development and inform plans for monitoring of P in groundwater.

2. Materials and methods

2.1. Global data

We compile 161 321 P measurements using data from 15 government agencies and eight peer-reviewed studies conducted in 12 different countries (table S1). Data is categorized by country/region and P concentration type. Where available, we provide the groundwater depth ranges at which samples have been collected, the range of sampling dates, and the phosphorus concentration detection limits (table S2). Samples that report concentrations less than the detection limit are assumed to have a concentration of 0 mg l<sup>−1</sup> (table S3, SM: Treatment of phosphorus data). We categorize the measurements based on the analysis methods: (a) total phosphorus (TP) after acid digestion followed by molybdate blue colorimetry or by inductively coupled plasma optical emission spectroscopy, (b) TP as determined by inductively coupled plasma mass spectrometry, (c) dissolved orthophosphate as P, (d) total dissolved P, and (e) particulate P [32] (table S4, SM: Treatment of phosphorus data, SM MS Excel Spreadsheet at https://osf.io/d96ak/?view_only=9c53811eb4d746c0862ea88b1b838b074).

2.2. Geospatial analysis

We geospatially analyze land use data, TP concentrations in groundwater, and oil and gas well locations in ArcGIS to link anthropogenic activity to increased TP concentrations in groundwater. To identify relationships between anthropogenic sources and enhanced P concentrations, we use all P concentration types. We analyze land use types throughout the US and Canada to evaluate the effects of anthropogenic activities such as agriculture and oil and gas on P concentrations in groundwater (SM: Geospatial analysis). We conduct an analysis of oil and gas well proximity and P concentrations in groundwater in the Canadian provinces of British Columbia (SM: Analysis of oil and gas well proximity impact on phosphorus concentrations in British Columbia), Alberta, and Ontario.

2.3. Statistical analysis

Due to the non-normality of the P and DO measurements, we use nonparametric methods to conduct statistical analysis on the data. We use Spearman rank correlation to measure the degree of association between DO and P measurements, and we use a chi-square (Chi<sup>2</sup>) test to compare the distribution of P measurements found at distances closer to oil and gas wells to P measurements found farther away from oil and gas wells [33, 34]. For the oil and gas well analysis, we analyze the top 30th percentile of P measurements because research has shown approximately 10% of oil and gas wells have reported leakage [23] and because of data sparsity (table S7).
3. Results

3.1. Global distribution of P in groundwater data
Of the 12 countries with P in groundwater data, we find that all countries have groundwater samples with concentrations >0.1 mg P l$^{-1}$. Figure 1 represents the sampling type and location of the wells used in this paper as well as the overall distribution of all samples within each country. China and Brazil have the largest percentage of measurements with concentrations >0.1 mg P l$^{-1}$ with 78% and 66%, respectively. Although these percentages are subject to bias in the sampling design, the high percentages clearly show that groundwater P contamination is a problem in some parts of China and Brazil. Wales, Mexico and the US are also found to have more than 20% of samples with concentrations >0.1 mg P l$^{-1}$. We find that South Africa and Ireland are the only countries with less than 10% of samples reporting concentrations >0.1 mg P l$^{-1}$.

The distribution of concentration values is skewed with most countries (11 out of 12) having larger means than medians (table S4 and figure S2). The US is found to have the highest sample mean of 0.66 mg P l$^{-1}$, followed by Brazil with 0.64 mg P l$^{-1}$. Brazil is also found to have the largest sample median with a value of 0.18 mg P l$^{-1}$, followed by China with a value of 0.16 mg P l$^{-1}$. All other countries (Germany, Ireland, Mexico, New Zealand, Northern Ireland, South Africa, Sweden, the US, and Wales) and the Canadian provinces (British Columbia, Alberta, Manitoba, Ontario, and Quebec) report median values less than 0.1 mg P l$^{-1}$. Alberta, British Columbia, Manitoba, Quebec, Ireland, New Zealand, and South Africa also report mean values less than 0.1 mg P l$^{-1}$. However, maximum concentrations in all tested regions can be well over 0.1 mg P l$^{-1}$ (table S4). The maximum concentration across the dataset is for Sweden, with a dissolved P concentration of 793 mg P l$^{-1}$. In Canada, the maximum reported concentration is 250 mg P l$^{-1}$ in Ontario (land use is settlement).

3.2. Association of DO and phosphorus concentrations in groundwater
Elevated P concentrations in groundwater have been attributed to anthropogenic sources [10, 14, 16, 35, 36] and natural processes that govern the movement of P in the subsurface [37]. Several natural factors including temperature and pH can impact P concentrations in groundwater, but DO, which affects the ability of iron oxides to adsorb and retain P [8], has been identified as a primary natural control on P concentrations in water [8, 38–40]. High DO concentrations in the subsurface allow for iron oxides to remain stable, but in low oxygen environments, iron oxides can dissolve, releasing adsorbed P back into the water and increasing P concentrations [8]. An inverse correlation between DO and P concentrations would therefore indicate a natural explanation for elevated P concentrations in groundwater.

Using Spearman’s Rank Correlation to judge the degree of association between DO concentrations and P concentrations, we find that the inverse correlation between DO and P is weaker for P concentrations >0.1 mg P l$^{-1}$. For the 1529 available locations with DO and P concentration data available in Canada and the US, we find that the overall correlation between DO and P is −0.61, suggesting a strong inverse relationship as expected. When looking at sites that report P concentrations <0.1 mg P l$^{-1}$ only, we again find a strong inverse relationship between DO and P with a Spearman rank correlation equal to −0.54. However, for sites that report >0.1 mg P l$^{-1}$, the correlation between DO concentrations and P concentrations is −0.16, suggesting a very weak inverse relationship. Moreover, decreasing the distance from P monitoring sites to cropland results in an even weaker correlation between DO and P (Spearman rank correlation of −0.11 if there is cropland within 50 m of the P monitoring site), supporting the notion that enhanced P concentrations in groundwater near cropland is likely linked to agricultural activities (table S10).

3.3. Land use types linked with higher phosphorus concentrations in groundwater
There are significant spatial gaps in the current location of P monitoring sites; only 4% of land area in Canada and 0.9% of land area in the US have at least one P monitoring site within a 30 km by 30 km area. In Canada, P monitoring sites are relatively dense in British Columbia, Alberta, Manitoba, Quebec, and Ontario but spatial gaps are still present with only 5%, 14%, 11%, 6%, and 9% of the provinces respectively having at least one monitoring site within a 30 km by 30 km search area. The maximum P monitoring site density in the US is 0.5 P sites km$^{-2}$ in Idaho and the largest in Canada is 1.3 P sites km$^{-2}$ in Alberta (white circled area in figure 2).

Correlations between land use types and areas with high P concentrations in groundwater may help identify the source of P contamination. Figure 2 shows the land use map for Canada and the US and P data collected in these regions separated into the CCME TP concentration ranges. According to a point density analysis on ArcGIS using a search area of 900 km$^2$, we find a high density of elevated P concentrations in Alberta (white circle), Manitoba (red circle), the Southern Ontario/Great Lakes region and Southern Quebec (black circle) and the urban areas, Vancouver and Victoria, in British Columbia (blue circle) (figures 2 and S4). We analyze 24 146 P concentrations located in Canada and the US to determine the relationship between land use and P concentrations. We find that 12% (2899) of all types of P concentrations across
Figure 1. Global distribution of P samples by type and country. Numbers in brackets represent total number of samples collected in that country. The pie charts show the distribution of samples with concentrations greater than (red) and less than (blue) 0.1 mg P l\(^{-1}\).
Figure 2. Land use map of Canada and the United States overlaid by TP in groundwater sample locations. Circled areas represent regions with high density of TP concentrations >0.1 mg P l\(^{-1}\) in the samples collected according to a point density analysis using ArcGIS (figure S4). The pie charts represent the distribution of sample concentrations greater than (red) or less than (blue) 0.1 mg P l\(^{-1}\).
Canada and the US are >0.1 mg P l$^{-1}$. For the P samples located directly on managed grasslands, we find that 33% of samples report concentrations >0.1 mg P l$^{-1}$ and the ‘other’ category reports 21% of samples to have >0.1 mg P l$^{-1}$, which are significantly higher percentages than the percentage associated with all samples >0.1 mg P l$^{-1}$. The remaining land use categories report having between 11% and 15% of samples >0.1 mg P l$^{-1}$. (table S13, SM: Representativeness of land use surrounding P monitoring sites in Canada and the United States). In other words, P concentrations in groundwater are more elevated than expected on managed grasslands and ‘other’ (including rocks, ice, and beaches) categories.

3.4. Relationship between oil and gas wells and phosphorus concentrations in groundwater

Based on figures 2, S4 and S5, we select Alberta and Ontario as regions of focus to identify the potential effects of oil and gas wells on P concentrations in groundwater. Both Alberta and Ontario have high densities of TP monitoring sites in addition to high densities of oil and gas wells. Although regions with elevated P concentrations in British Columbia do not have oil and gas wells, we evaluate the correlations between P concentrations and proximity to oil and gas wells in British Columbia as a basis for comparison to the Ontario and Alberta correlations (SM: Analysis of oil and gas well proximity impact on phosphorus concentrations in British Columbia).

There are 609 877 oil and gas wells of all statuses from active to abandoned in the province of Alberta, with all 306 TP monitoring sites located within 10 km radius of at least one oil and gas well as shown in figure 3. The maximum distance from a TP monitoring site to an oil and gas well occurs at 8.16 km with a TP measurement below detection limit. The maximum TP measurement in Alberta is found to be 1.67 mg P l$^{-1}$, more than ten times the required TP concentration for surface water eutrophication, and this is found at a distance of 0.41 km from an oil and gas well. In Ontario, there are 26 968 oil and gas wells, the majority (98%) of which are in the southwestern region of the province (figure 3). The maximum TP measurement in Ontario is more than 600 times the CCME recommendation at 68 mg P l$^{-1}$, and the monitoring site is located 10.8 km from the nearest oil and gas well. One of the lowest P measurements (0.005 mg P l$^{-1}$) in Ontario is found at the monitoring site located 428 km, the farthest distance, from an oil and gas well. Based on proximity of high P concentrations to oil and gas wells, we conduct a Chi$^2$ analysis to determine if there are statistical differences between P concentrations located closer to oil and gas wells.

Conducting a Chi$^2$ analysis on the top 30th percentile of TP concentrations using a 90% confidence interval, $\alpha = 0.1$, we find differences in populations of TP samples collected closer to oil and gas wells in Alberta and Ontario, depending on the cutoff distances used to identify wells located close to oil and gas wells. If we use the median distance in each province to select the cutoff distances at which TP monitoring sites are considered to be located closer to oil and gas wells, the cutoff distance is 0.3 km for Alberta and 15 km for Ontario. In Alberta, 97% of TP monitoring sites within 0.3 km of an oil and gas well report TP concentrations >0.1 mg P l$^{-1}$. At distances greater than 0.3 km, 61% of TP monitoring sites report concentrations >0.1 mg P l$^{-1}$ (table S14). In Ontario, we find 50% of TP concentrations at TP monitoring sites within 15 km of an oil and gas well are >0.1 mg P l$^{-1}$. At monitoring sites located farther than 15 km from an oil and gas well, 33% of TP concentrations are >0.1 mg P l$^{-1}$ (table S15). However, we see inconsistent trends when other cutoff distances are considered because the dataset is small and is strongly biased with few monitoring sites located far from oil and gas wells (table S7). Therefore, additional sampling such that we can represent the full range of distances from oil and gas wells is needed to understand if oil and gas development, both in terms of sources and pathways, is linked to elevated P concentrations in groundwater.

TP monitoring sites located close to oil and gas wells are typically also close to crop/pastureland in both Alberta and Ontario. In Alberta, 91% (278) of all TP monitoring sites are found within 1 km of any oil and gas wells, and of these 278 TP monitoring sites, 269 (97%) are found within 1 km of crop/pastureland. In Ontario, oil and gas wells are found at greater distances from TP monitoring wells than in Alberta. Only 12% (47) of all TP monitoring sites are found within 1 km of an oil and gas well, while 49% (196) of all TP monitoring sites are found within 5 km from any oil and gas wells. Of the 47 TP monitoring sites within 1 km of an oil and gas well in Ontario, 98% (46) are located within 1 km of crop/pastureland. In both provinces, this correlation makes it challenging to differentiate P contamination arising from oil and gas and agriculture.

4. Discussion

4.1. Limitations of global samples

Even in tropical regions, such as Brazil and Mexico, where P concentrations in groundwater are expected to be quite low due to the strong adsorption of P to soil particles, we find P concentrations >0.1 mg P l$^{-1}$. Although we do not have the detection limits from the Geological Survey of Brazil, Brazil has been responsible for the largest amount of deforestation in the Amazon [41]. 66 Mha (8% of Brazil’s land area [42]) of land has been converted for agricultural use in the decades between 1985 and 2017 [43]. Currently 28% of land in Brazil is used for pasture or agricultural purposes [43]. Moreover, multiple studies in recent years have focused on the degradation of soil and
groundwater quality as a result of excess P application from fertilizers [44–46]. In Mexico, we could not obtain a government database of P concentrations in groundwater, and the P data we collected was from site-specific peer reviewed studies. These studies are primarily conducted in areas of concern from P contamination, specifically from wastewater irrigation, which is suspected to be responsible for increased concentrations of nutrients in soils [47–50]. Overall, because P contamination from anthropogenic sources can be significant, relying on our understanding of natural P controls in tropical regions may lead to overlooking important anthropogenic groundwater P contamination.

Our database of P in groundwater samples lack data in the majority of South America, Europe and Asia. These regions have significant portions of land use designated to agricultural activities, a major source of P; 38% in South America [51], 39% in Europe [52] and 69% in Asia [53], meaning

Figure 3. Well density map of all oil and gas wells in Alberta and Ontario with TP concentration values separated by CCME concentration range. The dotted red lines on the scatter plots represent the CCME ‘hyper-eutrophic’ TP concentration of 0.1 mg P L⁻¹.
P contamination of groundwater supplies could be a significant problem. Moreover, there are many regions where high oil and gas well density coincides with intense agricultural areas such as the Middle East, Europe and several South American countries including Venezuela [54, 55]. Additional data collection and sampling campaigns in these regions could identify areas with potentially high P concentrations in groundwater.

Because the processes that govern N and P movement in the environment differ considerably, it is challenging to develop a suitable correlation between N and P to infer P concentrations in groundwater. Nitrogen is soluble in groundwater meaning that its respective forms, particularly nitrate (NO\textsubscript{3}^−), are highly mobile in the vadose zone and groundwater aquifers. In contrast, because P tends to be less soluble in groundwater and due to its high adsorption affinity in soils, it is difficult for P transport to occur through leaching through soils [56]. In future studies, with the collection of additional data such as soil properties, physically-based modeling of water flow and P and N transport through soils and groundwater aquifers may be used to relate N and P.

4.2. High P concentrations in groundwater linked to agricultural areas

The reduction in the strength of the inverse Spearman correlation between DO and P at P concentrations >0.1 mg P l\textsuperscript{−1} indicates that an anthropogenic factor is likely responsible for the elevated P concentrations. Based on the reduction in the strength of the Spearman correlation between DO and P with decreased proximity to crop/pastureland and the proximity of a majority of samples with P concentrations >0.1 mg P l\textsuperscript{−1} to crop/pastureland, agricultural areas appear to be linked to elevated P concentrations in groundwater. The agricultural areas are mostly cropland with pastureland (managed grasslands) representing <1% of the land use in Ontario and 7% of the land use in Alberta. Beyond DO concentrations, several other natural factors can impact P concentrations in water. The weathering of rocks releases P into terrestrial and aquatic ecosystems. Because rocks can have P concentrations that range from 120 ppm to 3000 ppm [57], geology can significantly influence the concentration of P in water. Therefore, geological information of the aquifers from which groundwater P samples are collected is needed to further clarify the impact of anthropogenic activities on P concentrations in groundwater.

4.3. Isolating the effects of oil and gas wells on P concentrations in groundwater

Two of the regions with elevated P concentrations in groundwater are also regions with extensive oil and gas development. In Alberta and Ontario, elevated P concentrations in groundwater are likely a result of agricultural activity. The extent to which oil and gas wells may be exacerbating the problem by acting as an open pathway for P to easily enter groundwater systems is difficult to determine because most TP monitoring sites in Alberta and Ontario coincide with regions with oil and gas development (figure 3) and the large range in statuses and conditions of oil and gas wells from abandoned unplugged wells to active wells. Moreover, in regions with a high density of oil and gas wells, potential contamination pathways may not necessarily be the nearest oil and gas wells but could be any one of a set of neighboring wells, some of which may not be documented. To better understand this relationship, additional sampling is needed, especially in areas located far from oil and gas wells, including in Alberta and Ontario. Moreover, analysis conjunctively exploring the relationship between other geochemical parameters such as total dissolved solids concentrations and P concentrations can help identify the mechanisms behind the relationship between oil and gas wells and elevated P concentrations in groundwater.

4.4. Policy implications

The identification of agricultural land as areas likely to have high P concentrations in groundwater in Canada suggests there may be policy-based and commercial mitigation opportunities [58]. Examples of mitigative action include limits on phosphorus application to the soil [3, 59] or the use of more advanced fertilizer formulations [60] to prevent phosphorus overfertilization in agricultural areas which may result in P loss to runoff and leaching to groundwater.

Currently, there are no legally imposed limits in Canada or the US to regulate the amount of phosphorus in drinking water, groundwater or water for use in agricultural industries, such as irrigation [61–63]. Phosphorus limits imposed on surface water bodies are implemented on a provincial basis in Canada, meaning that some provinces such as British Columbia and Manitoba have limits, while others, such as Saskatchewan, do not [64–66]. In Alberta, previously published numerical TP limits were redacted in 2018 and narrative statements have been developed for lakes, rivers and other water bodies [67]. The Ontario government has general numeric guidelines for TP concentrations, which state that TP concentrations should not exceed 0.02 mg P l\textsuperscript{−1} in lakes and 0.03 mg P l\textsuperscript{−1} in rivers and streams [68]. P limits in the US are proposed by individual states and approved by the Environmental Protection Agency (EPA). Currently, 27 states have no criteria for any types of surface waters. Three states (Minnesota, Wisconsin, and New Jersey) have statewide TP criterion for lakes/reservoirs and rivers/streams but lack TP criteria for estuaries. Florida has a statewide TP...
criterion for lakes/reservoirs and estuaries, but only partial TP criteria for rivers/streams, and Hawaii is the only state to have a complete numeric TP criterion for all waterbodies. The US EPA thresholds are generally lower than the CCME recommended range of 0.1 mg P l$^{-1}$, although some regions and water body types have limits as high as 0.14 mg P l$^{-1}$ [69]. Overall, establishing P criteria for groundwater would be helpful for maintaining surface ecological systems and will be an important aspect of establishing robust phosphorus monitoring programs.

5. Conclusion

In all studied regions (12 countries) around the world, groundwater P concentrations can be high enough to pose a eutrophication risk to surface waters, which is increasingly important as ecosystems face stresses from climate change and other anthropogenic activities. Data from Canada and the US show weak correlations between DO concentrations and elevated groundwater P concentrations (>0.1 mg P l$^{-1}$) that indicate anthropogenic factors are likely influencing groundwater TP concentrations. We show that elevated groundwater TP concentrations are more likely to be found closer to crop/pastureland. However, available data are insufficient to determine a relationship between oil and gas wells and elevated TP concentrations and the extent to which oil and gas wells may be acting as a pathway for anthropogenic phosphorus from the surface to reach groundwater. Moving forward, strategic data collection and an understanding of the mechanisms through which anthropogenic sources of P can contaminate groundwater will be useful to establish effective monitoring programs and implement mitigation efforts that preserve water quality and ecosystem health.

Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: https://osf.io/d96ak/?view_only=9c53811eb4d746c0862ea8b1b838b074.

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