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Atmospheric deposition of phosphorus to land and freshwater

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ABSTRACT

We compiled published and newly-obtained data on the directly-measured atmospheric deposition of total phosphorus (TP), filtered total phosphorus (FTP), and inorganic phosphorus (PO_4-P) to open land, lakes, and marine coasts. The resulting global data base includes data for c. 250 sites, covering the period 1954 to 2012. Most (82%) of the measurement locations are in Europe and North America, with 44 in Africa, Asia, Oceania, and South-Central America. The deposition rates are log-normally distributed, and for the whole data set the geometric mean deposition rates are 0.027, 0.019 and 0.14 g m^{-2} a^{-1} for TP, FTP and PO_4-P respectively. At smaller scales there is little systematic spatial variation, except for high deposition rates at some sites in Germany, likely due to local agricultural sources. In cases for which PO_4-P was determined as well as one of the other forms of P, strong parallels between logarithmic values were found. Based on the directly-measured deposition rates to land, and published estimates of P deposition to the oceans, we estimate a total annual transfer of P to and from the atmosphere of 3.7 Tg. However, much of the phosphorus in larger particles (principally primary biological aerosol particles) is probably redeposited near to its origin, so that long-range transport, important for tropical forests, large areas of peatland and the oceans, mainly involves fine dust from deserts and soils, as described by the simulations of Mahowald et al. (Global Biogeochemical Cycles 22, GB4026, 2008). We suggest that local release to the atmosphere and subsequent deposition bring about a pseudo-diffusive redistribution of P in the landscape, with P-poor ecosystems, for example ombrotrophic peatlands and oligotrophic lakes, gaining at the expense of P-rich ones. Simple calculations suggest that atmospheric transport could bring about significant local redistribution of P among terrestrial ecosystems. Although most atmospherically transported P is natural in origin, local transfers from fertilised farmland to P-poor ecosystems may be significant, and this requires further research.

Keywords: atmosphere, deposition, ecosystems, emissions, phosphorus

Introduction

The supply of phosphorus (P), a principal macronutrient, to ecosystems is a major factor governing productivity, especially in long-term ecosystem development.[1,2] The major non-anthropogenic source of P to terrestrial and freshwater ecosystems is mineral weathering, but there may be cases where P deposition is significant or even dominant.[2] According to the literature[2-6], the main components of P emitted to the atmosphere, and thence subsequently deposited, are dust from soils and deserts, marine aerosols, primary biological aerosol particles (microorganisms, dispersal units, fragments and excretions), ash from volcanoes, biomass burning, the combustion of oil and coal, and emissions from phosphate manufacture. Since small but sustained P input fluxes partially determine whether plant productivity is ultimately limited by P or nitrogen (N)[7], estimation of P deposition is important for understanding and modelling the dynamics of natural and semi-natural ecosystems.

Graham & Duce[3] assembled early available information (49 sites) on directly-measured (i.e. samples caught in collectors) P deposition, and used the results to close their estimated global P cycle. They estimated an annual input to the atmosphere, and loss from it, of 4.56 Tg, with 3.21 Tg deposited to land. The overall average deposition rate to the whole land area derived from this is 0.022 g m^{-2} a^{-1}, but if calculated omitting data for high latitudes (as preferred by Graham & Duce[3]), an overall average value of 0.027 g m^{-2} a^{-1} is obtained. This is less than half of the median, 0.063 g m^{-2} a^{-1}, of 20 published direct measurements in a later collation of data[2], but similar to the median values of collations by Gibson et al.[8] (0.033 g m^{-2} a^{-1}, n=33), Tsukada et al.[4] (0.025 g m^{-2} a^{-1}, n=45), and Mahowald et al.[5] (0.028 g m^{-2} a^{-1}, n=86).

In a landmark global modelling study, Mahowald et al.[5] estimated a total atmospheric P deposition of 1.39 Tg a^{-1}, i.e. only about one-third the value of Graham & Duce[3], attributing most atmospheric emission of P to dust mobilisation, with minor contributions from primary biogenic particles, different kinds of combustion, volcanoes, and sea-salts. They calculated a deposition rate to the oceans of 0.56 Tg a^{-1}, implying deposition of 0.83 Tg a^{-1} to land, which corresponds to an average deposition rate of 0.007 g m^{-2} a^{-1} (again assuming zero deposition to high-latitude areas). This simulated rate is only about one-quarter of the values from direct measurement, and so there appears to be disagreement about atmospheric transfers, and the likely inputs of P to terrestrial and freshwater ecosystems. Mahowald et al.[5] suggested that the discrepancy might be explained by emission and deposition involving relatively large (> 10μm) particles not considered in their simulations.

Thus, a key issue is the scale of atmospheric transport. For example, from a study of atmospheric P inputs to Tutuila Island (Samoa), Graham and Duce[9] estimated that only

about 20% of the directly measured deposition was actually a net depositional input, the remainder being recycled material of local biological origin. Such a distinction between external and local inputs makes sense in an island system, but is less obvious for larger land masses, with adjacent terrestrial ecosystems, or lakes, where short-distance movements of larger material might transfer P between neighbouring ecosystems differing in nutrient status.[2]

In the present study we attempted to improve the quantification of different forms of direct P deposition, and search for explanatory driving variables, spatial, temporal, meteorological etc. Our principal effort was to make a more comprehensive collation of published data on P deposition (we found data for 147 sites) and combine them with unpublished data (97 sites) from monitoring programmes for the UK and Germany. We also reviewed results about P sources and forms. We aimed to resolve the global P budget, and to use the results to consider how P deposition might affect the nutrient status of different terrestrial and freshwater ecosystems.

Methods

Collation of literature data

We searched for papers specifically mentioning deposition measurements in their titles and abstracts, and those reporting studies designed to measure terrestrial or freshwater ecosystem P budgets. We also used data compilations made by previous authors (see Introduction). Only annual values of total (unfiltered and filtered) or inorganic P were accepted. By filtering here we mean filter paper or sub-micron filters, not the coarse ones used for the exclusion of wind-blown twigs, leaves, pollen, insects etc. We refer to these three forms as TP, FTP and PO_4-P in the subsequent text. We did not collect separate wet and dry deposition data, since these are quite sparse. If both wet and dry deposition were reported they were summed to obtain TP.

Only open-field sites were considered. Fluxes of P in forest throughfall are nearly always found to be greater than nearby open-field values[10-24], which could be due to the scavenging effect of trees[2,25,26], causing an effectively greater P deposition rate in forests, or to recycling of P within the forest. Because the scavenging effect is neither widely demonstrated nor generally quantified, we did not attempt to include it in our analysis of the data.

In published work, contamination has usually been considered, and data removed when samples have high concentrations of P and other elements indicative of bird strike (i.e. N and K). Some authors have taken additional steps to minimise contamination, and devoted considerable effort to quantifying it. Perhaps the most comprehensive studies have been those investigating wet P deposition in Florida. Pollman et al.[27] went to considerable lengths to obtain data for wet P deposition in Florida, and reported that a flux over the years 1992 to 1996 (0.0075 g m-2 a-1) was 32% lower than a previous study of deposition in 1978-79.[28] However, Grimshaw and Doske[29] reported a flux of only 0.001 g m^{-2} a^{-1} in wet deposition for the period 1992-1993, while Ahn[30] analysed data to remove outliers statistically for Florida for the period 1992-1996 (15 sites) arriving at an average P concentration of 11.8 µg L^{-1}, which translates to a wet deposition flux of 0.016 g m^{-2} a^{-1}. These efforts to determine the same variable in the same area illustrate how results can vary, which may or may not be due to variations in contamination. In a study aimed at identifying and quantifying contamination in measurements of P deposition to a lake, Blake & Downing 2009[31] found appreciable insect contamination in floating collectors placed on a lake surface, but relatively little in collectors placed on the adjacent land; all results used in the present work refer to land-based collectors.

In view of widespread concerns about contamination, there may be a tendency for reported results to be overestimates of the deposition of P that has been transported by atmospheric processes. Overestimation of overall P deposition may also be caused if results are not reported in cases where deposition is too low to be measured. We found one paper that reported P concentrations to be below the detection limit[32], but that detection limit was not given. Newman[2] drew attention to possible losses of P to container walls, which would produce underestimation, therefore errors might not all be in the same direction.

We could not find an objective system to discriminate amongst reported data, and so did not apply any additional criteria to the reported data. Therefore our results represent a combined evaluation, with inevitable uncertainty, and without consistency among sites. For example, variability in the data could arise from differences in collector type and preparation, the frequency of sample collection (and possible exchange processes in the collection bottles), sample storage, the period between collection and analysis, and the contribution of snow to the samples[33].

Additional measurements made in the present work

Results for open-field sites in Germany were obtained using precipitation collectors, which were deployed as described in the International Cooperative Programme (ICP) Forests manual.[34] The collectors in the field contain a pre-filter mesh size 1000 μm. The data for PO_4-P were obtained on filtered samples by different laboratories using either molybdenum blue colorimetric analysis or anion chromatography with suppression. Contaminated samples were identified according to the checks proposed in the ICP Forests manual, which employ an ion charge balance check, and recognition of high concentrations of P, K, NH_4 and alkalinity.

Sampling by the Centre for Ecology and Hydrology (United Kingdom) involved placement of single bulk precipitation samplers each consisting of a 5 litre bottle, 14 cm polyethylene funnel and a debris filter at open-field sites for one to four weeks. Protocols for the preparation of sampling equipment and the deployment of bulk precipitation samplers follow those recommended by the European Monitoring and Evaluation Program (EMEP).[35] Analysis of bulk precipitation samples for total phosphorus was carried out within 28 days of collection. Unfiltered samples were digested in a matrix of $K_2S_2O_8$ and 1N sulphuric acid, with autoclave heating, then phosphorus was determined by molybdenum blue colorimetric analysis carried out on a Seal AQ2 discrete analyser. The limit of detection was 5 μg L^{-1}. Contaminated samples were identified from simultaneously high concentrations of P, NH_4 and K.

175 Weekly atmospheric bulk deposition samples are collected at the ten sites on the
176 terrestrial United Kingdom Environmental Change Network (ECN)[36,37]. Samples are
177 filtered at 0.45 μm prior to the measurement of phosphate-phosphorus, nitrate and
178 ammonium. Phosphate and ammonium concentrations are measured colorimetrically,
179 and nitrate by ion chromatography, although precise methods have varied between the
180 five ECN participant laboratories. The detection limit (DL) for PO_4-P is 1 μg L^{-1} at six of
181 the sites and 5 μg L^{-1} at the other four. Samples with concentrations <DL were
182 estimated as 0.5 x DL. Prior to the calculation of annual P and N fluxes samples were
183 removed if evidence for contamination by a bird strike was provided in the associated
184 quality code information. Weekly N and P fluxes were then estimated by multiplying
185 concentration by sample deposition sample volume and adjusting for funnel area.
186 Phosphorus-phosphate data for each site were sorted by the size of the weekly flux and
187 clear breaks in the cumulative sum curve were used to identify residual extreme outliers.
188 On average a further five additional samples per site were consequently removed using
189 this approach. Total annual N and P fluxes were consequently adjusted upwards by
190 multiplying the sum of weekly fluxes available for a given year by 52/n, where n = the
191 number of weeks in the year for which flux data were available, on the assumption that P
192 and N deposition fluxes for weeks when data were absent (as a result of excluded
193 samples or sample volumes were too low to allow analysis) equated to the average
194 weekly deposition for that year.

195 As with the German sites, monitoring and analysis by Forest Research (United Kingdom)
196 was carried out within the Level II program according to ICP protocols.[34] The collectors
197 in the open-field site are sited 1.5m above the ground. Each collection bottle is wrapped
198 in foil and contained within a ventilated plastic jacket to exclude light and reduce heating.
199 A bird wire attached to the jacket discourages bird perching. The collectors contain a
200 pre-filter with mesh size 1000 μm. Two samples were collected from each site, screened
201 for contamination and bulked before being filtered through a 0.45μm membrane filter
202 and stored at <4^{o}C prior to analysis. Total P was analysed by ICP-OES (Spectro flame,
203 Spectro Ltd) and PO_4-P by ion chromatography (Dionex DX-500). The limit of detection
204 was 1 μg L^{-1}. Contaminated samples were removed according to the checks proposed in
205 the ICP Forests manual, which employ an ion charge balance, Na/Cl balance and nitrogen
206 balance. Samples were also checked for plausibility within site specific ranges.

207 The James Hutton Institute (United Kingdom) analysed filtered bulk deposition samples
208 using the Konelab Aqua 20 discrete analyser or the Skalar facility to implement the
209 molybdenum blue method. The limit of detection was 1 μg L^{-1}. Contaminated samples
210 were identified from simultaneously high concentrations of P, N and K.

211

Results and Discussion

The full data set of annual direct measurements is compiled in Table S1, including references, and the results are summarised in Table 1. The deposition of P has been measured in six continental regions, with a total of 246 locations (Figure 1), and covering the years 1954 to 2012. The new compilation is an improvement on previous ones in that there are more sites and different forms of P are distinguished. Although the data cover most parts of the Earth's land surface, they are biased towards sites in Europe and North America, which between them account for 82% of the collector locations. It can also be noted that nearly all measurements refer to natural or semi-natural locations, mostly having been taken in studies aimed at understanding P inputs to ecosystems that might respond to P deposition. Thus they are probably biased and so extrapolation of the results to all land areas (see below) can only be regarded as an approximation. For example, there do not appear to be any measurements at places close to major dust sources such as the Sahara desert, the single largest source of dust to the atmosphere[38], where simulated TP deposition[5] is high, *c.* 0.1-0.5 g m^{-2} a^{-1}.

The average TP deposition rate of 0.043 g m^{-2} a^{-1} is somewhat greater than most of the values given in the Introduction, although lower than the average value from Newman.[2] However, a simple average is probably not the best value to use, because, taking all the data together, and treating the value for each site as representative (irrespective of the number of years of data), lognormal distributions of the deposition rates are obtained (Figure 2). Therefore geometric mean values provide a better summary statistic, avoiding bias towards larger values. For the entire data set, the geometric mean deposition rates increase in the expected order PO_4-P < FTP < TP. This sequence also applies to the relatively large data sets for Europe and North America, but for the other continental regions the data are too sparse to be generalised (Table 1). The overall geometric mean deposition rate of TP of 0.027 g m^{-2} a^{-1} is nearly identical to the average value that corresponds to the Graham and Duce[3] budget values (see Introduction).

Forms of P in deposition

Taking all the data together, the geometric mean FTP value is 73% of the TP, although there are only five instances of both the variables being determined at the same site. For PO_4-P a more detailed examination is possible because paired data are available for some sites (24 for TP, 21 for FTP), and strong positive relationships are evident (Figure 3). On average, 40% of TP is analysed as PO_4-P, and 59% of FTP. At the highest values of TP, the fraction that is PO_4-P is relatively low, but this applies only to a few points. Although most of the locations for comparison are in Europe (31 of the 45), the data are reasonably well globally-distributed, implying that the relationships may be general.

Thus a good deal of deposited P is either in the form of PO_4-P or is fairly quickly converted to that form after deposition. In either event, it can be concluded that a considerable fraction of deposited P would be readily bioavailable.

Several studies conducted in North America investigated wet and dry TP separately, through the use of collectors that respond to rainfall. The percentage of TP as dry deposition was found to be about 50% at Lake Huron[39], 53% at Haliburton-Muskoka[40], 63% in South Florida[30,41], 75% in Iowa[42], and 80% in Florida[43]. In other studies, Stoorvogel et al.[44] found that 85% of deposition at a site in Cote D'Ivoire was dry deposited, and Luo et al.[45] estimated that dry deposition fell within the range 0.004 to 0.044 g m^{-2} a^{-1} at Lake Taihu, China, while wet deposition was 0.033 g m^{-2} a^{-1}.

A comprehensive attribution of sources of P to a collection site Ashiu, Central Japan, was made by Tsukuda et al.[4] who determined, by comparison with data for other diagnostic elements, that 15±5% of TP was brought by lithogenic dust from East Eurasia, 39±4% was derived from coal combustion in China, and the remaining 47±6% might predominantly be attributed to the contribution of local biogenic particles. Nichols & Cox[46] reported that pollen contributed 20% of the total input of P to a lake in Ontario, while Rolff et al.[47] estimated that 20-40% of P deposition to the Baltic Sea is organic, and Hendry et al.[28] found that 50% of P deposition was organic at a site in Costa Rica.

Variations with site location, time, rainfall, temperature and season

Considering the continental regions in Table 1, there is not much variability, and the numbers of locations are too few to attempt to draw reliable conclusions about spatial variations. The high average values of TP and PO_4-P for Africa arise principally from the data for three sites around Lake Victoria with major local influences[48], and so do not reflect the influence of the Sahara desert.

Results from the two data-rich regions, Europe and N America, were examined for systematic spatial variation. We combined data for the three types of deposition TP, FTP, PO4, by normalising them to the geometric means. This can be justified given the strong relationships of Figure 3, and the advantage is that we have more data points, and all can be used for the analysis. We tested for spatial variation by kriging (Figure S1), which revealed sites in North Rhine-Westphalia and Saxony-Anhalt with high deposition rates, possibly due to local agricultural emissions from livestock farming[49]. There was also some evidence of generally higher deposition rates in central-northern England. A cluster of five locations in the mid-eastern USA had high deposition rates, but the low spatial coverage means that this cannot be considered significant. It can be noted that some of the North American locations are quite large areas, entire states in some cases, and a Canadian region of c. 10^4 km^2.[50] The latter study averaged results from 32 sites

and the results gave a relative standard deviation (RSD) of 75%, which is not much less than the RSD for North America as a whole (Table 1).

For some sites there are data covering up to 19 years. We performed regression analyses of TP and PO_4-P data for locations with 10 or more years' data, but found only six cases (of 65) with significant trends (Table S2), three of which were increases and three decreases. We conclude that there is no systematic evidence of change in P deposition rates over the available time periods. The annual loads vary about as much between years at a single site as do average deposition rates at different sites, which may go some way to explain why neither spatial nor temporal trends are apparent.

We examined trends with mean annual precipitation and temperature (MAP and MAT) using normalised data (Figure S2), combining the three forms of deposited P. There was no relationship to MAP, while a weak (r^2 = 0.02), marginally significant ($p<0.05$), positive dependence of deposition with MAT was found.

A number of papers report seasonal effects of P deposition. In temperate systems the main pattern is for TP depositon to be highest during spring and summer, which has been reported for sites in the Lake Michigan basin[50], Ontario[40], central Alberta[52], Austria[53], Colorado[54] and the Czech Republic.[55] Anderson and Downing[42] found that P deposition in Iowa was high during periods of agricultural planting and fertilization. However Rolff et al.[47] found little seasonal variation for Baltic locations, nor did Fish[56] in New Zealand. Brown et al.[57] found that P deposition occurred mainly during winter in South Africa, dependent upon rainfall. For non-temperate systems there are fewer data. Deposition of P occurred mainly in the wet season (June to November) in Mexico[58], but the dry season was more important in Costa Rica[28] and Venezuela.[59] Deposition was higher during May to October than other months at Lake Victoria[48], which was thought to be due to greater dust during the dry season, and more local burning of vegetation.

Global deposition budget

Given the lack of evidence for continental-scale variation in deposition, and the sparse data for Africa, Asia, Oceania and SC America, it is not justified to assign a representative deposition rate to each continental region. Using the geometric mean global directly-measured value of 0.027 g m^{-2} a^{-1} (Table 1) and a land area of 117 x 10^6 km^2 (omitting high latitudes[3]), we obtain 3.2 Tg a^{-1} for total deposition to land based on the measured values. As noted above, this is the same as the earlier estimate of Graham & Duce.[3] However, the latter authors estimated deposition to the oceans of 1.35 Tg a^{-1} which is likely too high; more recent estimates are 0.56 Tg a^{-1} and 0.35 Tg a^{-1} (refs 5 and 60 respectively). With a compromise value of 0.45 Tg a^{-1}, we obtain a total global annual deposition of 3.7 Tg a^{-1}, of which about 85% is to land and freshwater.

One reason that our estimate is greater than the 1.39 Tg a^{-1} simulated by Mahowald et al.[5] is that they deliberately confined their analysis to small particle sizes (< 10 μm) capable of long-range transport. In particular, they used an input of primary biological aerosol particles (PBAP) that corresponded to 0.164 Tg P a^{-1}. The value of c. 1000 Tg a^{-1} for the PBAP input given by Jaenicke[61], and including material over the complete size range[6], would give a P input of c. 2 Tg a^{-1}, using 0.2% as an average representative value of the P content of plant material[62,63] and pollen.[2] Substituting this higher PBAP-phosphorus input for the Mahowald et al.[5] value, and by implication including short range atmospheric transport, gives a total input of 3.4 Tg a^{-1}, in fair agreement with the 3.7 Tg a^{-1} that we estimate from directly-measured deposition (see above). The significant input of larger biological material is consistent with the seasonality in deposition rates and the substantial organic P in deposition, both mentioned above. The importance of local emission and deposition was highlighted by Newman[2] and Hendry et al.[28] Another possible factor is the higher P content of dust derived from intensively farmed, especially arable, soil, fertilised with P. This dust might contribute to directly-measured P deposition particularly in Europe and North America, and its P content may be greater than the single value of 720 mg kg^{-1} assumed by Mahowald et al.[5] in their global simulations, given that some intensively managed and fertilised soils have P contents of 1000 mg kg^{-1} or more[64-66]. Moreover the fraction of soil lost as dust is the smaller sized material, which is likely to be enriched relative to the bulk soil.[67]

Therefore, within the annual global budget of atmospheric P the majority undergoes short-distance transfers, while the finer material, simulated by Mahowald et al.[5], and most relevant for long-distance transfers, especially to the oceans, is dispersed over large distances. Given that the main difference between our larger input estimate and the fine material of Mahowald et al.[5], is biologically produced, their values for anthropogenic inputs are unaffected. Therefore their estimate that 4.8% of the 1.39 Tg a^{-1} is anthropogenic converts to about 2% of the input of P estimated here. However, they do not include inputs to the atmosphere associated with the extraction and processing activities involved in fertiliser productions, nor the subsequent dispersal of applied fertiliser, which may be especially significant for natural and semi-natural ecosystems near to intensive agricultural areas, and deserves attention.

The importance of P deposition in different ecosystems

Over the past 150 years, anthropogenic enrichment of ecosystems with nitrogen has occurred through fertiliser manufacture and application, and fossil fuel burning, and a substantial increase in N deposition.[68] In contrast, of the P circulating in the atmosphere, only a small proportion is from anthropogenic inputs (see above). Thus, the significance

of P as a nutrient source to ecosystems is more to do with transfers amongst ecosystems, the overall process being a kind of pseudo-diffusion, causing a net flux of P from P-rich to P-poor locations. In other words, P tends to be lost from some places and gained by others, and the net change is the key issue. As discussed above, P in larger, heavier particles can only move over short distances, whereas fine dust can travel thousands of kilometres. Whether or not a given ecosystem experiences a net gain of atmospherically-deposited P depends upon its own store of P, the proximity and pools of P in ecosystems that might supply new P, and its own emissions of P to the atmosphere. Perhaps surprisingly, few, if any, determinations of ecosystem budgets include the last term.

Globally, ecosystems initially acquire P by the chemical and physical weathering of mineral matter, and atmospheric transport contributes to its ultimate transfer to the ocean and burial in sediments. At the first intermediate scale, P in fine dust can be moved across or between continents. Then at sub-continental scales, P is atmospherically transported and deposited between different ecosystems over much shorter distances. Ultimately, atmospheric transfers occur within a single ecosystem, so there is no net gain or loss, although recycling is occurring; the return of P in litter to forest floor is typically in the range 0.2 to 0.5 g m^{-2} a^{-1}.[13,58,69,70,71]

Waters do not lose much P to the atmosphere[2,3,5], and so depositional inputs are essentially net gains. For example Mahowald et al.[5] calculate a depositional P input of 0.56 Tg a^{-1} to the global ocean, but a loss to the atmosphere of only 0.005 Tg a^{-1} in sea-salt spray. Deposition to freshwaters of P from the surrounding terrestrial landscape also represents a net gain, and a number of cases have been reported where this nutrient supply has significant effects, including lakes in Ontario[40], Oklahoma[72], Austria[52], New Hampshire[73], California-Nevada[74], the Rocky Mountains (Colorado)[75], Sierra Nevada, California[76], and the Tatra Mountains and Bohemian Forest of Central Europe[77]. Camarero and Catalan[78] reported that a four-fold increase in TP deposition between 1998 and 2004 P deposition caused a shift from P to N limitation in Pyrenean lakes. Clearly, the more oligotrophic is a lake, the relatively greater will be the effect of atmospherically-deposited phosphorus.

With regard to terrestrial ecosystems at large scales, much attention has been devoted to the contribution of P in dust deposition to tropical forests[25,26,79-81], including remote islands such as Hawaii[82]. Another likely sensitive terrestrial ecosystem is ombrotrophic peat.[83,84] Large areas of peatland, distant from other terrestrial ecosystems, notably in Canada, Scandinavia and Russia, might rely on long-distance dust deposition to supply phosphorus. We did not find any reported measurements of P deposition at such remote peatland sites, and this therefore seems to be a major gap in knowledge, especially

bearing in mind that phosphorus cycling in peatlands also requires further research[85]. Mahowald et al.[5] predict only low rates of dust deposition (<0.001 g m^{-2} a^{-1}) to the peatlands of Canada and N Russia. Atmospheric transport and deposition at regional scales has been reported to be significant for sites in the Rocky Mountains[86], nutrient-poor locations in north-central USA[87], semi-arid desert margins[88], and the Everglades wetlands of Florida.[41]

Short-distance atmospheric transfers of P will have their greatest effects where natural and semi-natural occur in heterogeneous landscapes, which are most prevalent in areas where land use is most affected by human activities. For example, the Land Cover Map of the UK[89] shows that of seven major terrestrial land-use types (broadleaved woodland, coniferous woodland, arable, improved grassland, unimproved grassland, heather, and bog) the median number found in 5x5 km grid squares is four, which implies considerable heterogeneity and scope for short-distance P transfers between terrestrial ecosystems. In Northern England, some ombrotrophic peats (blanket bog) are within 50 km of arable farmlands, and therefore could receive atmospherically-transported P applied as fertiliser. A clear demonstration of short-distance effects is a study of a super-humid forest in the Southern Alps of New Zealand[90], which showed the rejuvenating effect of dust deposition on soil P along an active dust flux gradient downwind of a braided river.

Ecosystems that acquire significant P via atmospheric deposition are also likely to do so via the activities of various living creatures. Most prominent in the literature are reports of the effects of defecation by birds[91-94], but mammals may also contribute, as well as insect migration. These processes operate similarly to deposition in that their effect depends upon the net movement of P between ecosystems, and they would fall mainly into the short-range category of P transfer. They are generally more difficult to quantify than P deposition, except for example in substantial transfers of nutrients from sea to land.[95,96] The results of Portnoy[92] give an annual loading by gull defecation to a freshwater pond in Massachusetts of 0.12 g m^{-2} a^{-1}, more than four times the overall geometric mean TP deposition value (Table 1). Longer-range transport of P and other nutrients by migratory fish can also be significant.[97-99]

Timescales

When considering the longer-term ecosystem processing and utilisation of P, it is pertinent to ask whether contemporary data are representative over time, e.g. over the Holocene. A number of reports show that dust deposition has varied in both amount and size over time. The largest single source of atmospheric dust, the Sahara Desert, only came into existence about 3-4000 years ago[100], while the Bodélé Depression in Chad,

identified as the single biggest source of dust on earth, has been eroded by the wind since the lake dried out around 1000 years ago.[81] A Holocene dust record at Baffin Island, Canada[101], relevant to the eastern Canadian Arctic, could be divided into three periods, with low inputs of dust from 11550 to 7500 BP, then increasing levels to 5000 BP, then higher still and more variable levels. Dust deposition in southern Belgium[102] was high during the periods 800 to 600 BC and from 3200 to 2800 BC, corresponding to cold periods, and was derived from local soils, distal volcanic and desert particles. More recent agricultural changes have had effects: Saharan dust deposition in West Africa increased over the last 200 yrs, compared to the previous 3000 years, due to the onset and expansion of agricultural activities in the Sahel region.[103]

Transfers of P in PBAP, which effectively redistribute P derived from weathering, will have been going on continuously over the Holocene, and before for non-glaciated regions of the Earth. As discussed above, their effects are most likely to have been felt through short-distance transfers. To explore this process, we conducted a thought experiment as follows. Consider two adjacent equally-sized land areas A and B (Figure 4). Area A receives P from mineral weathering. The element accumulates in soil and plant material, and is lost by leaching, erosion, and emission to the atmosphere as PBAP and dust. Area B receives P only from atmospherically transported P from area A, and loses P by leaching, erosion, and emission to the atmosphere, all emissions being transported back to area A. We characterised combined leaching and erosion losses of P, and emissions to the atmosphere, with first-order constants, i.e. the losses are proportional to the P pools. The model was parameterised by adjusting the weathering rate and the first-order constants (assumed equal for simplicity), so that the P pool in Area A was 50 g m^{-2} (representative of semi-natural topsoils in the UK; unpublished observations) and the average atmospheric emission/deposition rate was 0.027 g m^{-2} a^{-1} (i.e. the global average value from Table 1). The model outputs show that the steady state condition is reached in about 5000 years (Figure 4). By this time the Area B P pool is about 40% of the Area A pool, due only to atmospheric deposition, while the Area A P pool is about 70% of that expected in the absence of atmospheric losses (and returns from Area B). The results suggest that atmospheric transport can redistribute P over timescales which are relatively short compared to ecosystem development. This is of course a highly simplified picture, with over-constrained atmospheric transport, and neglect of the variable distribution of P within the soil profile which will affect leaching and erosion. Nonetheless, the basic premise is reasonable, and could be elaborated to take atmospheric P transport into account in simulations of long-term, large-scale nutrient behaviour and effects.

Conclusions

(1) Literature and newly-obtained data for 246 terrestrial locations and covering the period 1954-2012 gave geometric mean deposition rates of 0.027 (total P), 0.019 (filtered total P) and 0.014 (PO_4-P) g m^{-2} a^{-1}.

(2) Deposition rates of PO_4-P strongly parallel those of TP and FTP, and on average 40% of TP is analysed as PO_4-P, and 59% of FTP, implying considerable bioavailability.

(3) The data revealed no systematic spatial variation in P deposition rates, except for high deposition rates at 11 sites in an area of Germany, probably due to local agricultural emissions from livestock farming.

(4) No generally-significant temporal variations in P deposition, over periods of up to 19 years, were evident.

(5) The global atmosphere receives and loses approximately 3.7 Tg P a^{-1}, only a few percent of which is due to anthropogenic activities. Much of this flux appears to be accounted for by relatively coarse biological material, not considered in the global modelling by Mahowald et al.[5], and this is returned locally.

(6) When considering the effects of atmospherically-transported P on an ecosystem, both inputs and emissions should be considered, to obtain the net gain or loss.

(7) Oligotrophic lakes, tropical forests, and ombrotrophic peatlands are likely to be the most extensive ecosystems affected significantly by net inputs of atmospherically-deposited P.

(8) The global atmospheric transport of dust has varied over the last 10,000 years, but there may have been less variation in the transport of coarser, primary biological aerosol particles, and local transfer of P amongst ecosystems is likely a continual process. Results from a simple model suggest that local transfers effectively redistribute P over the terrestrial landscape.

(9) Research into the atmospheric transport of P from fertilised agricultural land to natural and semi-natural ecosystems is warranted.

Acknowledgements

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Supplementary Information

Table S1 Global phosphorus deposition database

Table S2 Temporal variations of P deposition at different locations

Figure S1 Variation of normalised P deposition with latitude and longitude in North America and Europe

Figure S2 Dependence of log (normalised P deposition) on mean annual precipitation and temperature (MAP, MAT)

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691 Table 1. Summary of deposition fluxes (gP m^{-2} a^{-1}). Key: TP, total P; FTP, total P after filtering; PO_4-P phosphate-P; Sim TP, total P
692 simulated by Mahowald et al.[5]; n, number of different sites; SD standard deviation.

		Africa	Asia	Europe	N America	Oceania	SC America	All
TP	n	10	7	54	38	5	6	120
	mean	0.110	0.020	0.033	0.042	0.030	0.063	0.043
	SD	0.103	0.017	0.031	0.039	0.019	0.062	0.049
	median	0.069	0.017	0.022	0.032	0.036	0.032	0.026
	geometric mean	0.062	0.015	0.022	0.029	0.024	0.043	0.027
FTP	n	0	1	21	5	3	2	32
	mean		0.030	0.029	0.022	0.029	0.019	0.028
	SD			0.040	0.016	0.008	0.016	0.033
	median		0.030	0.023	0.022	0.026	0.019	0.024
	geometric mean		0.030	0.020	0.016	0.028	0.014	0.019
PO_4-P	n	3	4	109	11	3	10	138
	mean	0.067	0.005	0.027	0.019	0.003	0.028	0.026
	SD	0.020	0.001	0.034	0.022	0.002	0.024	0.032
	median	0.068	0.005	0.013	0.007	0.002	0.021	0.013
	geometric mean	0.065	0.004	0.014	0.011	0.002	0.018	0.014
Sim TP	n	10	7	54	38	5	6	120
	mean	0.012	0.006	0.003	0.001	0.001	0.005	0.003
	SD	0.008	0.004	0.002	0.000	0.001	0.001	0.004
	median	0.014	0.006	0.002	0.001	0.001	0.006	0.002
	geometric mean	0.008	0.006	0.003	0.001	0.001	0.005	0.002

693

694 **Figure captions**

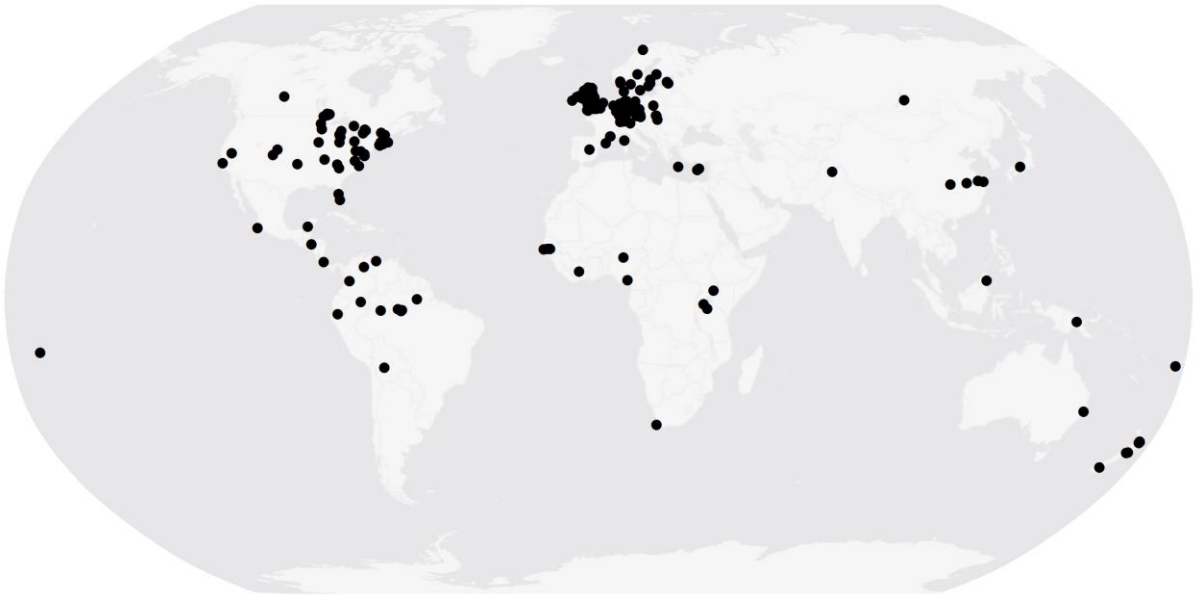
695 Figure 1. Locations with measurements of P deposition

696 Figure 2. Distributions of P deposition values. For each category the values are ordered
697 by increasing magnitude; the y-variable is the fraction of the total locations with
698 deposition values less than or equal to a given deposition.

699 Figure 3. Deposition of PO_4-P vs TP (22 points, circles) and FTP (19 points, squares) for
700 sites where two deposition classes were measured. The line shows the 1:1 relationship.

701 Figure 4. Structure of, and outputs from, a simple model to explore atmospheric P
702 transfers between land areas (upper panel), and plots of P pools over time since the start
703 of the weathering input (lower panel). The weathering input is constant at 0.054 gP m^{-2}
704 a^{-1}. The fractional loss rates of the P pools to leaching/erosion and atmospheric emission
705 are both set to 0.00072 a^{-1}.

706

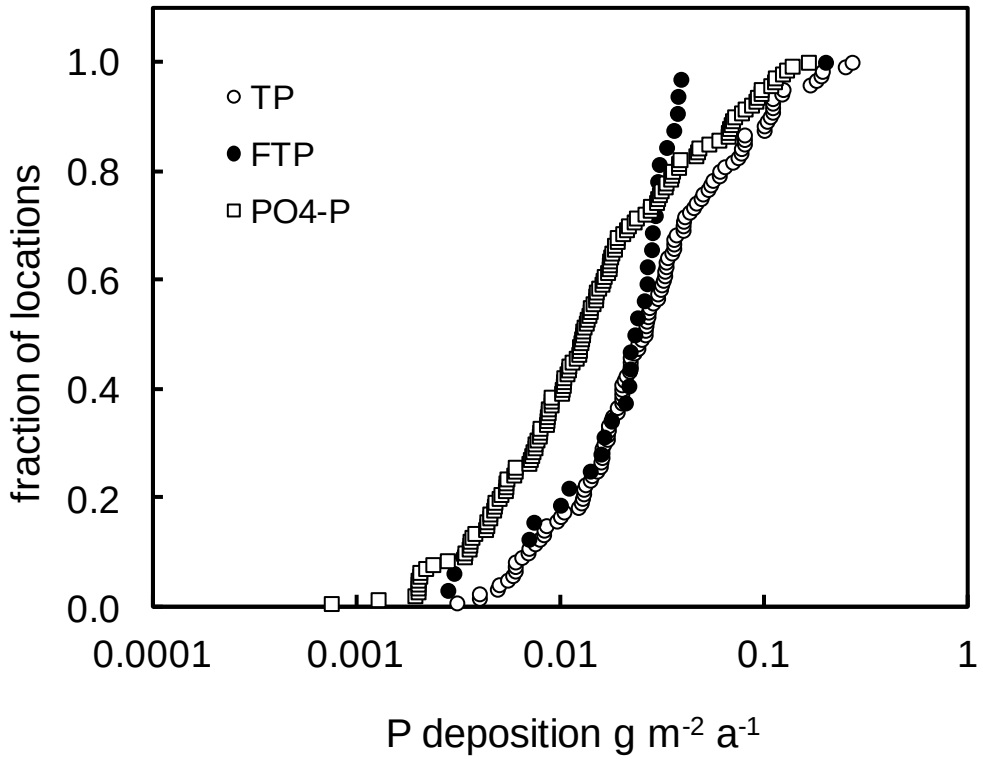


707

708

709 Figure 1.

710

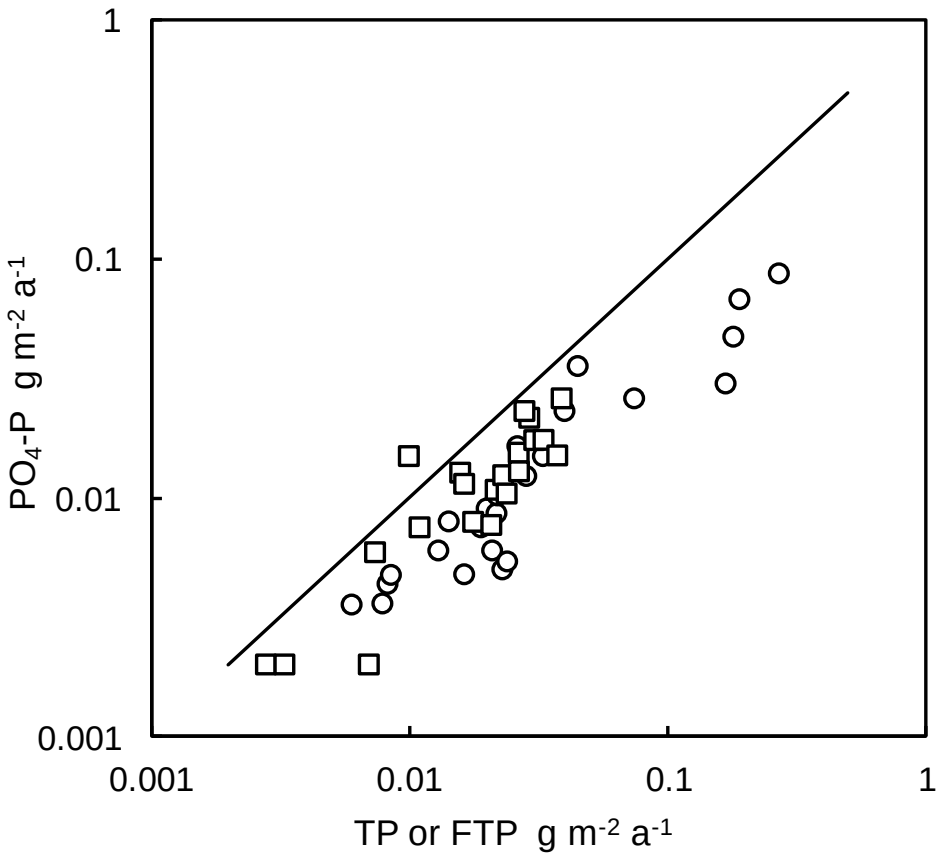


711

712 Figure 2.

713

714



715

716 Figure 3.

717

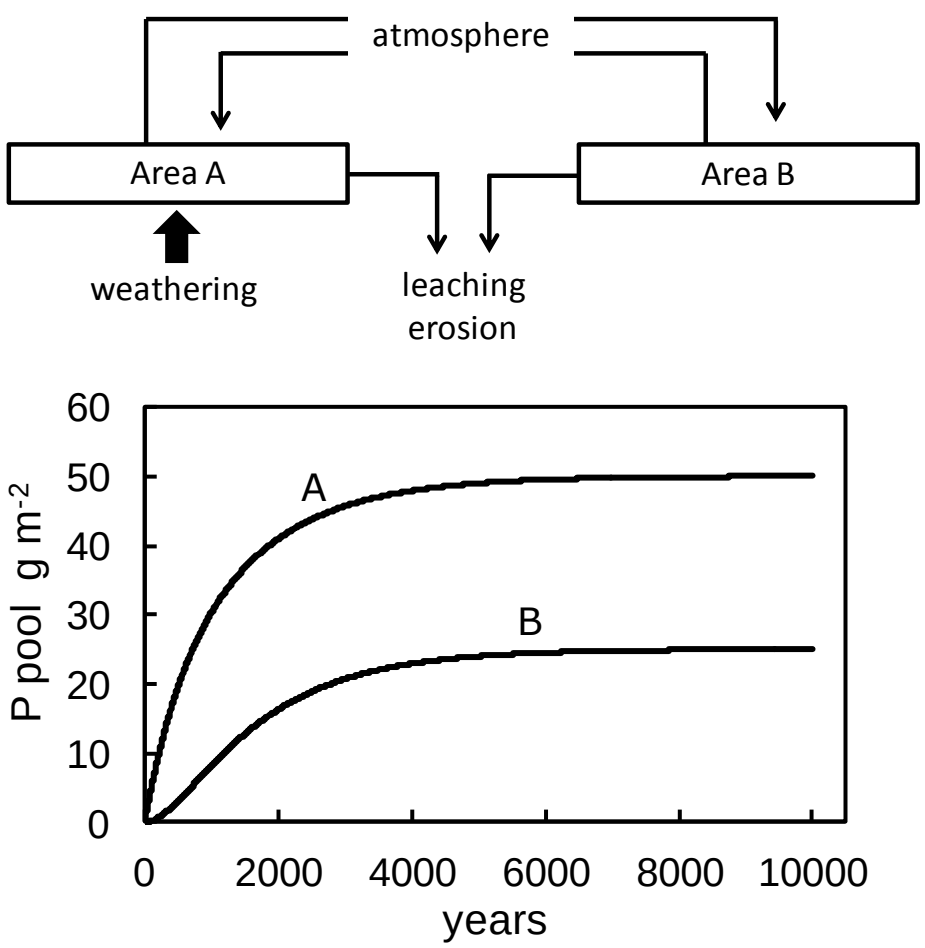

atmosphere
Area A
Area B
weathering
leaching
erosion
P pool g m-2
60
50
40
30
20
10
0
A
B
0
2000
4000
6000
8000
10000
years

Figure 4.