General comment to the Editor

Dear Associate Editor Sönke Zaehle,

We sincerely thank you for re-evaluating our manuscript and thank the reviewers for the significant improvements they promoted on the manuscript through their comments. We responded to each comment and revised the manuscript. We made appropriate changes in the manuscript or have provided an explanation below where we did not make a change suggested by a reviewer. To the best of our understanding, no remaining issues are left in this corrected version.

Responses to the reviewers comments are described below under "**Responses by the authors to the specific comments**". Changes made on the manuscript were all based on these responses. These responses were adapted from the ones provided on the response to the reviewers ('Response to the Anonymous Referees #1 and #2, on 18 Nov 2017). To facilitate the evaluation, we provided the comments made by the reviewers before each response by the authors. Responses made by the authors are in blue and indicated with the terms "***Response to".*** After the responses to the reviewers comments one can find the manuscript with all changes marked (after page 15 on this document). Changes made on the supplementary file are also being provided.

The major changes made on the manuscript were that we focused on specific organic P compounds (i.e. DNA and IHP) instead of its respective functional groups (diester and monoester). All statistical analyses regarding these variables were re-made, and we added a new path analysis regarding inorganic P compounds. New results and discussions were added, and we provided new references to support our discussions. We also updated our data set to November 17, 2017 (previously January 2017). We added more supplementary material as appendices to better explain the random factor effects over the studied variables, and also added the Whittaker's diagram to determine the main biomes comprised in our dataset. We reduced the number of figures on the main document, and have provided them as supplementary material. Finally a native language specialist revised the English. All the other specific changes can be found on the responses to the reviewers or the marked document, both provided below.

Just for clarification, changes were made in two steps, the first was before the language revision, and the second was after the language revision, but the documents regarding the two steps are now merged to facilitate the evaluation process (instead of providing separate documents for each step as we have done in the first attempt to submit the revised manuscript). The alterations made by a non-author refer to the language correction personnel (marked changes made by Cindy Joyac). We apologize for the confusion on the first attempt to submit the revised manuscript. We now followed all requested steps and they are specifically described along this document.

Responses by the authors to the specific comments made by the reviewer 1.

R1 General comment

Organic phosphorus (P) cycling in soils is a topic that has received attention in recent years. As more papers are published, meta-analyses that link the data from these papers together to identify trends in organic P cycling become possible, at least in theory, and a paper presenting novel findings could be of interest to readers. However, deriving meaningful interpretations from a meta-analysis of soil P-NMR studies requires a clear understanding of the P-NMR method and its limitations, in order to correct for known artifacts of analysis. This was not done for this manuscript. As such, it cannot be published in its present form, and will require a major revision, including reanalysis of data, to make it publishable.

Response to R1 General comment

We understand the point the reviewer made about using the correction for potentially degraded peaks (of diesters converted to monoesters). Just to clarify, we did not use the correction previously because 39% of inositol phosphate (comprehending all tropical results and other locates) and 12% of DNA results were absent from the compiled data. We knew that correction was possible through adding to the total diesters concentration, the α - and β -glycerophosphate concentrations (potentialy degraded peaks), but the reviewer also provided additional details that could improve our analysis. To address the issue, we followed reviewer suggestion. Using the available data, we focused on specific organic P compounds (i.e. DNA and IHP) instead of its respective functional groups (diester and monoester). Given the huge proportions of potentially degraded peaks (non-inositol monoesters), and the uncertain about which compounds were present in this potentially degraded fraction, we choose to not to work with the corrected di-to-mono ratio, focusing on DNA and IHP compounds instead.

R1 Comment 1.

Writing quality: a) The quality of English in the manuscript is poor in many places. If the authors revise this manuscript, I suggest they have it read by someone more familiar with English, who also understands the research field. b) Please check that you are using the correct spelling of the names of authors whose papers are cited. For example, “Vincent” is repeatedly cited as “Vicent”, including in the supplemental files. c) Be specific with terminology. The term “P” is an abbreviation for the element for phosphorus. However, the authors use it interchangeably for phosphate, which is incorrect.

Response to R1 Comment 1

a) In the new manuscript version, a native language specialist revised the English.

b) “Vicent” will be replaced by “Vincent”, and all the names of the other authors whose papers are cited were checked.
c) In the new manuscript version, the terminology was revised regarding the proper use of abbreviations. “P” was used as an abbreviation for the element phosphorus, Po and Pi were used for the respective organic and inorganic pools, and the other P compounds were described by their proper names.

R1 Comment 2.

As P-NMR has become more widely used to characterize soil P forms, enough data has become available to indicate the possibility of using these data in meta-analyses to look at soil factors controlling P forms, especially organic P. However, those of us who use this technique the most also recognize its limitations. Although the use of P-NMR has advanced our understanding of soil organic P cycling more than almost any other method to date, the technique is not perfect. It is important to understand the artifacts of the method. It is also important to separate P-NMR results on a soil extract from the P forms that would have been present in the original soil sample prior to extraction. After all, isn’t that the objective of a soil science study? Unfortunately, it isn’t clear to me that the authors of this manuscript are familiar enough with the soil P-NMR technique to understand its limitations and address them. This has produced a study that clearly involved a lot of work by the authors, but which ultimately has not produced any new insights with respect to soil P.

Response to R1 Comment 2

- We recognize that P-NMR can have limitations, and we have addressed them in specific parts of the manuscript. We emphasized those limitations according to the suggested comments.
- Regarding the separations of P-NMR results from other P forms present in the original soil sample, we worked with P-NMR results obtained from NaOH-EDTA extracts only (Y axis on figures 2, 3, 4, 5, and 7, which do not include the residual P, i.e. difference between soil total P and NaOH-EDTA P). The total P of NaOH-EDTA extracts could be obtained by adding Organic P (e.g. figure 3A) to Inorganic P (e.g. figure 2A), but it does not correspond to the soil total P. The total P (obtained with other method – not P-NMR, e.g. digestion) was also presented in the manuscript, but acknowledging that it was obtained by a different method. In the new manuscript version, added more information in the figure captions to avoid misunderstandings, i.e., results in the Y axis are from NaOH-EDTA P-NMR results.

Some specific areas of concern are:
a) Concentration: It is not possible to determine absolute concentrations of P forms or compound classes using NMR; only relative percentages can be determined, because it is a compositional analysis in which the total must be 100%.

Concentrations of P forms are then determined by multiplying by the total extracted P concentration by the percentage of each P form, which is still based on the compositional analysis. This is why the proportions and concentrations of total organic P and total inorganic P (Figs. 2 and 3) show inverse relationships to one another – together they have to add to 100%. This is exactly what would be expected, so it is strange to me that the authors would comment on this (p. 6, lines 13-16). The authors also do not seem to understand the relationship between total P in the soils and P extraction in NaOH- EDTA. In natural (non-tilled) samples, P is stratified, such that concentrations are higher at the soil surface and lower with depth. There will also be an increase in organic P at the soil surface from inputs of plant material, which will decrease with depth – especially in forests with limited mixing and with greater fungal activity in mats in the forest floor (as is typical for temperate forests, where the majority of these studies were conducted). This needs to be accounted for somehow.

Response to R1 Comment 2a

Our total organic and inorganic P results, on mg kg-1 basis, are from NaOH–EDTA extracts only (do not include residual P, i.e. difference between soil total P and total P of NaOH-EDTA extracts). Based on our understanding, the results on mg kg-1 basis were determined from the proportion (%) of each P compound or functional group on spectra (determined by integration of peaks area or deconvolution) multiplied by the total P extracted with NaOH–EDTA. Most authors have presented their P-NMR results (forms and compounds) on both % and mg kg-1 basis (from P-NMR results of NaOH–EDTA extracts), including most of the ones we compiled data from. In the new manuscript version, we added more information on figure captions to state that results on Y axis are from NaOH–EDTA extracts only. Usually, P-NMR results from NaOH-EDTA soil extracts are presented in both ways: (a) on mg kg−1 basis (non-including residual P), and (b) relative distribution of P (%). We followed the same criteria used by those papers to present our results.

We do understand that results are based on a compositional analysis (i.e. P forms are determined by multiplying the total P extracted with NaOH-EDTA by the percentage of each P form), but the description of the inverse relation (obviously a inverse relation) between organic and inorganic concentration (% of total NaOH EDTA P) meant to explore the phenomena of pH or other variable impacting these forms. It was the way we found to describe our results. In the new manuscript version, we reformulated the text avoiding the obviousness on describing results from percentages. In the specific case, the sentence containing "they showed a contrasting behavior" was excluded.

We do understand that soil total P is different than soil P extracted with NaOH- EDTA. We have mentioned that on Page 7 lines 10-13 "It's important to note that the reported total P is the one obtained by digestion and usually comprise the residual P non-recovered by the NaOH EDTA extractant. The recovery of total P by NaOH EDTA extraction is variable depending on soil characteristics and laboratory procedures (Cade-Menun and Liu, 2014)." Moreover, knowing that there is a potential effect of soil conditions and laboratory procedures, we used the P recovery

(percentage of P extracted with NaOH EDTA from soil total P) as a random factor in all bivariate regression models.

We agree that natural (non-tilled) samples have stratified nutrient distributions. Our supplemental Figure S2 presented the results obtained regarding this effect. But contrary to what was expected, we found no effect of sampling depth over organic P concentration in mg kg^{-1}, neither for both organic and inorganic on % basis (even though functional groups of organic and inorganic P responded dynamically to soil depth, even having contrasting responses for organic and mineral soil layers). We did find a sampling depth effect for inorganic P concentration in mg kg^{-1}. Therefore, knowing that there is a potential effect of sampling depth, we used it as a random factor in all bivariate regression models.

b) Extraction efficiency and soil pH: It has been very well established that the recovery of total P from soil samples with NaOH-EDTA extraction is never 100%, and is higher from samples with lower pH. The extraction seems to favor samples high in iron and aluminum, with generally poor P recovery from samples high in calcium; the reasons for this are unclear. As such, any meta-analysis comparing across a range of sample must take into account differences in P recovery among studies, and even among depths within the same soil profile or at different points along a soil chronosequence. For example, the recovery of total P in the samples for the Turner et al. (2003) paper ranged from 14-45%, in the Turner et al. (2007) paper 63-91%, and in the McDowell et al. (2007) paper 11-75%. If the purpose of this meta-analysis is to look at factors controlling soil P, then these differences in recovery must be factored in. Is it even possible to compare the results for a soil where only 11% of the total P was extracted to one with 91% extraction? What about the 89% of total P that wasn't extracted? The authors of this manuscript don't even mention this as a factor, let alone correct for it. And that, unfortunately, undermines their results.

Response to R1 Comment 2b

We do understand that soil chemical characteristics can impact the recovery of P with a NaOH-EDTA extraction. We also agree that an "analysis comparing across a range of sample must take into account differences in P recovery among studies, and even among depths within the same soil profile". We have already addressed that using the 1) P recovery, and 2) sampling depths as random factors (and also latitude for other purpose not directly associated with the comment) in the analysis (which were described in the methods section Page 5 lines 7-15). An example of the impact of a random factor is described in the Page 6 lines 18-20: "There was no pH effect over this inorganic compounds in the organic layer (even though there is an apparent trend, these relationships became non-significant after including sampling depth as random effect on models; Supplementary Appendix S2 shows the sampling depth effect over soil P composition)."

c) Degradation: As noted, it is important for any soil study to ensure that the forms discussed, or the ratios of compound classes such as orthophosphate monoesters and diesters, are based on what was in the original soil sample, and not what was produced during extraction and analysis. It is well established that some orthophosphate diesters such as RNA and phospholipids can degrade to the orthophosphate monoesters α- and β-glycerophosphates (phospholipids) and various monophosphates (RNA) when analyzed at the high pH required for good peak separation in P-NMR spectra [e.g. Turner et al. 2003; Doolette et al. 2009; He et al. 2011, Vincent et al. 2013; Schneider et al. 2016. The degree of degradation will vary depending on the length of NMR experiment and other factors [see Cade-Menun and Liu (2014) and Cade-Menun (2015) for more details]. It is essential that these degradation peaks are identified and quantified in order to determine the correct concentrations of orthophosphate monoesters and diesters that were in the original soil sample; doing so improves any comparison of these P forms to other soil properties (e.g. Young et al., 2013; Liu et al. 2013 J. Environ. Qual. 42:1763-1770). Unfortunately, most studies before 2010 did not identify these compounds and correct for degradation. The authors of this manuscript acknowledge that degradation can occur (p. 4), but for some reason have chosen to ignore it, which is a major problem. The issue of degradation MUST be addressed for any study of edaphic and climatic characteristics to have any meaning. If the concentrations of orthophosphate monoesters and diesters were not corrected in the original study, then the authors of this manuscript could have applied some correction factor to compensate. For example, Vincent et al. (2013) note that most non-inositol phosphate monoesters were diester breakdown products (p. 160). The studies used by the authors here all included some measurement of inositol phosphates (at least myo-IHP and scyllo-IHP). As such, the authors could have assumed that those were the only true monoesters, and corrected the remaining proportion of monoesters to diesters. It would have at least been more meaningful that what they did, which was to ignore degradation but then reach the conclusion that the ratio of diesters to monoesters was a significant factor in the study.

Response to R1 Comment 2c

We understand and agree with the reviewer's comment. But, as described in the methods section Page 4 lines 14-16: "We know that it is possible to correct degraded peaks of diesters converted to monoesters (e.g., Young et al., 2013 and Cade-Menun et al., 2010), but since some papers only showed functional groups like monoesters and diesters, and not species (specific P compounds) inside these functional groups, this correction was not done." Not all studies used in this manuscript included some measurement of inositol phosphates (at least myo-IHP and scyllo-IHP).

Specifically, the following papers did not present P species (including myo-IHP and scyllo-IHP) inside these functional groups (monoesters and diesters) are: Celi et al., 2013, n=4; Vincent et al. 2010, n=1; Turner, 2008b, n=1; Turner et al 2003 (native soil sample), n=1; Turner and Engelbrecht, 2011, n=19; Turner et al 2014, n=10; and therefore correction was not possible to be addressed properly based on

our previous knowledge. Some of these authors acknowledge that there is a small contribution of inositol phosphates (most tropical soils) while others have provided no explanation about with they did not present specific P compounds results. Therefore, we thought it will still be biased to assume something that we were not certain of (i.e. amount of inositol phosphates).

As described earlier, to address the issue, we followed the reviewer suggestion. Using the available data, we focused on specific organic P compounds (i.e. DNA and IHP) instead of its respective functional groups (diester and monoester).

According to the gathered data, non-inositol monoesters (potentially degraded peaks, as suggested by the reviewer) corresponded to 66.76 % in average of the total amount of non-corrected monoesters (ranging from 7.8 to 100%), previously reported as total monoesters content, from papers that presented IHP results (n=61). The same non-inositol monoesters (potentially degraded peaks) corresponded to 53.94 % in average of the total NaOH EDTA organic P amount (ranging from 6.47 to 100%) from papers that presented IHP results (n=61).

Based of the results presented by the authors we could not calculate how much of the potentially degraded peaks were: α - and β -glycerophosphate (Doolette et al., 2009), nor RNA and phospholipid (which includes glycerophosphates) (Vincent et al., 2013); which were determined as degraded peaks by those authors.

Therefore, given the proportions, correcting for potentially degraded peaks has a huge impact on the results, and it is a not completely unbiased calculation, since we don't know if all potentially degraded peaks were α - and β -glycerophosphate (Doolette et al., 2009), or RNA and phospholipid (Vincent et al., 2013), so we choose to not work with the di-to-mono ratio.

Inositol plus DNA represented 59.20% in average of total NaOH EDTA organic P (n=51) from papers that presented both DNA and IHP results. Therefore, it is also a huge proportion and could be an unbiased approach for those results.

The reported proportions are not closing exactly due to the different datasets (n = 51 and n=61).

To re-analyze data, IHP was not considered for tropical soil results because they have non-detected concentrations of this compound (but tropical results will be maintained for the other variables).

The following two paragraphs were written just to clarify why we have done the analysis in the previous way.

We tried to be as clear as possible about this issue, as it is written in the Page 4 lines 18-20: "We expect for future researches to provide results of as much soil P species they can find rather than functional groups only, even when species concentrations are low (and describe when species are not detected), what may enable future analysis to avoid possible confounding effects of organic P species inside functional groups (e.g., inositol and monoesters)." So, we believe that some questions will still remain to be addressed regarding soil P composition in terrestrial natural ecosystems, but our manuscript will provide significant and robust information using currently available results from literature.

We understand the importance of what the reviewer is asking for, and recognize that in the manuscript, but as described we could not reach that level of detail due to absence of data (all specific P compounds). We have used an approach used by other authors. The same approach of not correcting for potentially degraded peaks was used in another recent paper, for example, that combined results from pasture soils using P-NMR results of NaOH–EDTA extracts (Nash et al., 2014). Essentially, they did not corrected for any degraded peak to determine the diester-to-monoester ratio, and described that this was out of their scope, but we agree that their approach is also not optimal.

R1 Comment 3.

Selection of studies: The authors indicate in the methods that they were careful in their selection of papers to include in their meta-analysis, such as native vegetation. As such, I am puzzled as to why the Turner et al. 2003 paper was included as the only study from the USA, because it used agricultural soils. And while the abstract and elsewhere in the text indicate a "dataset including 88 sites", these are overwhelmingly biased to sites in New Zealand (59) and Panama (21), which does not cover a range of "temporal, edaphic and climatic characteristics". The sites selected are also mainly from chronosequence studies, which may also have affected the P forms and their relationship to soil properties.

Response to R1 Comment 3

The Turner et al. 2003 paper included most soils under arable cropping, although there was a native site, and this was the one we included in our analysis. We understand that we were not able to cover a vast representative sample, at global level, but we included as much as we could, given the data availability on the literature. This compilation made this study to have the wider geographical coverage on the topic (terrestrial environments with native vegetation - P-NMR results of NaOH–EDTA extracts).

R1 Comment 4.

Introduction:
a) Please include references for all statements of fact, and make sure those facts are correct. For example, p. 1, lines 24-25: "Once P has been dissolved as free orthophosphate" It isn't possible for free orthophosphate to exist in the soil solution; it will still be associated with cations, although as more soluble forms.
b) Be careful with terminology. Page 2, line 1: "inorganic and organic P pools are each composed by fractions or functional groups". No, they are composed of specific P compounds. The term "functional group" is used elsewhere in the introduction. Please indicate what is meant by this term, which isn't one used for soil P chemistry. And note that fractionation measures operationally-defined P pools, rather than specific P forms.

c) Page 2, line 10: Turner 2007 is not cited in the references.

Response to R1 Comment 4

a) In the new manuscript version, all statements of fact were referenced, and it was make sure that those facts were correct. Specifically, "as free orthophosphate" will be excluded from the sentence. In other occurrences we used "available" instead of "free" when referring to P that could be potentially taken up by plants.

b) In the new manuscript version, the statement was reviewed clarifying that inorganic and organic P pools are composed of specific P compounds. "Functional groups" were changed to compounds in the whole manuscript when describing P compounds.

c) It was corrected in the new manuscript version. The correction is "Turner et al., 2007", which was previously cited in other parts of the manuscript.

R1 Comment 5.

Methods:
a) See comments above about site selection.
b) Page 4, lines 14-23: This discussion about degradation belongs in the Discussion section, not the methods section.
c) The authors have made a lot of assumptions here, particularly for soil classification. Please justify these assumptions in the Discussion section of the manuscript.

Response to R1 Comment 5

a) It was answered on Authors' response to comment 3.

b) In the new manuscript version, we moved the part about degradation to the Discussion section.

c) In the new manuscript version, the assumptions about soil classification were justified in the discussion section. The assumptions include:

- The soil total P content depends on both weathering stages and parent material, but generally decreases with increasingly weathered soil orders (Yang and Post, 2011).
- The soil weathering stages classification also takes into account changes in soil P composition, and generally follows the Walker and Syers (1976) conceptual model: there is a gradual decrease and eventual depletion of primary mineral P (mainly apatite P), a decrease of total P, an increase and then decrease of total organic P, and a increase and eventual dominance of occluded P during soil development (Yang and Post, 2011).

- In highly weathered soils, occluded P increases at the expense of organic P through by encapsulation of mineralized P inside of Fe and Al minerals (Crews et al., 1995).

R1 Comment 6.

Results: a) I am puzzled by the phrase "concentration (% of total NaOH EDTA P)", page 6 line 30. Do you mean % or concentration in mg/kg? They are not the same thing, although they are derived from the same data (% of P forms multiplied by extract concentration).
b) As noted above, any results related to total concentrations or percentages of orthophosphate monoesters, orthophosphate diesters and the diester:monoester ratio are meaningless if not corrected for degradation. The authors must remove all reference to uncorrected concentrations and ratios. They could correct them as suggested above, or they could focus on specific P forms (e.g. DNA or IHP).

Response to R1 Comment 6
a) We meant % of NaOH EDTA P in %.

b) We consider to be the same response of "Authors' response to comment 2c)". We did not correct them for degradation in the previous manuscript version. As described earlier, to address the issue, we will follow the reviewer suggestion. Using the available data, we now focused on specific organic P compounds (i.e. DNA and IHP) instead of its respective functional groups (diester and monoester).

R1 Comment 7. Discussion: Given the issues noted above, I am not sure there is anything meaningful in the discussion section, which as written is a review of the temporal, edaphic and climatic characteristics affecting P forms in NaOH-EDTA extracts, rather than in the original soils themselves. This is really unfortunate given the amount of work the authors put into this study. I hope the authors will address these issues. When they do, I expect much of the discussion section to change.

Response to R1 Comment 7
As described above, we focused on specific organic P compounds (i.e. DNA and IHP). Specifically, we deleted discussion about the mechanisms that prompted the inverse response of monoesters and diesters as P limitation increased (since those functional groups results were excluded from the manuscript). Discussion was added about why DNA concentration increased as both P limitation and soil acidity increased in older, more weathered soil systems. Discussion was also added about the increase in inositol phosphates concentrations at more acidic soil environments.

R1 Comment 8.

Figures:
a) The two figures used for Figure 1 were both published elsewhere, and thus are covered by copyright. However, the authors do not indicate anywhere that they have permission to use these figures in their manuscript, which must be obtained from the publishers of the original papers.
b) All figures containing references to total orthophosphate monoesters and diesters, and the diester:monoester ratio (e.g. 3, 5, 7, 8, 9, S4.1, S4.2, S4.3, S4.4) must be corrected for degradation. And all figures will likely change when the authors have normalized the data used in this study for P recovery.

Response to R1 Comment 8

a) In the new manuscript version, we provided the coverage by copyright. License Numbers: 4210920836823 (Elsevier) and 4210930550479 (John Wiley and Sons).

b) The response about the correction for degradation is on "Authors' response to comment 2c)", and regarding the normalization for P recovery is addressed on the "Authors' response to comment 2a)".

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Responses by the authors to the specific comments made by the reviewer 2.

- The writing of the paper needs to be improved. The paper is dense and hard to read like it is.
- The main messages to take home are not clear, these must be highlighted.
- There are to many bivariate graphs that distract to understand the main messages. I would suggest to add most of them for the supplementary material and keep in the main text the ones that are significant and are used to describe main processes in the text.
- The authors present the patterns shown as global, but there is no reference on the role of different biomes and plant communities, which are in turn related to soil properties. Ecological implications for the relations seen are missing.
- Because of the distribution of the dataset, where most of the samples are from New Zealand, the authors should address the associated bias that the data could have.
- The authors consider the weathering status as a temporal proxy (as it is said in the abstract) to be crossed with soil and climate properties. However, weathering status in this paper is defined by soil type, which makes this classification at certain point redundant with soil properties and climate. The authors should clarify this decision.
- To assume organic C as total C is only acceptable in organic soils. This assumption can lead to large errors in calcareous soils.
- Why the path analysis is used to explain exclusively diester/monoester ratio and not other P-form? Is this ratio providing specific information on nutrient state of the ecosystem? Is significant for understanding P-limitation or inorganic control over the P cycle? This should be argued.
- I miss a clear explanation on the role of the basement/parent material.

Response to R2 General comments

- The writing of the paper was improved making it less dense and easier to read, and a native language specialist revised it.

- We believe that the main messages were responses related to the increasing complexity of phosphorus compounds as pedogenesis progress; therefore, we emphasized those aspects across the manuscript to make more clear the take home messages.
- We agree that we have too many bivariate graphs, so we reduced one variable in Figure 3 (phosphonate, adding it as supplementary material), and added both climatic figures as supplementary material (organic and inorganic P vs. climatic drivers, Figures 4 and 5).
- Ecological implications of different biomes and plant communities over soil P composition were added to the manuscript discussion. This was done through determining the main biomes comprised in our data set using the annual precipitation – temperature diagram (whittaker diagram).
- The potential associated bias that the data could have because most of the samples were from New Zealand will be added to the manuscript. We related this discussion with the soil orders and biomes comprised.

- Regarding the redundancy between soil properties and climate with weathering, this is one reason why we used the path analysis. We can control for the redundancy using such statistical analysis. The figure of soil weathering stage relationship with soil age also showed that our classification followed the patterns that could be expected between soil weathering and soil age and between soil weathering and soil C. However, we do not have enough data to base our weathering classification only based on soil age.
-We checked all data from soils with pH higher than 7 and they have measured organic C, instead of total C. We added this description in the Material and Methods section to clarify this issue for readers.
- We agree that it is not explaining the inorganic P composition, but to fill this gap we have determined the path analysis for the inorganic P compounds.
- We added more explanation on the role of the parent material over soil P composition and implications related to the soil weathering stages classification. The soil total P content depends on both weathering stages and parent material, but generally decreases with increasingly weathered soil orders (Yang and Post, 2011). The soil weathering stages classification also takes into account changes in soil P composition, and generally follows the Walker and Syers (1976) conceptual model: there is a gradual decrease and eventual depletion of primary mineral P (mainly apatite P), a decrease of total P, an increase and then decrease of total organic P, and a increase and eventual dominance of occluded P during soil development (Yang and Post, 2011). In highly weathered soils, occluded P increases at the expense of organic P through by encapsulation of mineralized P inside of Fe and Al minerals (Crews et al., 1995).

R2 Specific comments
- The last sentence of the abstract is not telling anything new "organic and inorganic P pools as well as their functional groups composition are determined by distinctive drivers that regulate key ecological governing their presence. . ."
- Pag 2, line 22, which 5 factors?
- Pag 4, line 27, starts a list with "a) " but no more items are listed
- Pag 7, line 25, the no effect of many climatic variables can be related to the geographic bias of the dataset. Should be argued.
- Pag 8, line 10. Is obvious that poorly crystalline Fe and Al, do not correspond to weathering status if we consider the classification status than the authors have used. However, the presence of these oxides can deeply influence the P pools and cycles in Oxisols and Ultisols but also Andosols.
- Pag 9, line 10. This is a too ambitious sentence. There is no information presented in this study about the variability among communities or different biomes. It is not explained neither how some edaphic variables depend on climate.
- Precipitation and moisture index give similar bivariate relations, maybe with one of both variables would be enough.

Response to R2 Specific comments

- The last sentence of the abstract was modified to be more meaningful. It was changed to "We conclude that soil P composition is determined by edaphic and climatic drivers that regulate key ecological processes on terrestrial natural ecosystems. These processes are related to the source of P inputs, primarily determined by parent material and soil forming factors, and after altogether with plants and microbes coexistence, the bio-physico-chemical properties governing soil phosphatase activity, soil solid surface specific reactivity and P losses through leaching, and the P persistence induced by increasing complexity of P organic and inorganic compounds as pedogenesis evolve.
- Pag 2, line 22. We added the five state factors determining soil weathering.
- Pag 4, line 27. The "a)" was remaining from a previous version. It was excluded in the current version.
- Pag 7, line 25. The potential bias promoted by the geographic concentration of the studied sites was added to the manuscript.
- Pag 8, line 10. We added this explanation to the manuscript.
- Pag 9, line 10. We attenuated the sentence following the reviewer suggestion. "Unprecedented" was changed to "wide". Moreover, as described in the Response to R2 General comments we will improve discussion about what are the biomes represented in our dataset (whittaker diagram), and will add this information on that specific part of discussion. How edaphic variables depend on climate is being explored on the path analysis, but we will improve discussion on theoretical aspects of those relationships.
- Given the similarity between precipitation and the moisture index, the latter has been excluded from the manuscript.

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Soil phosphorus dynamics on terrestrial natural ecosystems

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Abstract. Soil organic and inorganic phosphorus (P) compounds can be modified by distinctive ecosystem properties. This study aims to analyze soil P dynamics on terrestrial natural ecosystems, relating its organic (inositolinositol hexakisphosphate, DNA and phosphonate) and inorganic (orthophosphate, polyphosphate and pyrophosphate) compoundscompounds with major temporal (weathering), edaphic and climatic characteristics. A dataset including 88 sites was assembled from published papers that determined the soil P composition using one-dimensional liquid state ^{31}P nuclear magnetic resonance of soils extracted with NaOH EDTA. Bivariate and multivariate regression models were used to better understand the soil P. In bivariate relationships, soil P compounds had similar overall behaviors on mineral and organic layers but with different slopes. Independent and combined effects of weathering, edaphic and climatic properties of ecosystems explained up to 86DNA78% (inositol) and 8989% (orthophosphate) of organic and inorganic P compound variationsacross the ecosystems, likely deriving from parent material differences. Soil, particularly pH, total carbon and carbon-to-phosphorus ratio, over climate and weathering mainly explained the P variation. We conclude that edaphic and climatic drivers regulate key ecological processes that determine the soil P composition on terrestrial natural ecosystems. These processes are related to the source of P inputs, primarily determined by the parent material and soil forming factors, and after altogether with plant and microbe coexistence, the bio-physico-chemical properties governing soil phosphatase activity, soil solid surface specific reactivity and P losses through leaching, and finally the P persistence induced by the increasing complexity of P organic and inorganic compounds as the pedogenesis evolveevolves.

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1 Introduction

Phosphorus (P) is a key nutrient in animal, microbial and plant nutrition and 'bears light' to terrestrial ecosystem functioning, regulating primary and secondary productivities (Walker and Adams, 1958; Vitousek et al., 2010). Phosphorus input into an ecosystem derives essentially from the weathering of rocks, with little input as an eolian deposit. Once P has been dissolved from primary minerals, plants and microorganisms access it from the soil solution. Phosphorus goes back to soil as organic materials (Noack et al., 2012; Damon et al., 2014) and is then processed both through physico-chemical and biological reactions, determining its forms and bioavailability for the next cycle loop. Each new cycle loop leads to more complex and less bioavailable P compoundscompounds, ultimately seriously limiting ecosystem productivity in the absence of 'fresh' P input as primary minerals. Quantifying organic and mineral forms and their relative abundance as well as their main drivers among the five state factors of ecosystem functioning (time, parent rock, climate, topography, biota) is crucial to understand the historic and present dynamics of P cycling (Jenny, 1941).

In soils, inorganic and organic PP (P_i and P_o, respectively) pools are each composed by specific P compounds compounds (species). The main P_o compoundcompounds are i) orthophosphate monoester (single ester linkage to orthophosphate) such as inositol phosphates, ii) orthophosphate diester (two ester linkages to orthophosphate) such as ribonucleic acid, deoxyribonucleic acid, lipoteichoic acid, phospholipid fatty acids, and iii) phosphonate (Nash et al., 2014). Inorganic P compoundcompounds are orthophosphate, polyphosphate and pyrophosphate (Cade-Menun and Preston, 1996). Specific phosphatase enzymes are required to transform the different P_o and complex P_i forms into orthophosphate, which is the P compound used by plants and microbes. Obviously, enzyme activity is very sensitive to soil pH with specific enzyme optimum. Phosphomonoesterase is more active in acidic soils while phosphodiesterase is optimized in basic soils (Turner and Haygarth, 2005). Other soil variables are also involved in regulating Pi and Po transformations. Inositol phosphates bind strongly to metal oxides and other soil components, which strongly contrained their bioavailability (Turner et al., 2007). Amino group protonation of adenine, guanine and cytosine bases in the DNA molecule can cause adsorption of charged DNA by the charged clay surface (Yu et al., 2013). As such, several soil properties regulate soil P composition but it is unclear which ones predominate.

The absolute and relative abundances of P_o and P_i forms and compounds are likely related to ecosystem development and soil weathering, as conceptualized by the Walker and Syers model (e.g., Walker and Syers, 1976; Yang and Post, 2011) (Figure 1, upper panel). As soil ages (through pedogenesis), ecosystem productivity progresses from nitrogen (N) to P limitation with ecosystem productivity peaking at the N-P colimitation intermediate stage. Parallel changes occur in soil chemistry including total exchangeable bases and soil pH decrease and soil Al and Fe oxide concentration increase (Albrecht, 1957; Walker, 1965). As a result, some P_o and P_i compounds increasingly react with the mineral surface and progressively become occluded P (Yang and Post, 2011). Consequently, complex P_o and P_i forms may increase during ecosystem

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development compared to their simpler forms (McDowell et al. 2007, Figure 1, bottom panel). Soil weathering depends inherently on the state factor 'time' as demonstrated along many chronosequences (e.g. Turner and Laliberté 2015) but it can either increase or decrease through other state factors (Albrecht 1957), such as along climosequences (e.g. Feng et al. 2016) and toposequences (e.g. Agbenin and Tiessen 1995). Along an aridity gradient, increasing precipitations increased total base loss and soil weathering while increasing potential evapotranspiration slowed them down (Feng et al. 2016). However, this study did not focus strictly on P compounds, but on their reactivity (or fractions) through the Hedley analytical procedure, which analyses P release in solutions following a series of acid extraction. Most importantly, we need to investigate the hierarchical nature of causal effects between state factors, soil weathering, soil properties and Po and Pi compositioncomposition.

As an overlooked state factor, parent rock may also determine differences in edaphic properties between soils for a given soil weathering stage. First, parent rocks differ in total P concentration, which may impact absolute P forms. Parent rocks also differ in total exchangeable base concentration and mineral composition, which likely leads to differences in soil pH, soil texture and Al and Fe oxides, all having mandatory control over soil P cycling and Pcomposition. For instance, P_i and P_o absorb and react differently to clays and Ca, Al and Fe oxyhydroxides, which define the soil P retention potential (Batjes 2011). As such, the parent rock factor is important to consider to understand the soil P cycling.

Soil P composition has been studied in soils from ecosystems worldwide, and nuclear magnetic resonance spectroscopy (NMR) was a widely used method to access complex Po and Pi compounds (Kizewski et al. 2011). This technique can be used for both qualitative and quantitative estimates of P species in soil (Cade-Menun and Preston 1996), and the more effective extractant for NMR analysis has been NaOH and chelating agent EDTA (Cade-Menun and Liu 2014). This does not imply that NaOH EDTA is the best extractant for ^{31}P NMR; however, because of its widespread use, it is a good baseline for comparison (Cade-Menun and Liu 2014). According to Cade-Menun and Preston (1996), NaOH can solubilize organic and inorganic P while EDTA chelates metallic cations to increase P extraction efficiency from the soil. The NaOH EDTA extraction method is recognized to quantitatively extract P compounds from the soil because its extract concentrations are similar to other methods considered to provide accurate measurements of the soil P (Turner and Blackwell 2013).

There is a lack of broader understanding of how soil P composition is simultaneously affected by different state factors of ecosystem functioning because known responses were obtained from case-specific conditions. A larger-scale comparative geographical approach could help better understand soil P dynamics. Therefore, we suggest combining the soil organic P results obtained with ^{31}P NMR using NaOH EDTA from different studies, to be able to determine how the soil P composition is determined in terrestrial natural ecosystems. We hypothesize that soil P_i and P_o and compoundtheir compounds can be modified by distinctive edaphic and climatic properties because they regulate key ecological processes

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coupled with soil P cycling. We aim to determine the causal paths through which climate, parent rock and time influence soil properties, and their impact on P_i and P_o pools and specific P compounds.

2 Methods

2.1 Dataset assembling

A search was conducted until November 1717, 2017, to identify published papers that accurately determined soil P species through one-dimensional liquid state ^{31}P NMR on NaOH EDTA extracts. According to McDowell et al. (2006) and Cade-Menun and Liu (2014), we consider accurate the papers that namely estimated an adequate delay time prior to the NMR analysis, therefore enabling the production of quantitative data on the NMR instrument. We used two platforms and specific search terms for each one. The first platform was the Web of Knowledge. The following terms were used: "soil* phosphorus or P or 31P* nuclear-magnetic-resonance or NMR* naoh or sodium hydroxide* edta or ethylenediaminetetraacetic" from which 129 results were obtained. The second platform was Google Scholar. The following terms were used: "soil* phosphorus* "nuclear magnetic resonance"* naoh* edta", which yielded 2,190 results (excluding patents and citations).

We followed pre-defined eligibility criteria to consider the papers, and then to select or reject these papers. The eligibility criteria used to select or reject papers had the following steps: I) Original field native vegetation (native forest or scrub) soil samples (manure, pot soil, soil leachate and sediment samples were excluded), and when studies changed natural conditions, only control (unchanged) was used, i.e., original condition (e.g., litter removal in Vincent et al. 2010 was excluded); II) Samples analyzed using one-dimensional liquid state ^{31}P NMR, according to the following features: a) NaOH EDTA extractor without pretreatment (0.5 or 0.25 M NaOH and 0.1 or 0.05 M EDTA), b) delay times > 2.00 s (i.e., quantitative data, see Cade-Menun and Liu, 2014). Papers that did not show c) NMR features or explanations according to ^{31}P NMR principles (see Cade-Menun and Liu, 2014) using delay times less than 2.0 s were excluded. Papers that did not show d) total NaOH EDTA extracted P and total P were also excluded. Note that we considered both top mineral and organic layers.

The following results were compiled from the papers selected: total P, total NaOH EDTA P, NaOH EDTA organic P and its compound inositol hexakisphosphate (*myo*-, *scyllo*-, *neo*-, and D-*chiro*-IHP, when available), deoxyribonucleic acid (DNA) and phosphonates, NaOH EDTA inorganic P, and its compound orthophosphate, pyrophosphate and polyphosphate.

2.2 Characterization of the sites

Soil texture, total C, total N and pH, short range ordered Al and Fe minerals (poorly crystalline) estimated with oxalate extraction, climate characteristic mean annual precipitation (mm) and mean annual temperatures (ºC) as well as the temporal variable soil age, when available, were also collected from the papers. To extract these results, we used the following assumption: when the total C was unavailable, the organic C was assumed to be the total C. This assumption only occurred

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for non-calcareous soils. No duplicity was found in the papers selected, i.e., results repeated in different papers. Some variables were unavailable for some results and the plot number used for each analysis is presented in the results section. Missing texture and total C data (representing 12 sites and one site, respectively) were extracted from a global soil dataset, SoilGrids, which is now at 250 m resolution (0-20 cm topsoil, Hengl et al. 2017). The resulting dataset is available in Supplementary Appendix S1. We used Whittaker'Whittaker's diagram (Whittaker, 1975) to determine the main biomes comprised in our dataset (Appendix S2). It was created using "BIOMEplot" package (Kunstler 2014).

Soil weathering stages were derived from the soil type according to Cross and Schlesinger (1995) and Yang and Post (2011) as well as from chronosequence positions. A low weathering stage was attributed to Entisol, Mollisols and Inceptisols forming the first stages of chronosequences and gleyed Acrisols. An intermediate weathering stage was attributed to Alfisol, Aridisol, Mollisols and Inceptisols forming the intermediate stages of chronosequences and orthic Acrisol. Finally, a high weathering stage was attributed to Oxisol, Spodosol, Ultisol and humic Acrisol.

2.3 Data analysis

Statistical analyses were conducted on R Version 3.1.0 (© 2014 The R Foundation for Statistical Computing) using mixed-models including edaphic and climatic variables as continuous or categorical fixed effects. The latitude, the percentage of P extracted with NaOH EDTA and the soil sampling depth were considered as random effects. Latitude was used because of the potential biogeographical factors influencing soil P composition once knowing that soil, climate, fauna and flora can modify P dynamics (Turner et al. 2002; Turner 2008a, Turner et al. 2014). The percentage of P extracted with NaOH EDTA was used because the NaOH EDTA extraction process varies according to soil characteristics and experimental conditions (i.e., pretreatment, soil-to-solution ratio and soil characteristics) (Cade-Menun and Liu 2014, see Fig. S5). Sampling depth was used because of potential differences in organic matter inputs in the soil profile. The bivariate effects of latitude, percentage of P extracted and sampling depth on the soil P composition areare presented in Appendices3additional Appendices SS53-S5. Statistical models of soil P compounds were adjusted considering variables as outcome measures in decimal units, where 1 = 100%.
compound.

We used variation partitioning and Venn diagrams (Legendre and Legendre 2012) to partition the total variation explained uniquely by the matrix of soil variables, climate variables or soil weathering stages or variation explained (jointly) by the combined effect of these matrices. The unique effect of soil, climate or soil weathering stages was calculated as the adjusted r^2 value (r_a^2) difference between the full model and unique model. The joint effect of these matrices was calculated as the difference between the summed r_a^2 of unique models and the r_a^2 of the full model.

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Structural equation modeling (path analysis, the '*lavaan*' package) was used to explore how the soil inorganic and organic P compounds variation can best be understood as driven by both direct and indirect effects of key environmental drivers (soil, climate and parent rock), selecting the model that differed least from the observations (*P*-value > 0.0001).

Different units were used across statistics to analyze soil P composition. The bivariate relationships (Figures 2, 3, and 5, and Appendices S3-S8) considered: i) total P_i or P_o concentration in NaOH EDTA extracts (mg kg^{-1} soil), ii) proportion of total P_i or P_o as percentage of total NaOH EDTA P (% of NaOH EDTA P), and iii) proportion of soil P compounds as percentage of their respective pools (% of NaOH EDTA P_i or P_o). Differently, in both Venn diagrams (Figure 6) and structural equation modeling (Figure 7) soil P compounds were in mg kg^{-1}. In bivariate relationships, our objective was to compare the relative composition of P along environmental variables that are linked with the weathering of soils. For that, we used values as percentage. In Venn diagram and path analysis, the objective was to explain soil P composition either partitioning the variation among state factors or accounting for the causal structure of environment. For that, we used the mg kg^{-1} unit so that the distribution of our variables was not constrained as a proportion.

3 Results

Our search resulted in 100 native vegetation outcomes from 13 references (Appendixadditional S1) (Backnäs et al. 2012, n=1; Celi et al. 2013, n=4; Doolette et al. 2016, n=5; Li et al. 2015, n=1; McDowell and Steward 2006, n=4; McDowell et al. 2007, n=26, Turner and Engelbrecht 2011, n=19; Turner et al. 2003, n=1, Turner et al. 2007, n=8; Turner 2008b, n=1; Turner et al. 2014, n=20; VincentVincent et al. 2013, n=8; VincentVincent et al. 2010, n=1). Most of the papers were excluded (from more than 2,000 papers found during the search) because they failed to meet the eligibility criteria including land use (e.g., crop, pasture, planted forest or wetlands) and ^{31}P NMR features. The results selected were from the following countries: Australia (n=5), Finland (n=1), Italy (n=1), New Zealand (n=59), Republic of Panama (n=21), Russia (n=4), Sweden (n=8) and the United States of America (n=1). These results comprised most of the global biomes classified according to Whittaker'Whittaker's diagram (Whittaker 1975), exceptfor the subtropical desert, tundra and temperate rain forest. The six chronosequences studies (5 in New Zealand, 1 in Sweden; 5 on A layer, 2 on O layer) were the most important contributors to the data (45/74 sites on A layer, 18/20 sites on O layer).

In the compiled data, 80% of results were from mineral layers and the remaining 20% from organic layers; 39% did not contain inositol phosphate results (including all tropical regions), and 12% of DNA results were absented (including both non-tropical and tropical regions). All compiled results included the total P, total NaOH EDTA P, NaOH EDTA P_o, and its compound phosphonates, NaOH EDTA P_i, and its compound orthophosphate, pyrophosphate and polyphosphate.

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3.1 Edaphic properties

All edaphic properties affected soil P_i and P_o pools and compounds. These results are summarized in Figures 2 and 3. Both total P_i (Figure 2A) and P_o (Figure 3A) concentrations in NaOH EDTA extracts (mg kg^{-1} soil) had a quadratic response to soil pH, with higher values occurring at an intermediate pH, but only in the mineral layers. No effect was observed in the organic layers. As a percentage of NaOH EDTA P, P_i %(% of NaOH EDTA P) decreased (Figure 2B) and P_o (% of NaOH EDTA P) increased (Figure 3B) as the pH decreased (from right to left), and there was no pH effect on both pools in the organic layers. The compoundcompounds proportions into the P_i (Figure 2C-E) and P_o (Figure 3C-DD) pools responded dynamically to the pH. Into the P_i pool (% of P_i) of mineral layers, the orthophosphate decreased, and pyrophosphate accounted for the remaining P_i as the pH decreased. The pH had no effect eeon these inorganic compounds in the organic layer (even though there is an apparent trend, these relationships became insignificant after including sampling depth as a random effect on models; additional Appendix 3S3 shows the sampling depth effect over the soil P composition). In the P_o pool (% of P_o), both inositolinositol phosphates (mineral layer) and DNA (mineral and organic layers) proportions increased proportionsas the pH decreased. Phosphonate response to edaphic properties (significantinsignificant) is presented Supplementaryin additional Appendix S6.

Both total P_i and P_o concentrations in NaOH EDTA extracts (mg kg^{-1} soil) responded quadratically to the clay concentration, with higher values occurring at intermediate textural classes (Figures 2F and F3E). There was no clay effect on both P_i and P_o and their compoundcompounds proportions (% of NaOH EDTA P) (Figures 2G-J and F3F-H).

Total P_i and P_o concentrations in NaOH EDTA extracts (mg kg^{-1} soil) increased as the soil C concentration increased in mineral layers, whereas in organic layers there was no C concentration effect on P_i and P_o concentrations (Figures 2K and I3I). As a percentage of NaOH EDTA P, P_i decreased and P_o increased (% of NaOH EDTA P) as the soil C concentration increased in mineral layers, and there was no C concentration effect onon both P_i and P_o proportions in organic layers (Figures 2L and I3J). In the P_i pool (% of P_i) of mineral layers, orthophosphate and pyrophosphate proportions decreased and increased, respectively, as the soil C concentration increased (Figure 2M-O). As the soil C concentration increased in the organic layer, orthophosphate decreased, at a greater extent when compared to the mineral layer, pyrophosphate decreased (in contrast to the mineral layer, in which it increased), while the polyphosphate proportion increased, and gradually dominated the P_i pool at greater soil C concentrations. In the P_o pool (% of P_o) there was no C concentration effect on the soil organic P composition (phosphonate, inositol phosphates and DNA) in both mineral and organic layers (Figure 3K-L; additional Appendix S6S6).

Both total P_i and P_o concentrations in NaOH EDTA extracts (mg kg^{-1} soil) from both mineral and organic layers increased as the total soil P concentration increased (Figures 2P and M3M). Only the DNA compoundcompoundfrom the P_o pool (% of

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P_o) in the mineral layer aswas affected by the total soil P concentration (Figure P3P). As the total soil P concentration increased, the DNADNAproportion in the P_o pool decreased. It is important to note that the reported total P (x axis on Figures 2 and 3) is the one obtained by digestion and usually comprises the residual P not recovered by the NaOH EDTA extractant. The recovery of the total P by NaOH EDTA extraction varies depending on soil characteristics and laboratory procedures (Cade-Menun and Liu 2014).

Total P_i and P_o concentrations in NaOH EDTA extracts (mg kg^{-1} soil) were only affected by the soil CP ratio in organic layers. Both total P_i and P_o concentrations decreased as the soil CP ratio increased (Figures 2U and 3Q3Q). As the percentage of NaOH EDTA extract (% of NaOH EDTA P), P_i decreased while P_o increased, both exponentially, as the soil CP ratio increased in mineral layers (Figures 2V and R3R). As proportions iin the P_i pool (% of P_i) of mineral layers, orthophosphatedecreased and pyrophosphate increased as the soil CP ratio increased (Figure 2W-X). In the P_i pool (% of P_i) of organic layers, proportions of orthophosphatedecreased and polyphosphate increased, gradually dominating the P_i pool as the soil CP ratio increased (Figure 2W-Y). In the P_o pool (% of P_oDNA), the DNA proportion increased as the soil CP ratio increased, only in the mineral layer only(Figure 3T3T).

3.2 Climatic properties

Climatic properties affected soil P_i and P_o pools and their composition only through the mean annual precipitation. These results are summarized in Supplementaryadditional Appendices S7 and S8S8. The mean annual temperature, ranging from -0.4 to 27 C^o, did not promote any change on the soil P composition on terrestrial natural ecosystems. There was no effect of climatic variables over total P_i and P_o concentrations in NaOH EDTA extracts (mg kg^{-1} soil) (additional Appendices S7S7A and S8S8A). As a fraction of the NaOH EDTA extract (% of NaOH EDTA P), P_i decreased and P_o increased as theprecipitation increased (AppendicesS7additional Appendices S7B and S8S8B). As theprecipitation increased, proportions of orthophosphate decreased and pyrophosphate increased as compounds of the P_i pool (% of P_i) (additional Appendix S7C-D).

3.3 Soil weathering stages

Soil weathering stages determined from the soil type and chronosequence positions affected the soil age and CP ratio following an expected effect of pedogenesis (Figure 44). As soil weathering stages increased, the soil age and CP ratio also increased. Both P_i and P_o pools were affected by the soil weathering stage (Figure 55). Total P_i and P_o in NaOH EDTA extracts (mg kg^{-1} soil) erewere more concentrated in soils at moderate weathering stages when compared to low and high weathering stages (n=79, Figure 5A,5A, F). As percentages iin the P_i pool (% of P_i), orthophosphate decreased and pyrophosphate increased as the soil weathering stage increased (n=79 for all inorganic compounds, Figure 55C-D). In the P_o pool (% of P_o), DNAthe DNA (n=64) proportion aswasgreater in in more edweathered stages, and there was no effect of weathering stages on phosphonate (n=79) and inositol phosphate (n=52) proportions (Figure 5H-J). Using available data

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(n=49)) we observed no effect of soil weathering stages on short range ordered (poorly crystalline) Al and Fe minerals estimated with oxalate extraction (p>0.1, additional Appendix 9S9).

3.4 Variation partitioning among edaphic, climatic and weathering on the soil P composition

The variation partitioning of the ecosystem's properties governing the soil P composition (in mg kg^{-1} soil) was generally more pronounced for soil variables (pH, clay concentration and total P and C concentrations) than climatic variables (precipitation and temperature) and soil weathering (Figure 66). For the total P_i concentration and its compoundcompound orthophosphate and pyrophosphate, the total variation explained by models ranged from 46% to 889%, and they were mostly explained by soil variables and combined effects of soil and weathering. Polyphosphates had a poorly defined response to the variation partitioning of the ecosystem's properties (<0.01% of the total variation explained).

In the P_o pool, the total variation explained by models ranged from 41 to 86% (Figure 66). The total P_o, inositol phosphates and DNA had theirtheir total variation mostly explained by soil variables, and to a lower degree, but more pronounced for the DNA compound, by combined effects of soil variables and weathering. In contrast, phosphonate had most of its variation explained by combined effects of climate and soil variables, followed by uniquely soil variables.

3.5 Interdependences between environmental variables and soil soil P compounds

We used path analyses to explore the interdependences between edaphic and climatic variables and how they relate to the soil organic and inorganic P compounds (Figure 7; Appendicesadditional Appendix S10). The parent rock was used as a latent variable (set by the pH) in both models (organic and inorganic P). Climate and soil weathering drivers were independently related to soil variables (total P, pH, clay and total soil C), and soil variables were considered direct effects in the models. The most parsimonious path analysis model explained 83up to 78% of P_o compounds variation and 89% of P_i compounds variation and 31%.

Following an expected effect of pedogenesis, the path analysis indicated that the parent rock (latent variable) was positively related to the soil total P, clay and pH. Greater mean annual precipitation was negatively related to the soil total P, pH, and it positively influenced soil total C, while clay was negatively influenced by precipitation into the P_o model only. In the P_o model, precipitation promoted soil weathering, whereas in the P_i model, soil weathering was positively affected by temperature. The mean annual temperature positively affected the clay and pH. Soil weathering was negatively related to the soil pH, and positively related to the soil clay and total C. Into the P_i model only, soil weathering negatively affected soil total P. There were also significant direct and positive effects between soil total C and clay, and total P, in both P_i and P_o models, and there was a positive relationship between soil total C and pH in the P_o model only.

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In the P_i model, orthophosphate was negatively related to precipitation, and it was positively influenced by soil total P and total C. Pyrophosphate had a positive influence of precipitation, soil total P and total C. Polyphosphate was negatively influenced by temperature, and it was positively related to soil pH. affected Srelatedvariablesofand soil pH and soil total PIn the P_o model, inositol was negatively affected by precipitation and temperature, but positively affected by the soil total P, total C and pH. In contrast, total P and total C positively affected DNA, and there were no effects of climatic variables over DNA. Phosphonate was negatively affected by temperature and weathering, but positively affected by precipitation and soil total C.

4 Discussion

Our results showed how soil P_i and P_o compounds respond to edaphic variables (Figures 2 and 3), climatic variables (additional Appendices S7 and S8S8), and soil weathering stages as a proxy for pedogenesis (Figures 55), at awide geographical scale, including a variety of terrestrial natural ecosystems. The soil P composition was generally more influenced by soil variablesand to a lesser degree, but not less importantly, by climatic variables (precipitation), and weathering stages (Figures 6 and 7S7). Soil P_i and P_o compounds responded differently to these groups of factors indicating that each P compound has specific factors governing its presence, transformation and persistence on ecosystems. This could be due to the source of P inputs, primarily by minerals, and then altogether with plants and microbes, the presence of specific phosphatase enzymes that are required to transform P_i and P_o compounds into orthophosphate soil specific reactivity and P losses governed by physico-chemical properties (e.g., clay, short-range ordered oxides, and pH,), and the P persistence induced by the increasing complexity of P organic and inorganic compounds as pedogenesis evolvevolves.

As time passes after the onset of pedogenesis and the ecosystem accumulates organic matter up to a maximum, and then starts to decline, the decaying degree of C element is lower than the P, and concomitantly there is an increasingly acidic environment, resulting in the slowed decomposition of the older soil systems (Walker 1965; Turner and Condron 2013). In addition, parent rock supplies cations and orthophosphate to young soils, whereas more weathered soils are remote from the parent material (Maire et al. 2015). Consequently, highly weathered soils generally have higher CP ratios, a lower pH, and greater clay concentration. The soil total P content depends on both weathering stages and parent material, but generally decreases with increasingly weathered soil orders (Yang and Post 2011). Our data theincluded soil orders ranging from all three stages of soil weathering (low, intermediate and high)), according to Cross and Schlesinger (1995) and Yang and Post (2011). The soil weathering stage classification also takes into account changes in the soil P composition, and generally follows the Walker and Syers (1976) conceptual model: there is gradual decrease and eventual depletion of primary mineral P (mainly apatite P), decrease of total P, increase and then decrease of total organic P, and increase and eventual dominance of occluded P during the soil development (Yang and Post 2011). In highly weathered soils, occluded P increases at the expense of organic P through the encapsulation of the mineralized P inside of Fe and Al minerals (Crews et al. 1995).

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Even though most results were from New Zealand and Panama, our dataset comprised several biomes according to the Whittaker'Whittaker's diagram (Whittaker, 1975)), including the temperate grassland desert, woodland shrubland, temperate forest, boreal forest, tropical rain forest, tropical forest savanna, and intermediates between the temperate rain forest and boreal forest, and tropical rain forest and temperate rain forest (additional Appendix S2). However), however, quantitative data on the feedback between phosphorus compounds and biological communities during pedogenesis is still incipient to conclusions ondrawn from the influence of vegetation and organisms on the soil P composition (Huang et al., 2017), especially for ^{31}P NMR results. What is clearer is how soil P availability shapes the ecosystem's overall primary productivity, and to a lesser extendextent, soil food webs. In a global analysis, Maire et al. (2015) demonstrated that the soil available P is a key environmental dimension increaseincreasing leaf phosphorus content along with species maximum photosynthetic rates and lower stomatal conductance, but a trend that is expected to gradually decline in more weathered soils, due to a lower P availability. conductedConducted at a narrower scale, Laliberté et al.'s study (2017) showed that soil fertility (including P availability) strongly shapes underground food webs, promoting changes such as a shift in dominance from bacterial to fungal energy channels with increasing soil age.

4.1 Soil properties and the soil P composition

As soil aged, pyrophosphate and polyphosphate duemay have accumulated because of the incorporation and stabilization of these compounds (biological origin) into soil organic matter (Turner et al., 2007). The soil pH, total carbon and CP ratio, as well as soil weathering stages had a major role on the soil P_i pool dynamic and composition. As the orthophosphate proportion decreased in more weathered, acidic, organic-rich and P-limited soil environments (Figures 2C, M, W and 55C), pyrophosphate and, or, polyphosphate proportion increased and dominated the P_i pool (Figures 2D, N, O, X and Y and 55D). Even though pyrophosphate and polyphosphate are inorganic compounds, they have a biological origin (Turner and Engelbrecht, 2011). Condensed forms of P (including pyrophosphate and polyphosphates) are found in every bacterial, archaeal and eukaryotic cell, but in highly variable amounts (Kornberg et al., 1999). Bunemann et al. (2008) found a positive relationship between the proportion of fungi and the amount of pyrophosphate, and Reitzel and Turner (2014) found a positive link between the pyrophosphate proportionproportion and soil microbial P. Polyphosphate can originate from ectomycorrhizal fungi (Koukol et al. 2008), and there are some ectomycorrhizal fungi specialized for P uptake in low P_i acidified soil conditions (Wang and Qiu, 2006). Therefore, we believe that pyrophosphate and polyphosphate dominated the P_i pool in acidic, P-limiting (CP ratio) and high organic matter (total C) soils because of the microbial origin of these P sources. These organisms could have helped to deplete and transform the bioavailable orthophosphate, turning it into more complex forms of P as microbial biomass as pedogenesis progressed thosein these environments.

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Moreover, polyphosphates tend to occur in abundance only in soils where decomposition is slowed, such as acidified soil conditions, or cold and wet soils high in organic matter (e.g., Cade-Menun et al. 2000; Turner et al. 2004). Studying wetland soils, Cheesman et al. (2014)found that polyphosphates played a preeminent role in P-limited systems, predominantly in acidic, high-organic-matter systems. Adding to that, pyrophosphate hydrolysis was found to be more rapid with greater biological activity and higher agricultural soilssoil pH (Sutton and Larsen 1964), and this may have contributed to reduce reducing the pyrophosphate proportion at a higher pH in mineral soils (Figure 2D). As the C concentration increased in organic layers, polyphosphate dominated the soil P_i pool (Figure 2M-O), possibly because of its lesser lability when compared to orthophosphate and polyphosphate. Pyrophosphate is less polymerized and potentially more susceptible to hydrolysis than polyphosphate. According to Savant and Racz (1972), Subbarao et al. (1977), and Dick (1985)), pyrophosphate is hydrolyzed more rapidly than polyphosphate because pyrophosphate is an intermediate product of polyphosphate hydrolysis until the final orthophosphate. produced.

As time passes after the onset of pedogenesis, modifications in the soil P_o composition were possibly related to the investment necessary for the P acquisition, and the acidifying environment in soils. microorganismsPlant and microorganism breakdown diesters need a higher investment for the P acquisition than monoesters since they require hydrolysis by both phosphodiesterase and phosphomonoesterase to release available phosphate, whereas monoesters require only the last one, i.e., lower investment (Turner 2008a). According to Turner and Haygarth (2005) and Kunito et al. (2012), P limitation increased phosphoesterases synthesis as a way to increase the organic P breakdown to the bioavailable P. In our results, the DNA proportion increased as soil acidity got stronger and the P limitation increased (Figure 3D, T) in more weathered soil systems (Figure 4 and 5J). Investigations have shown that diester proportionproportions, including phospholipids and DNA, increased as soil aged (Turner et al. 2014; Vincent et al. 2013; Turner et al. 2007; McDowell et al. 2007). Therefore, we hypothesize that as P got scarcer, plant and soil microorganisms may have been stimulated to produce phosphomonoesterases in greater amounts compared to phosphodiesterase because of the lower investment required for the organic P acquisition. Even though acid phosphatases require greater activation energy than alkaline phosphatases (Hui et al. 2013), breaking down diesters would require both enzymes; therefore, a greater investment in energy.

Moreoverthe increasingly acidic pH could have favored phosphomonoesterase activity (Turner and Haygarth 2005), and therefore facilitated DNA accumulation. As demonstrated for temperate pasture soils, phosphomonoesterase activity increased in acidic soil environments, and phosphodiesterase is higher in neutral to basic soils (Turner and Haygarth 2005). Fungi are organismswell known for their capacity to secrete acid phosphatases (Rosling et al. 2016), and are usually the predominant microorganisms in acidic natural soils, while alkaline phosphatase and phytase genes are distributed across a broad phylogenetic range and display a high level of microdiversity (Zimmerman et al. 2013, George et al. 2017). Our findings of pH influence on DNA (Figure 3D) are in keeping with Turner and EngelbrechtEngelbrecht's observations (2011)

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for tropical forest soils, and with Turner and BlackwellBlackwell's observations (2013) for temperate arable soils, where the most acidic soils contained an increasing proportion of P_o as diesters (mostly DNA).

Although inositol phosphates are also classified as part of monoesters, they need a higher investment in organic P acquisition than other monoesters and diesters because they cancan be strongly bounded by metal oxides, clays and organic matter, requiring both solubilization and hydrolysis by the phytase to release a free orthophosphate (Turner, 2008a). This would suggest that as soil gets more weathered, inositol phosphates accumulate extendmore than other organic P compounds. However; however, some authors described that inositol phosphates declined to lower concentrations in older soils (Turner et al., 2014; Turner et al., 2007). They found that for fine textured soils, a decrease in inositol phosphate concentrations (mg kg^{-1}) was mirrored by a decline in amorphous Al and Fe oxides, which may have declined sorption sites for inositol phosphates and weakened protection from plant and microbial enzymatic attack (Turner et al., 2014; Turner et al., 2007). In addition, mineralization of myo-inositol hexakisphosphate by ectomycorrhizal fungi (Chen et al. 2004; Huang et al., 2017) may also have contributed to its decline in more weathered, acidic soils, due to fungi predominance in thosethese environments. In fact, under natural conditions, most tropical soils have negligible inositol hexakisphosphate contribution (e.g., Turner and Engelbrecht, 2011). In contrast, coarse textured soils had an increase in Al and Fe oxide concentrations as soils aged, and inositol phosphate decline was attributed to changes in its inputs into soil, either from plant seeds or microbial synthesis, including through shifts in plant or microbial communities (Jangid et al. 2013, Turner et al. 2012, and Turner et al., 2014).

Inositol phosphate concentration (mg kg^{-1}) may decrease in more weathered, acidic soil systems, mirroring the decline of the soil P_o (mg kg^{-1}) (Figure 3A) and soil organic matter concentrations, but we found that there is an increasing proportion of inositol phosphates in the P_o pool (% of NaOH EDTA P), in acidic soils at, in non-tropical ecosystems (Figure 3C). Therefore, we believe that as pedogenesis progressed, the decaying degree of inositol phosphates was lower than the other monoesters mostly because of the hierarchy of investment for the P acquisition. Nonetheless, inositol phosphates can persist up to a certain point and then decline because there were no inositol phosphates on tropical, more weathered soils. Declining phosphorus availability favorsmay favor organisms' use of inositol hexakisphosphate despite its abundant stabilization potential (Vincent et al. 2013; Turner et al. 2014). Other P compounds such as DNA (Figure 3D) and pyrophosphate (Figure 2D) will prevail onin more weathered systems.

Soil clay concentration affected bothboth soil total P_o and P_i concentrationsconcentrations (Figures F2F and F3E), but had a minor association with soil P compounds. Recent investigations have contradicted the often-cited literature that clay concentration is a major driver of organic compound stabilization, which in fact may be mainly driven by other factors, such as short-range-ordered minerals (i.e., poorly crystalline minerals often estimated with oxalate extraction) that are typically a minor component of the clay-sized fraction by mass, but a highly reactive component. Vogel et al. (2014) showed that

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organic matter is preferentially stabilized in certain hot-spot zones (i.e., rough surfaces), and that only a limited portion of clay-sized surfaces contributed to soil organic matter stabilization. This concept was further tested for P by Werner et al. (2017),) who found that microscale spatial heterogeneity influences P accessibility and bioavailability in soil aggregates, depending on soil substrate and depth. were They also found that in P-rich areas of soil aggregates, the P was predominantly co-located with Al and Fe oxides, while in low-P topsoil aggregateaggregates, majoritymost of the P was organically bound (Werner et al., 2017). Yang et al. (2016) also showed that only limited portions of fine mineral surfaces contributed to soil organic matter stabilization. So, these facts could justify the minor association between bulk soil clay concentration and soil P compounds.

Soil organic matter stabilization could be facilitated in more weathered soils by the potential increase in amorphous Al and Fe oxides (Albrecht, 1957; Walker, 1965), and consequently more reactive surface area availability to sorbabsorb and stabilize soil organic matter. However; however, we did not observe a significant overall soil weathering effect on Al and Fe oxide concentrations (additional Appendix S9), suggesting mightthat it may depend on other factors such as the parent rock material or specific soil orders. Moreover, contrasting soil organic matter responses to short-range-ordered (amorphous) Fe or Al oxides have been found in literature. Some investigations found a pronounced role promoted by Al oxides (Heister, 2016; Kaiser et al., 2016), whereas otherothers found Fe oxides as the main soil organic matter stabilizing mechanism (Wilson et al., 2013; Catoni et al., 2016; Deiss et al., 2017). Investigations also found no apparent relationship between soil organic matter and both Al and Fe oxides (Cloy et al., 2014; Vogel et al., 2015; Rumpel et al., 2016). Therefore, we could not confirm the role of Al and Fe oxides as influenced by soil weathering stages over the soil P composition.

4.22 Climate and the soil P composition

Climatic variables exerted an important role on the soil P compositioncomposition, but to a lesser extent, when compared to soil variables (Figure 66). Contradicting what we expected, our results showed that temperatures ranging from -0.4 to 27 Cº had no effect on both P_i and P_o pools and itstheir compounds (additional Appendices S7 and S8S8). It was expected that the soil organic P concentration would decrease with increasing temperatures, because higher temperatures are optimal for the breakdown of the soil organic P by the microbial biomass through phosphatase enzymes release (Turner et al., 2002). AnoAnother investigation sconfirmed that greater maximum phosphatasephosphatase activity occurred at incubation temperatures >25ºC when compared to 20ºC, but no differences were observed among temperatures greater than 25ºC (Hui et al., 2013). Therefore, phosphatasephosphatase activity may depend on the range and magnitude of temperatures; and our results covered a greater range of markedly lower temperatures, which may reduce microbial activity variability even more due to a slowdown in the microorganisms' metabolism.

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In contrast, precipitation affected several variables in the soil P_i pool. This result was also expected based on the classic paper of Walker and Syers (1976), which suggested that pedogenesis depends predominantly on the volume of water leached through soil. In our results, precipitation was negatively related to the soil total P concentration and pH (Figure 7), and decreased orthophosphate proportion (additional Appendix S7C). As precipitation increased (additional Appendix 7C) and the soil was in a higher weathering stage (Figure 5C), the orthophosphate proportion (% of NaOH EDTA P) possibly decreased because of increased leaching (Walker and Syers, 1976; Feng et al., 2016). Feng et al. (2016) evaluated P transformations along a climosequence and observed that greater soil wetness reduced the inorganic concentration of P linked to Ca, corresponding to a marked decline in soil exchangeable Ca and suggesting an enhanced leaching of P along with weatherable cations. Moreover, greater water availability, and consequent greater primary productivity, may have increased the demand for P in its bioavailable form and contributed to the orthophosphate depletion.

As the orthophosphate percentage percentage decreased following greater precipitation pyrophosphate percentage increased suggesting that this compoundcompound predominates in soils under greater soil wetness. As previously described, this may be due to the incorporation of these compounds into recalcitrant soil organic matter (Turner et al. 2007). Moreover, given the microbial origin of pyrophosphate and its association with the microbial P biomass (Koukol et al. 2008; Turner and Engelbrecht, 2011; Reitzel and Turner, 2014), pyrophosphate (Appendix7additional Appendix 7D) possibly mirrored the response of the organic P (additional Appendix 88B) to climatic variables, which may have resulted from greater soil organic matter accumulation following greater productivity (i.e., plants and organisms) on these ecosystems with greater water availability. Evaluating the phosphorus budget of the whole ecosystem, Turner et al. (2013) demonstrated the dominance of microbial phosphorus in mature soils. Wang et al. (2014) found that greater organic P concentrations were associated with increasing biomass production (i.e., primary production and microbial biomass), because plants and microbes incorporate P into biomass and return it to the soil.

Finally, we expect future research to provide results of as many soil P species as they can find rather than functional groups only (i.e., diesters and monoesters), even when species concentrations are low (and describe when main soil species are not detected), which may enable future analysisanalyses to avoid possible confounding effects of P compounds inside functional groups (e.g., inositol phosphates and monoesters), and to make a more precise correction for potential degraded peaks occurring during the alkaline extraction and reading process. We also urge researchers to determine variances or standard errors for soils with distinctive properties. Then, as stated by Stewart (2010), future analysis cananalyses could use the different information provided by studies of different scopes and quality in a meta-analyticanalytical approach.

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5 Conclusion

We conclude that edaphic and climatic properties are important factors in determining soil P_i and P_o pools as well as their compoundcompounds, since they regulate key ecological processes governing their presence, transformation and persistence on soils. These processes are related to the source of P inputs, primarily determined by parent material and soil forming factors, after altogether with plant and microbe coexistence, the bio-physico-chemical properties governing soil phosphatase activity, soil solid surface specific reactivity and P losses through leaching, and finally the P persistence induced by increasing complexity of P organic and inorganic compounds as pedogenesis evolve. Soil drivers that played a preeminent role were soil acidification, C concentration, P limitation determined as CP ratio, soil weathering as the temporal variable, while precipitation aswas the climatic variables that most influenced soil P composition. Soil P composition was more influenced by soil variables than both climatic variables and weathering stages, however combined effects among these factors also contributed to explain soil P variability on these ecosystems. Therefore, knowing how environmental drivers affect soil P composition enabled a comprehensive understanding of soil P dynamics in terrestrial natural ecosystems.

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6 Appendices (supplementary files)

Appendix S1 – Dataset: The dataset is attached as a supplementary Excel file.

Appendix S2 – Global biomes comprised in our dataset according to the Whittaker' diagram.

Appendix 3S3 – Soil depth effect on soil P composition.

Appendix S4 – Latitude effect on soil P composition.

Appendix S5 – Percentage of P extracted with NaOH EDTA effect on soil P composition.

Appendix S6 – Soil properties and soil organic phosphonates.

Appendix S7 – Climatic properties and soil inorganic phosphorus.

Appendix S8 – Climatic properties and soil organic phosphorus.

Appendix 9S9 – Soil weathering stages and poorly crystalline Al and Fe concentration.

Appendix 10S10 – Models tested to explore the interdependences between edaphic and climatic variables (path analysis) as the main environmental predictors of soil inorganic and organic P compounds.

7 Acknowledgements

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Wilson, C.A., Cloy, J.M., Graham, M.C., and Hamlet, L.E.: A microanalytical study of iron, aluminium and organic matter relationships in soils with contrasting hydrological regimes, Geoderma, 202, 71–81, doi: 10.1016/j.geoderma.2013.03.020, 2013.

Whittaker, R.H.: Communities and Ecosystems, 2nd ed. Macmillan, New York, 1975.

Zimmerman, A.E., Martiny, A.C., Allison, S.D.: Microdiversity of extracellular enzyme genes among sequenced prokaryotic genomes. ISME, 7, 1187–1199, https://doi.org/10.1038 /ismej.2012.176, 2013.

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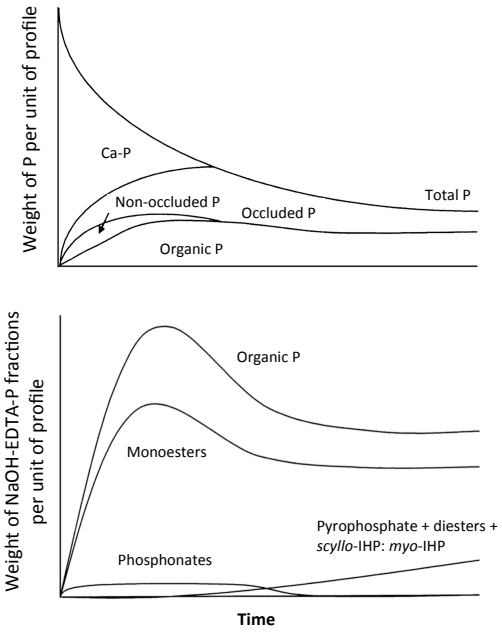

Figure 1: Conceptual diagrams of the changes in soil P fractions (above) and NaOH-EDTA extractable P compoundscompounds (excluding orthophosphate) (below) with time (redrawn from Walker and Syers, 1976, with permission from Elsevier, and McDowell et al., 2007, with permission from John Wiley and Sons, respectively).

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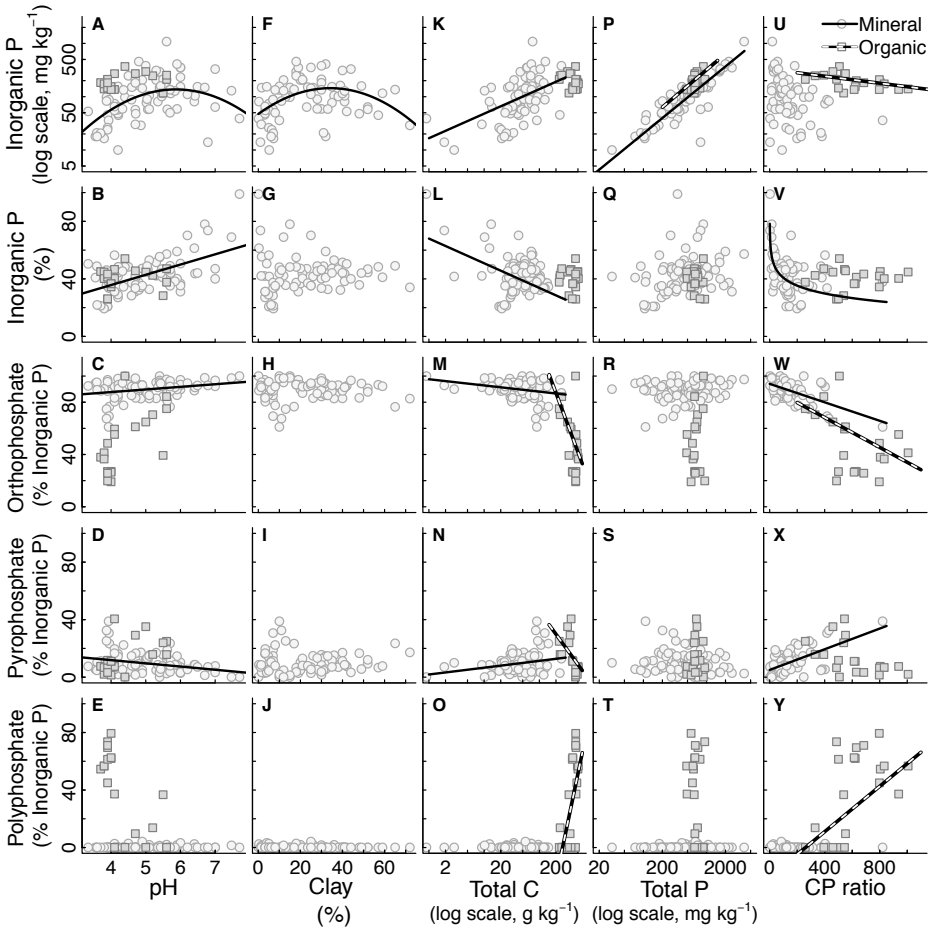
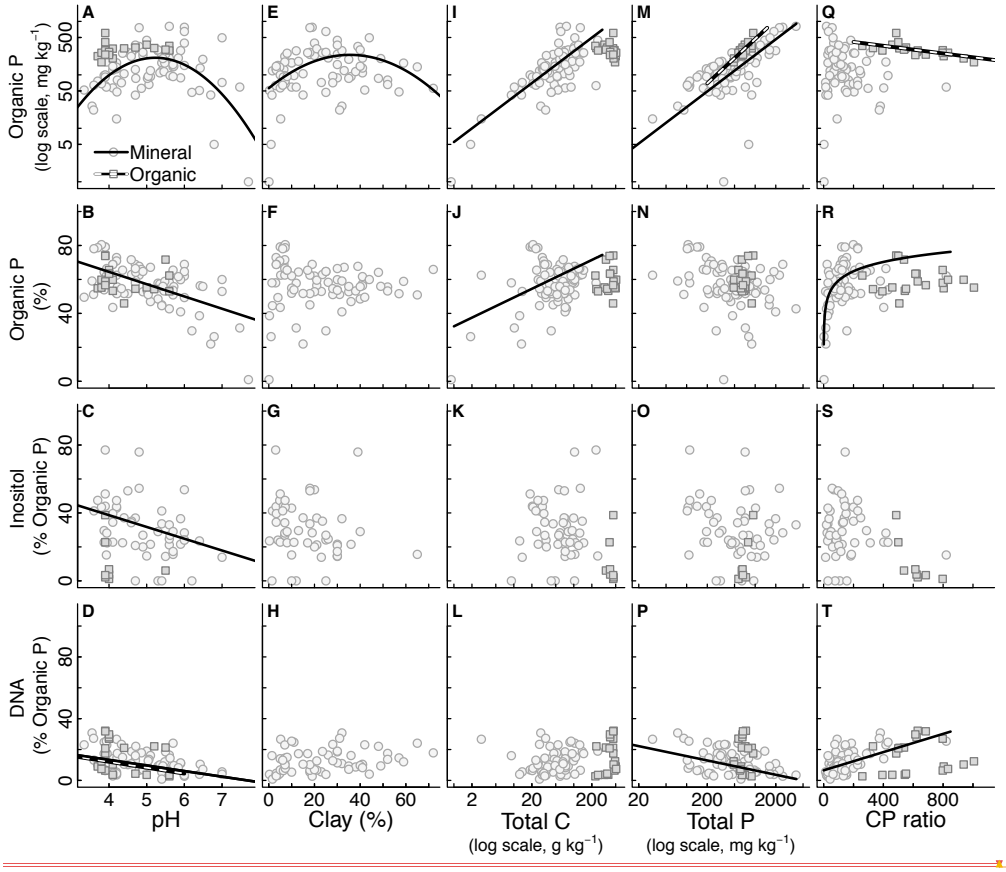


Figure 2: Relationship between edaphic properties and soil inorganic phosphorus (P) composition in NaOH EDTA extract from soil mineral and organic layers on terrestrial natural ecosystems. Note that the reported total P is the one obtained by digestion and usually comprise the residual P non-recovered by the NaOH EDTA extractant. Regression models (n = 80 mineral layer and n = 20 mineral layer): mineral layer, log(total P_i mg kg^{-1}) = -1.62 + 1.28 pH – 0.11 pH ^{2}, r^2 = 0.33; mineral layer, total P_i (%) = 7.21 + 7.12 pH, r^2 = 0.34; mineral layer, orthophosphate = 79.7 + 2.00 pH, r^2 = 0.11; mineral layer, pyrophosphate = 20.8 – 2.23 pH, r^2 = 0.11; mineral layer, log(total P_i mg kg^{-1}) = 1.68 + 0.028 – 0.00041 clay ^{2}, r^2 = 0.23; mineral layer, log(total P_i mg kg^{-1}) = 1.22 + 0.46 log(total C), r^2 = 0.32; mineral layer, total P_i (%) = 68.0 - 17.2 log(total C), r^2 = 0.14; mineral layer, orthophosphate = 97.6 – 4.74 log(total C), r^2 = 0.08; organic layer, orthophosphate = 348.0 – 113.4 log(total C), r^2 = 0.30; mineral layer, pyrophosphate = 1.85 + 4.74 log(total C), r^2 = 0.08; organic layer, pyrophosphate = 151.7 – 53.0 * log(total C), r^2 = 0.34; organic layer, polyphosphate = -446.4 + 184.4 log(total C), r^2 = 0.45; mineral layer, log(total P_i mg kg^{-1}) = -0.63 + 0.97 log(total P), r^2 = 0.73; organic layer, log(total

P_i mg kg^{-1}) = -0.50 + 1.00 log(total P), r^2 =0.27; organic layer, log(total P_i mg kg^{-1}) = 2.51 – 0.00033 CP ratio, r^2 = 0.26; mineral layer, total P_i (%) = 77.0 – 7.88 * log(CP ratio), r^2 = 0.33; mineral layer, orthophosphate = 94.0 – 0.0353 CP ratio, r^2 = 0.37; organic layer, orthophosphate = 91.1 – 0.00057 CP ratio, r^2 = 0.19; mineral layer, pyrophosphate = 5.04+ 0.0359 CP ratio, r^2 = 0.37; organic layer, polyphosphate = -20.5 + 0.079 CP ratio, r^2 = 0.31.



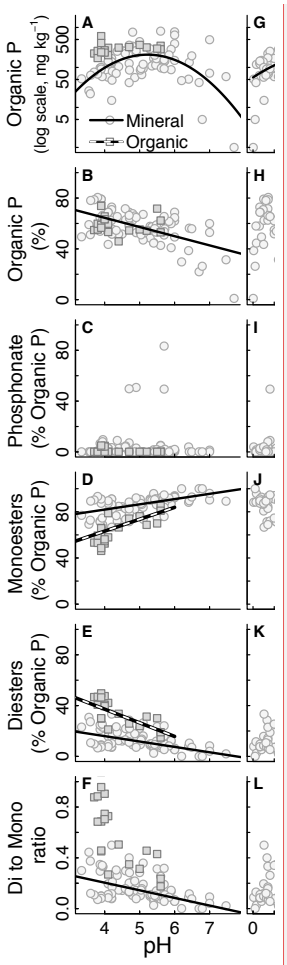


Figure 3: Relationship between edaphic properties and soil organic phosphorus (P) composition in NaOH EDTA extract from soil mineral and organic layers on terrestrial natural ecosystems. Note that the reported total P (x axis) is the one obtained by digestion and usually comprise the residual P non-recovered by the NaOH EDTA extractant. Regression models: mineral layer, log(total P_o mg kg^{-1}) = -3.61 + 2.27 pH – 0.22 pH ^{2}, r^2 = 0.34 (n=80); mineral layer, total P_o (%) = 93.4 – 7.24 pH, r^2 = 0.35 (n=80); mineral layer, inositol =64.6946.4 - 6.94 pH, r^2 = 0.1616 (n=52); DNA 2783319 (n=64)DNA = 27.44 – 3.8383 pH, r^2 =0.10 (n=20); mineral layer, log(total P_o mg kg^{-1}) = 1.75 + 0.035 clay – 0.00049 clay ^{2}, r^2 = 0.16 (n=80); mineral layer, log(total P_o mg kg^{-1}) = 31.2 + 120.5 log(total C), r^2 = 0.60 (n=80); mineral layer, total P_o (%) = 32.4 + 17.0 log(total C), r^2 = 0.12 (n=80); log(total P_o mg kg^{-1}) = -0.55 + 0.97 log(total P), r^2 = 0.29 (n=80); organic layer, log(total P_o mg kg^{-1}) = -0.89 + 1.19 log(total P), r^2 = 0.68 (n=20); DNA 2927DNA = 34.2 – 9.27 log(total P), r^2 = 0.18 (n=64); organic layer, log(total P_o mg kg^{-1}) = 2.69 – 3.58*10^{-4} CP ratio, r^2 =0.48 (n=20); mineral layer, total P_o (%) =23.0 + 7.90 * log(CP ratio), r^2 = 0.33 (n=80); DNA = 6.53 +DNA 653294 (n=64),34 (n=64).

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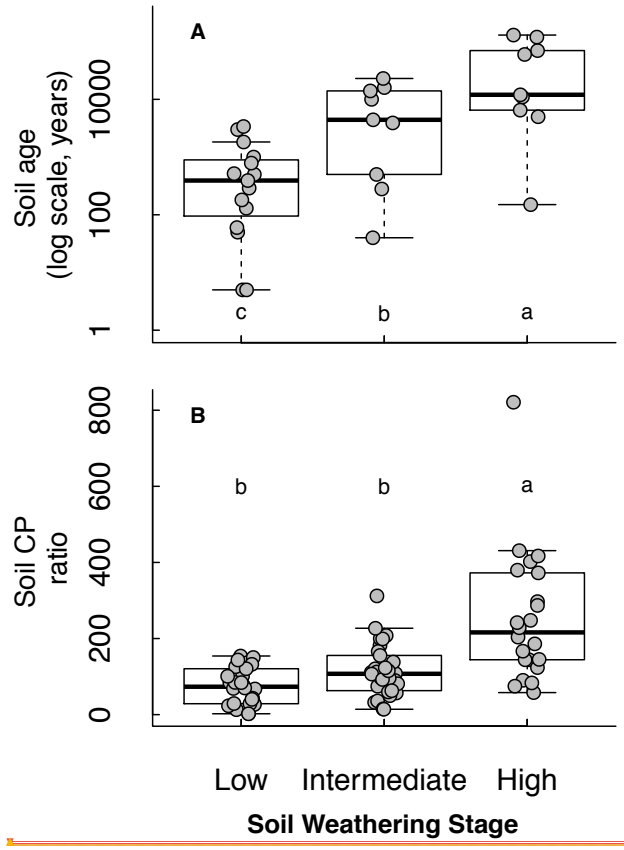

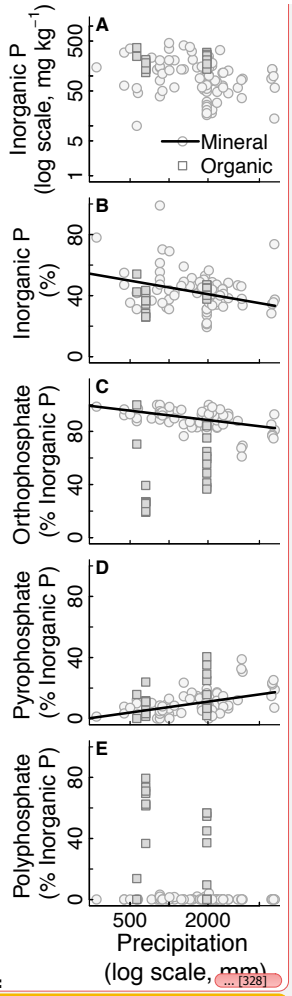

5 **Figure 4: Soil weathering stage relationship with soil age (n = 333) and CP ratio (n = 878) on terrestrial natural ecosystems.**

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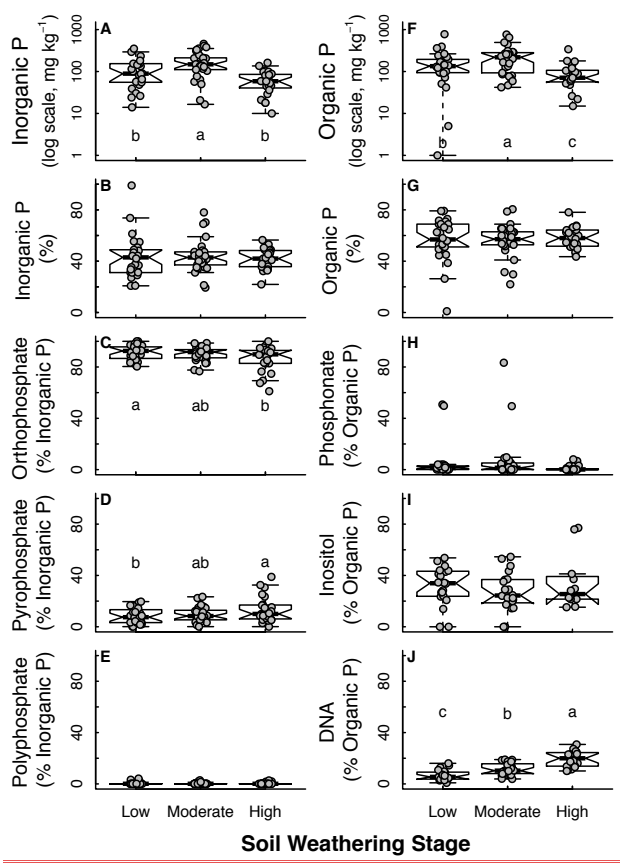

Figure 5: Soil inorganic and organic phosphorus (P) composition in NaOH EDTA extract as influenced by weathering stages on terrestrial natural ecosystems.

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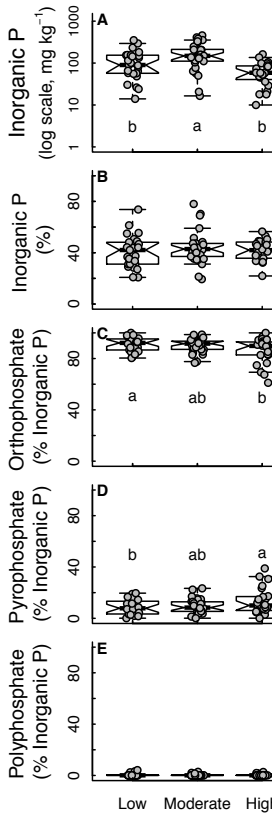

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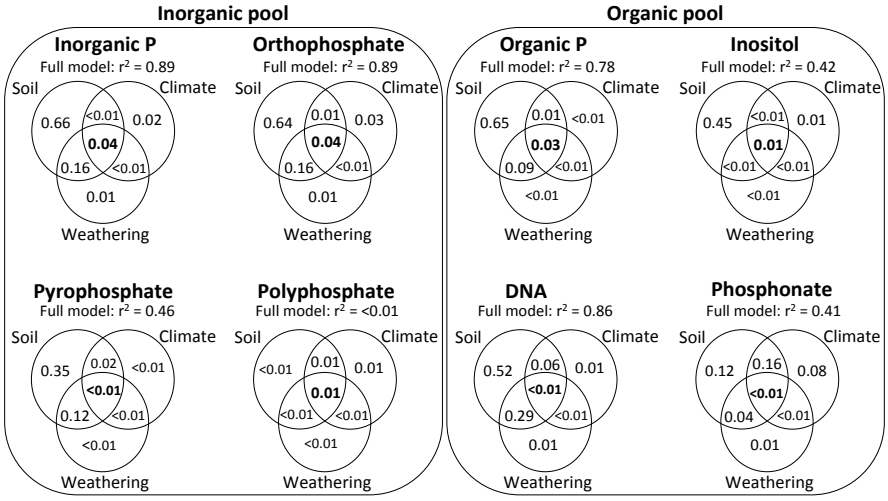
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Organic pool
Inorganic P
Full model: r² = 0.89
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Climate
0.66
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0.01
Weathering
Orthophosphate
Full model: r² = 0.89
Soil
Climate
0.64
0.01
0.03
0.04
0.16
<0.01
0.01
Weathering
Organic P
Full model: r² = 0.78
Soil
Climate
0.65
0.01
<0.01
0.03
0.09
<0.01
<0.01
Weathering
Inositol
Full model: r² = 0.42
Soil
Climate
0.45
<0.01
0.01
0.01
<0.01
<0.01
<0.01
Weathering
Pyrophosphate
Full model: r² = 0.46
Soil
Climate
0.35
0.02
<0.01
<0.01
0.12
<0.01
<0.01
Weathering
Polyphosphate
Full model: r² = <0.01
Soil
Climate
<0.01
0.01
0.01
0.01
<0.01
<0.01
<0.01
Weathering
DNA
Full model: r² = 0.86
Soil
Climate
0.52
0.06
0.01
<0.01
0.29
<0.01
0.01
Weathering
Phosphonate
Full model: r² = 0.41
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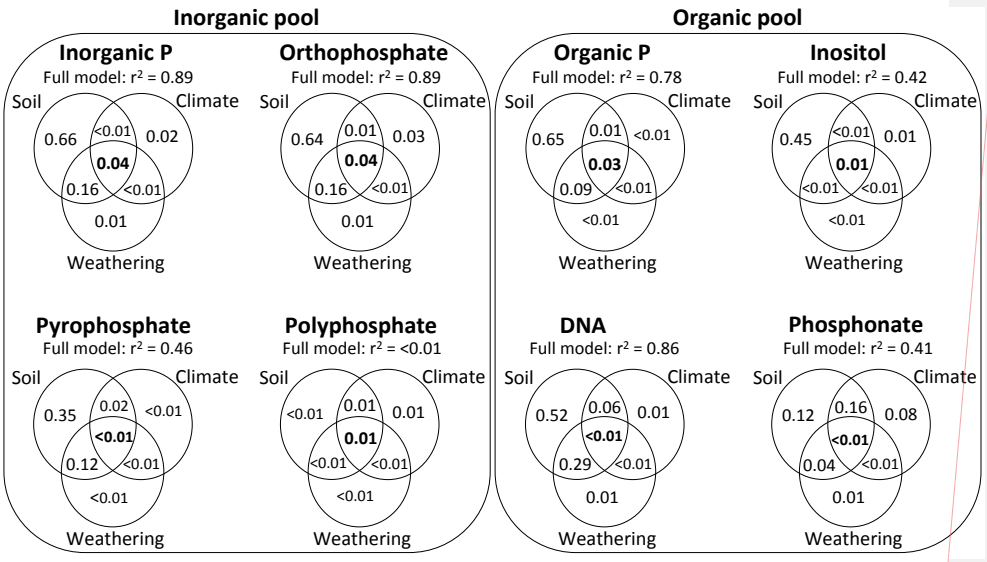

Figure 6: Variation partitioning among edaphic, climatic, and weathering stages on soil inorganic and organic P composition in NaOH EDTA extract on terrestrial natural ecosystems. Soil organic and inorganic P forms and compounds were in mg kg^{-1}, and the other variables followed units described on Figures 2 and 3.

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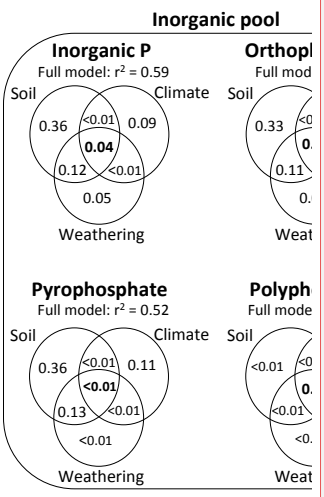

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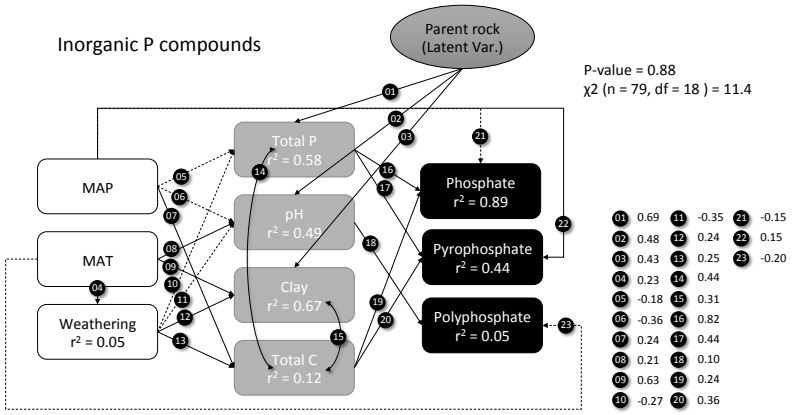

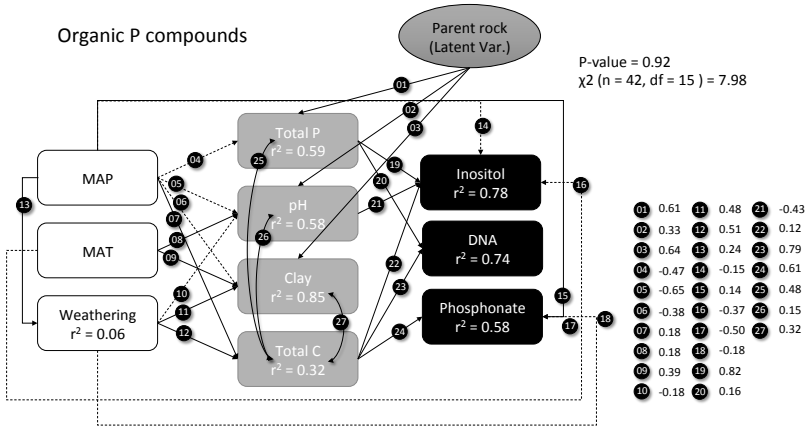

Figure 7: Path analysis describing the direct and indirect effects of the main environmental predictors of soil DNA, inorganic and organic P compounds (mg kg^{-1}) as influenced by edaphic and climatic drivers on terrestrial natural ecosystems. Solid and dashed lines represent positive and negative relationships, respectively. Soil organic and inorganic P compounds were mg kg^{-1}, and other variables followed units described on Figures 2 and 3.

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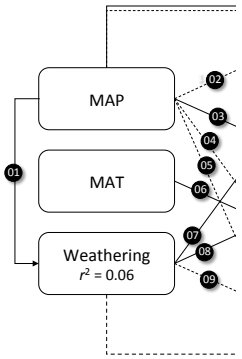

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We conclude that soil organic and inorganic P pools as well as their functional groups composition are determined by distinctive drivers that regulate key ecological processes governing their presence, transformation and persistence on terrestrial natural ecosystems.

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We know that it is possible to correct degraded peaks of diesters converted to monoesters (e.g., Young et al., 2013 and Cade-Menun et al., 2010), but since some papers only showed fractions like monoesters and diesters, and not species inside these functional groups, this correction was not done. Doolette et al. (2009) showed that monoesters α- and β-glycerophosphate are diesters derived species, products of phospholipid hydrolysis occurred during the alkaline extraction and reading process.

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We expect for future researches to provide results of as much soil P species they can find rather than functional groups only, even when species concentrations are low (and describe when species are not detected), what may enable future analysis to avoid possible confounding effects of organic P species inside functional groups (e.g., inositol and monoesters). And researchers must determine variances or standard errors for soils with distinctive properties. Then, as stated by Stewart (2010), future analysis can use the different amounts of information that studies of different sizes and different quality present in a meta-analytic approach.

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The resultant resulting dataset is available in the SupplementarySupplementaryadditional Appendix S1.

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The resultant dataset is available in the Supplementary Appendix S1.

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The moisture index which is the ratio between precipitation and evapotranspiration was considered as another climatic variable that likely impact soil weathering and properties. It was calculated based on Wang et al. (2014a).

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Appendix

Page 5: [80] Moved from page 5 (Move #4) **L D** **1/17/18 12:11 PM**

Statistical models of soil P compounds were adjusted considering variables as outcome measures in decimal units, where 1 = 100%.

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Based on the Wald-type chi-square test, we verified the significance of factor effects significance. Where any factors or interactions effect were was detected, we excluded factors and interactioninteractionsthem of the model, and used only and only used the covariate as model moderator, to test covariate effectthe overall results. of covariate effects overall results. A likelihood ratio test (LRT) was used to test the significance of excluding moderators. Statistical models of soil P

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Statistical models of soil P compoundfunctional groupscompounds were adjusted considering variables as outcome measures in decimal units, where 1 = 100 %.

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functional groupscompounds were adjusted considering variables as outcome measures in decimal units, where 1 = 100 %.

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I'm confused with the mg/kg -- % thing when analyzing the variation of P compounds. First, regarding %, do we consider the perc to total P or to total Pi or Po. Second, do we consider P compounds only with % across analyses ? For instance, for the variation partitioning and the path analysis, I have the feeling that we use the mg/kg unit, do we?
It would be important to be consistent with the unit to use across analyses.
Finally, it would be important to have an argument to focus either on % or mg/kg unit.

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i) Pi or Po mg kg-1: total P amount in NaOH EDTA extract
ii) Pi or Po %: percentage to total NaOH EDTA P
iii) organic P compounds - percentage to total Pi or Po
iv) total P mg kg-1: soil total P in mg kg-1 after acid extraction and digestion

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When these values were normalized as a percentage of NaOH EDTA total P, they showed a contrasting behavior.

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When these values were normalized as a percentage of NaOH EDTA total P, they showed a contrasting behavior.

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When these values were normalized as a percentage of NaOH EDTA total P, they showed a contrasting behavior.

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This response resulted in the increase of diester-to-monoester ratio as pH decreased, and changes occurred in greater intensity in organic layers. There was no pH effect on phosphonates concentration.

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(% of total NaOH EDTA P) concentrations

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(% of total NaOH EDTA P) concentrations

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Inorganic P concentration (% of total NaOH EDTA P)

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Inorganic P concentration (% of total NaOH EDTA P)

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concentrations (% of total NaOH EDTA P)

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concentrations (% of total NaOH EDTA P)

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concentrations (% of total NaOH EDTA P)

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monoesters concentration decreased and diesters concentration as well as diester-to-monoester ratio increased as soil C concentration increased, to a greater extent in the organic layers. There was no soil C concentration effect on phosphonates

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, monoesters concentration increased, and both diesters

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, monoesters concentration increased, and both diesters

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, monoesters concentration increased, and both diesters

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and diester-to-monoester ratio

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and diester-to-monoester ratio

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concentrations

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Into the P_o pool (% of P_o), monoesters decreased as both precipitation (Figure 5D) and the moisture index (Figure 5P) increased and diester-to-monoester ratio increased as the moisture index increased (Figure 5R).

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Page 8: [146] Comment [3] **Maire, Vincent** **1/9/18 11:19 AM**

Here, we should consider the weathering stage the other way around. Precipitation, temperature and time should impact the weathering stage, while the weathering stage should impact edaphic properties. Briefly, I would propose to replace the Fig 4 by a MANOVA, where we try to explain the variation of soil weathering stage by precipitation, temperature and soil age. It would be nice to show that soil age is the main factor to echo the modification I made in the introduction…

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monoesters were more abundant at lower weathering stages, and both diesters and diester-to-monoester ratio

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monoesters were more abundant at lower weathering stages, and both diesters and diester-to-monoester ratio

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monoesters were more abundant at lower weathering stages, and both diesters and diester-to-monoester ratio

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the remaining being explained at lower extent by weathering, climatic variables, and their combined effects.

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Diesters concentration had the greatest amount of variation explained by models achieving 78% of the total variation, being mostly explained by soil variables and combined effects of soil and weathering. The diester-to-monoester ratio had 50% of total variance mostly explained by soil and weathering.

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, but when it was present, the models were consistently far weaker ($p<0.0001$; Supplementary additional Appendices S10.1 and S10.3; S11.1 to S11.3)

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The most parsimonious path analysis model explained 44% of the diester-to-monoester ratio variation, and the causal effects are described in the following. There was no significance on models tested for the soil pyrophosphate + polyphosphate to orthophosphate ratio (chi-square statistics, $p<0.05$); hence they are not presented here (see Supplementary additional Appendix S11).

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;, and the causal effects are described in the following (Figure 7). Climate and soil weathering drivers were independently related to soil variables (total P, pH, clay, and total soil C), and soil varaibles were considered as direct effets in the model.

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The parent rock was used as latent variable, which was fixed by pH in this model.

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The parent rock was used as latent variable, which was fixed by pH in this model.

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that greater mean annual precipitation promoted soil weathering, and negatively influenced reducedthe soil total P, pH and clay

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,. The Mmean annual temperature was positively related to

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soil pH andaffected the clay concentration

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oil. Soil weathering was negatively related to reducedthe soil total P and pH, and positively

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increasedrelated to the soil clay and total C. The parent rock was strongly and positively related to soil total P and pH, and to a lesser extent clay concentration. It's important to note that when parent rock was absent the models were consistently far weaker ($p<0.0001$; Supplementary Appendix S4); hence they are not presented. There were also significant direct and positive effects among all soil

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soil total P that was positively related toofexcept for the soil total C, and soil total C

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that was negatively related to soil pH, which were negatively related. Inositol was negatively affected by precipitation, and soil weathering, but positively affected by the soil C. In contrast, DNA was positively affected by precipitation, soil weathering, soil C and clay, and it was negatively affected by

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Finally, soil diester-to-monoester ratio was positively influenced by both soil weathering and pH.

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Therefore, we assume that that pyrophosphate and polyphosphate dominated the P_i in acidic, P-limiting (CP ratio), and high organic matter soils (total C) because of the microbial origin of these P sources. These

organisms could have helped to deplete and transform the bioavailable orthophosphate, turning it into more complex forms of P as microbial biomass at those environments.

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As soil aged and soil P limitation increased (higher CP ratios) the accumulation of pyrophosphate (Figures 2X and 7D) and polyphosphate (Figure 2Y) could be due to the incorporation and stabilization of these compounds (biological origin) into soil organic matter (Turner et al. 2007). Adding to that, pyrophosphate hydrolysis was found to be more rapid with greater biological activity and higher pH of agricultural soils (Sutton and Larsen, 1964), and this might have contributed to reduce pyrophosphate concentration at higher pH in mineral soils (Figure 2D).

This could be facilitated in older, weathered soils by the potential increase in amorphous Al and Fe oxides (Albrecht, 1957; Walker, 1965), and consequently more reactive surface area availability to sorb and stabilize soil organic matter. However, we did not verify a significant soil weathering effect overall the Al and Fe oxides concentrations (Supplementary Appendix S3), suggesting it might depend on other factors such as the parent rock. Other investigation also found a decline in Al and Fe oxides concentrations in older, fine textured soils (Turner et al., 2007). Moreover, contrasting soil organic matter responses to amorphous Fe or Al oxides have been found in the literature. Some investigations found a pronounced role promoted by Al oxides (Heister, 2016; Kaiser et al., 2016), whereas other found Fe oxides as the main soil organic matter stabilizing mechanism (Wilson et al., 2013; Catoni et al., 2016; Deiss et al., 2017). Investigations also found no apparent relationship between soil organic matter and both Al and Fe oxides (Cloy et al., 2014; Vogel et al., 2015; Rumpel et al., 2016). Therefore, we could not confirm the role of Al and Fe oxides as influenced by soil weathering stages over soil P composition.

Adding to that, pyrophosphate hydrolysis was found to be more rapid with greater biological activity and higher pH of agricultural soils (Sutton and Larsen, 1964), and this might have contributed to reduce pyrophosphate concentration at higher pH in mineral soils (Figure 2D). In organic layers (higher soil C levels), polyphosphate dominated the soil P_i pool (Figure 2O), and the pyrophosphate concentration decline (Figure 2N) was possibly caused by its greater lability. Pyrophosphate is less polymerized and potentially more susceptible to hydrolysis than polyphosphate. According to Savant and Racz (1972), Subbarao et al. (1977), and Dick (1985) pyrophosphate is hydrolyzed more rapidly than polyphosphate because pyrophosphate is an intermediate product of the polyphosphate hydrolysis until the final product orthophosphate.

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Adding to that, pyrophosphate hydrolysis was found to be more rapid with greater biological activity and higher pH of agricultural soils (Sutton and Larsen, 1964), and this might have contributed to reduce pyrophosphate concentration at higher pH in mineral soils (Figure 2D).

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Adding to that, pyrophosphate hydrolysis was found to be more rapid with greater biological activity and higher pH of agricultural soils (Sutton and Larsen, 1964), and this might have contributed to reduce pyrophosphate concentration at higher pH in mineral soils (Figure 2D).

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Into the P_o pool, the mechanisms that prompted the inverse response of monoesters and diesters as P limitation increased (Figure 3 AB-AD) in older, more weathered soil systems (Figure 6) are also possibly related to the development stage of those ecosystems, and its acidifying impact in those soils (Figure 9). Investigations have shown that diesters proportion, including phospholipids and DNA, increased as soil aged and monoesters concentration declined at older stages of pedogenesis (Turner et al., 2014; Vincent et al., 2013; Turner et al., 2007; McDowell et al., 2007). To plants and

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Into the P_o pool, the mechanisms that prompted the inverse response of monoesters and diesters as P limitation increased (Figure 3 AB-AD) in older, more weathered soil systems (Figure 6) are also possibly related to the development stage of those ecosystems, and its acidifying impact in those soils (Figure 9). Investigations have shown that diesters proportion, including phospholipids and DNA, increased as soil aged and monoesters concentration declined at older stages of pedogenesis (Turner et al., 2014; Vincent et al., 2013; Turner et al., 2007; McDowell et al., 2007). To plants and

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Our findings of pH influence on soil P_o functional groups (Figure 3) corroborate with Turner and Engelbrecht (2011) for tropical forest soils, where the most acidic soils contained greater proportion of the P_o as diesters (mainly DNA in that case), while neutral soils contained greater proportion as monoesters (with negligible inositol hexakisphosphate contribution in that study). It also supported Turner and Blackwell (2013) results for temperate arable soils, where greater proportion of DNA in acidic soils and greater proportion of monoesters (non-inositol hexakisphosphate for that study) in neutral soils were found. Moreover, as demonstrated for temperate pasture soils, phosphomonoesterase activity increased in acidic soil environments and phosphodiesterase is higher in neutral to basic soils (Turner and Haygarth, 2005). Therefore, this could have contributed to break down organic P molecules where its required enzymatic activity was greater.

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However, according to Turner et al. (2014) and Turner et al. (2007), inositol phosphates declined to lower concentrations in older soils, which in fine textured soils it was mirrored by a decline in amorphous Al and Fe oxides, what may have declined sorption sites for inositol phosphates and weakened protection from plant and microbial enzymatic attack. In contrast, coarse textured soils had an increase in Al and Fe oxides concentrations as soils aged, and inositol phosphates decline was attributed to changes in its inputs into soil, either from plant seeds or microbial synthesis, including through shifts in plant or microbial communities (Jangid et al. 2013, Turner et al. 2012, and Turner et al., 2014). Therefore, both soil and plant characteristics could have contributed to the inositol phosphates decline (here represented along with other monoesters) in soil ecosystems depleted with P.

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Curtin et al. (2016) found no correlation between C concentration in whole soil and clay fraction.

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However, even with the not always straightforward relationship between clay concentration and organic compounds stabilization, we verified that greater diesters concentration occurred in more weathered soils (Figure 7J), which had a greater clay concentration (Figure 9). Diesters such as DNA can be protected from dephosphorylation by adsorption into the soil matrix (Nash et al., 2014). The amino groups protonation of adenine, guanine and cytosine bases in DNA molecule can cause adsorption of charged DNA by the charged inorganic solid soil surface (Yu et al., 2013).

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However, even with the not always straightforward relationship between clay concentration and organic compounds stabilization, we verified that greater diesters concentration occurred in more weathered soils (Figure 7J), which had a greater clay concentration (Figure 9). Diesters such as DNA can be protected from dephosphorylation by adsorption into the soil matrix (Nash et al., 2014). The amino groups protonation of adenine, guanine and cytosine bases in DNA molecule can cause adsorption of charged DNA by the charged inorganic solid soil surface (Yu et al., 2013).

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In the variance partitioning of most soil P_i and P_o results, there was a smaller contribution prompted by uniquely climatic variables, or these combined with soil variables and the soil weathering stages. But the contribution of the soil weathering stage (combined or not with soil variables) was markedly high for diester-to-monoester ratio (Figures 8 and 9), and climate is a main driver of soil weathering.

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In the variance partitioning of most soil P_i and P_o results, there was a smaller contribution prompted by uniquely climatic variables, or these combined with soil variables and the soil weathering stages. But the contribution of the soil weathering stage (combined or not with soil variables) was markedly high for diester-to-monoester ratio (Figures 8 and 9), and climate is a main driver of soil weathering.

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In the variance partitioning of most soil P_i and P_o results, there was a smaller contribution prompted by uniquely climatic variables, or these combined with soil variables and the soil weathering stages. But the contribution of the soil weathering stage (combined or not with soil variables) was markedly high for diester-to-monoester ratio (Figures 8 and 9), and climate is a main driver of soil weathering.

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Finally, we expect for future researches to provide results of as much many soil P species as they can find rather than functional groups only (i.e., diesters and monoesters), even when species concentrations are low (and describe when main soil species are not detected), what which may enable future analysisanalyseis to avoid possible confounding effects of P compounds inside functional groups (e.g., inositol phosphates and monoesters), and to make a more precise correction for potential degraded peaks occurred occurring during the alkaline extraction and reading process. We also urge researchers to determine variances or standard errors for soils with distinctive properties. Then, as stated by Stewart (2010), future analysis cananalyseis couldan use the different amounts of information that provided by studies of different sizes scopes and different quality present in a meta-analyticanalytical approach.

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Models tested to explore the interdependences between edaphic and climatic variables (path analysis) as the main environmental predictors of soil inositol and DNA.

Appendix S11 – Models tested to explore the interdependences between edaphic and climatic variables (path analysis) as the main environmental predictors of of soil inorganic P compounds.

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Young, E.O., Ross, S.S., Cade-Menun, B.J., and Liu, C.W.: Phosphorus speciation in riparian soils: a phosphorus-31 nuclear magnetic resonance spectroscopy and enzyme hydrolysis study, Soil Sci. Soc. Am. J., 77, 1636–1647, doi: 10.2136/sssaj2012.0313, 2013.

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Wang, C., Wang, X., Liu, D., Wu, H., Lu, X., Fang, Y., Cheng, W., Luo, W., Jiang, P., Shi, J., Yin, H., Zhou, J., Han, X., and Bai, E.: Aridity threshold in controlling ecosystem nitrogen cycling in arid and semi-arid grasslands, Nat. Commun., 5, 4799, doi:10.1038/ncomms5799, 2014a.

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55; mineral layer, di-to-mono= 44.5 – 6.00 pH, r^2 = 0.26;

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55; mineral layer, di-to-mono= 44.5 – 6.00 pH, r^2 = 0.26;

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mineral layer, monoesters = 97.6 – 7.30 log(total C), r^2 = 0.08; organic layer, monoesters = 265.0 – 77.0 log(total C), r^2 = 0.59; mineral layer, diesters = 2.86 + 5.82 log(total C), r^2 = 0.06; organic layer, diesters = -16.0 + 74.6 log(total C), r^2 = 0.60; mineral layer, di-to-mono =0.03 + 0.073 log(total C), r^2 = 0.04; organic layer, di-to-mono = -3.71 + 1.64 log(total C), r^2 =0.53; mineral layer,

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mineral layer, monoesters = 69.0 + 6.64 log(total P), r^2 = 0.10; mineral layer, diesters

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mineral layer, monoesters = 69.0 + 6.64 log(total P), r^2 = 0.10; mineral layer, diesters

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mineral layer, monoesters = 69.0 + 6.64 log(total P), r^2 = 0.10; mineral layer, diesters

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mineral layer, monoesters = 69.0 + 6.64 log(total P), r^2 = 0.10; mineral layer, diesters

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mineral layer, monoesters = 69.0 + 6.64 log(total P), r^2 = 0.10; mineral layer, diesters

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3; mineral layer, di-to-mono = 0.47 – 0.12 log(total P), r^2 = 0.14;

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3; mineral layer, di-to-mono = 0.47 – 0.12 log(total P), r^2 = 0.14;

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mineral layer, monoesters = 90.1 –

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mineral layer, monoesters = 90.1 –

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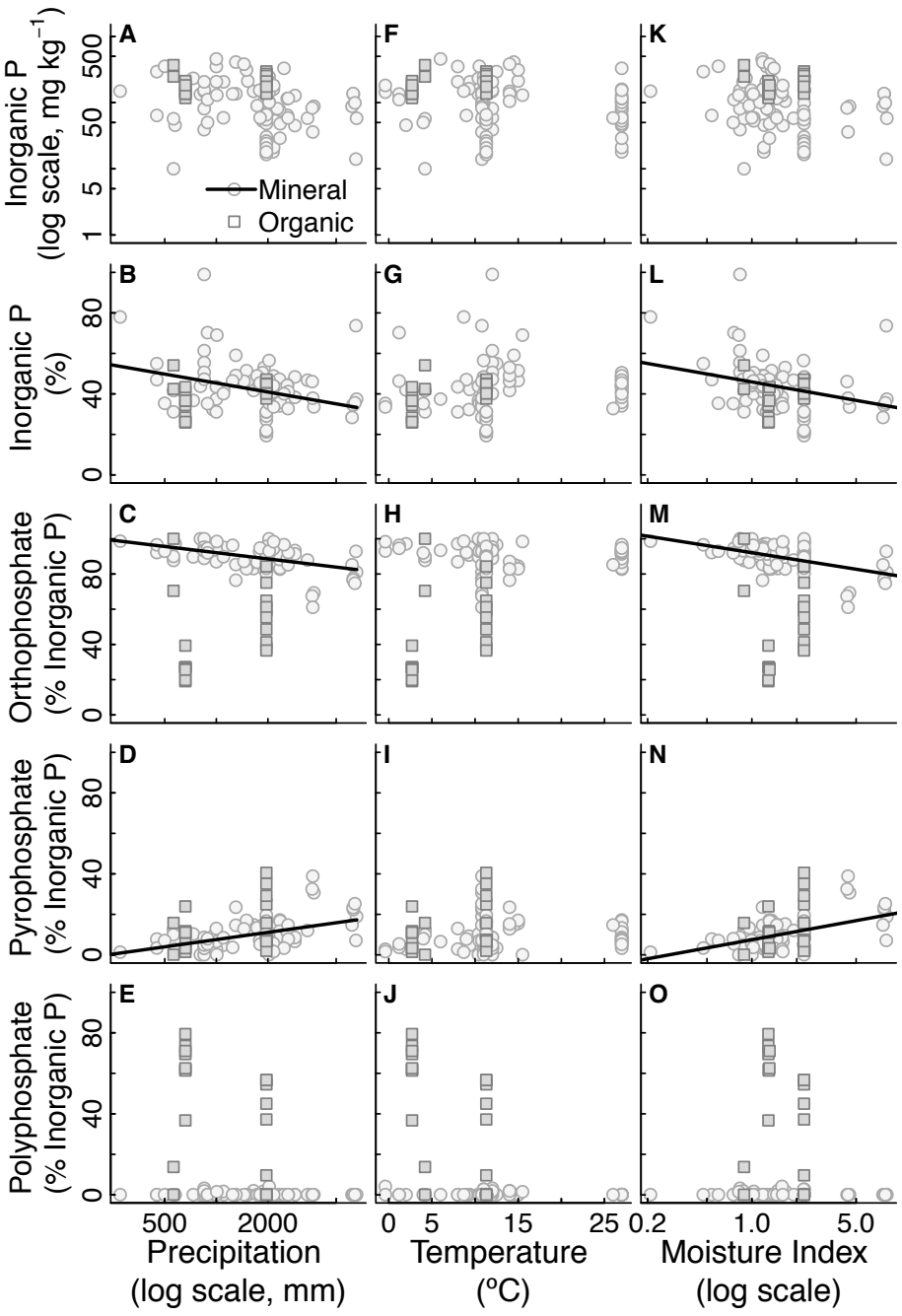


Figure 4: Relationship between climatic properties and soil inorganic phosphorus (P) composition from soil mineral and organic layers on terrestrial natural ecosystems.

Regression models (n = 80 mineral layer and n = 20 mineral layer): mineral layer, total P_i (%) = 89.4 – 14.7 log(precipitation), r^2 = 0.08; mineral layer, orthophosphate = 127 – 11.7 log(precipitation), r^2 = 0.24; mineral layer, pyrophosphate = -28.1 + 11.9 log(precipitation), r^2 = 0.24; mineral layer, total P_i (%) = 45.9 - 13.0 log(moisture), r^2 = 0.13; mineral layer, orthophosphate = 92.1 – 13.4 log(moisture), r^2 = 0.32; mineral layer, pyrophosphate = 7.49 + 13.5 log(moisture), r^2 = 0.32.

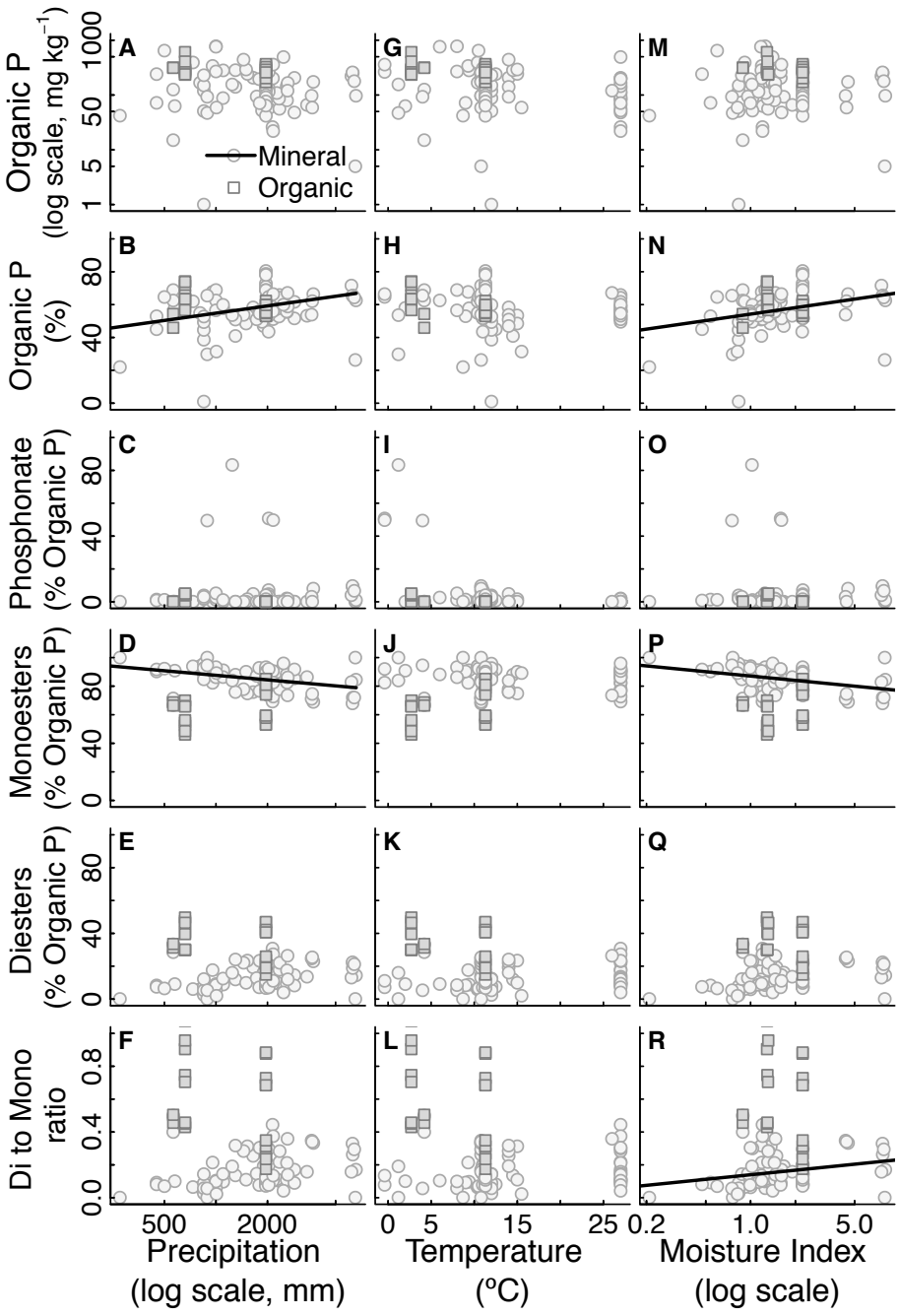
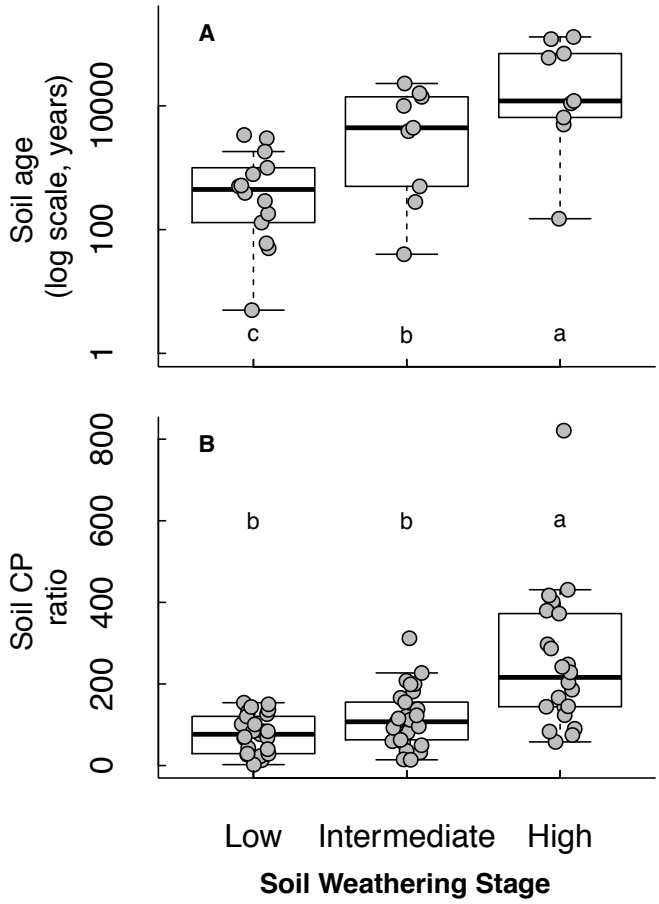


Figure 5: Relationship between climatic properties and soil organic phosphorus (P) composition from soil mineral and organic layers on terrestrial natural ecosystems. Regression models (n = 80 mineral layer and n = 20 mineral layer): mineral layer, total P_o (%) = 10.6 + 14.7 log(precipitation), r^2 = 0.08; mineral layer, monoesters = 119 – 10.6 log(precipitation), r^2 = 0.11; mineral layer, total P_o (%) = 54.2 + 13.1 log(moisture), r^2 = 013; mineral layer, monoesters = 87.1 – 10.1 log(moisture), r^2 = 0.13; mineral layer, di-to-mono = 0.14 + 0.092 log(moisture), r^2 = 0.06.



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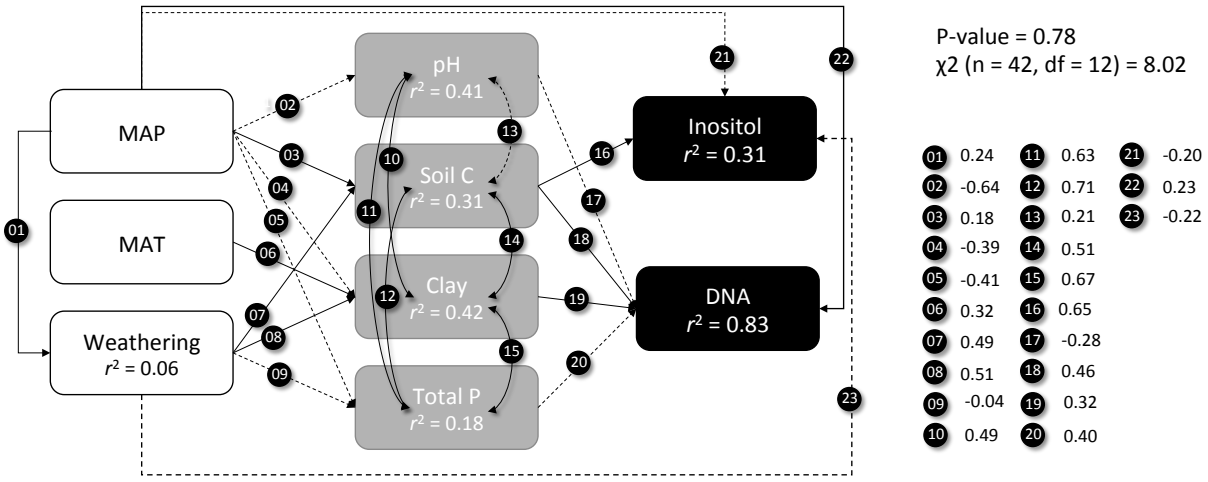

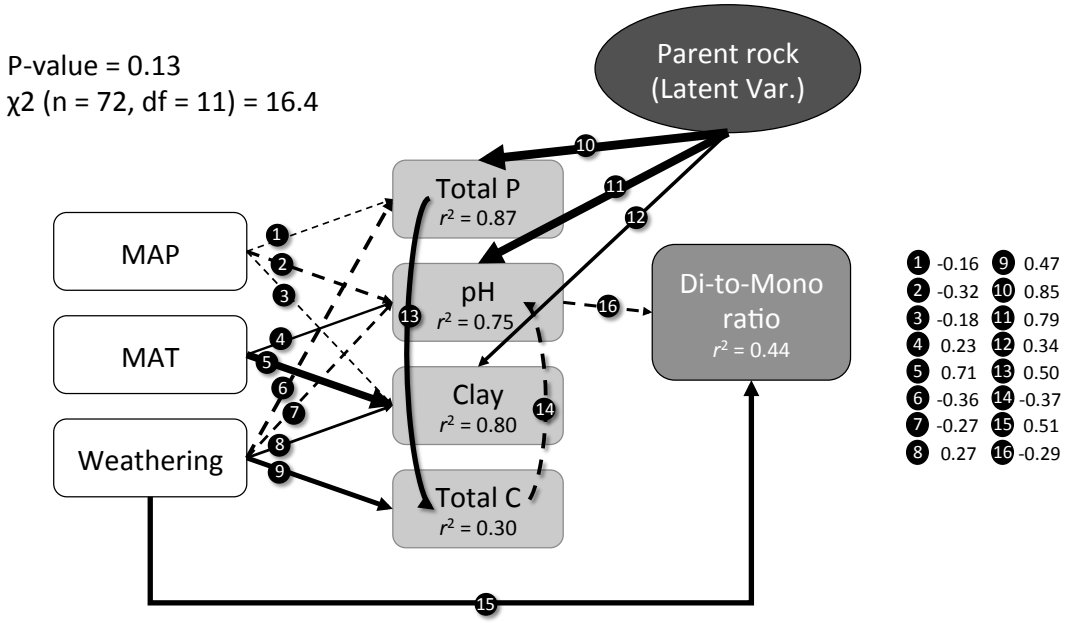

Page 32: [332] Comment [8] **Maire, Vincent** 1/9/18 10:02 PM

I've checked path analyses and I've got different results with better P-value when parent rock is included. For that, I transform the variable so that the variance of each variable is standardized and get a pvalue <

0.05 (required to meet statistical requirement of the path analysis). In addition, I let the MAT and MAP covaried. You can check the r file and the doc file with final result.

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the model.

Appendices - Soil phosphorus dynamics on terrestrial natural ecosystems

Appendix S1 - Dataset

The dataset of “Soil phosphorus dynamics on terrestrial natural ecosystems” is attached as a supplementary Excel file.

Appendix S2 – Global biomes according to the Whittaker' diagram

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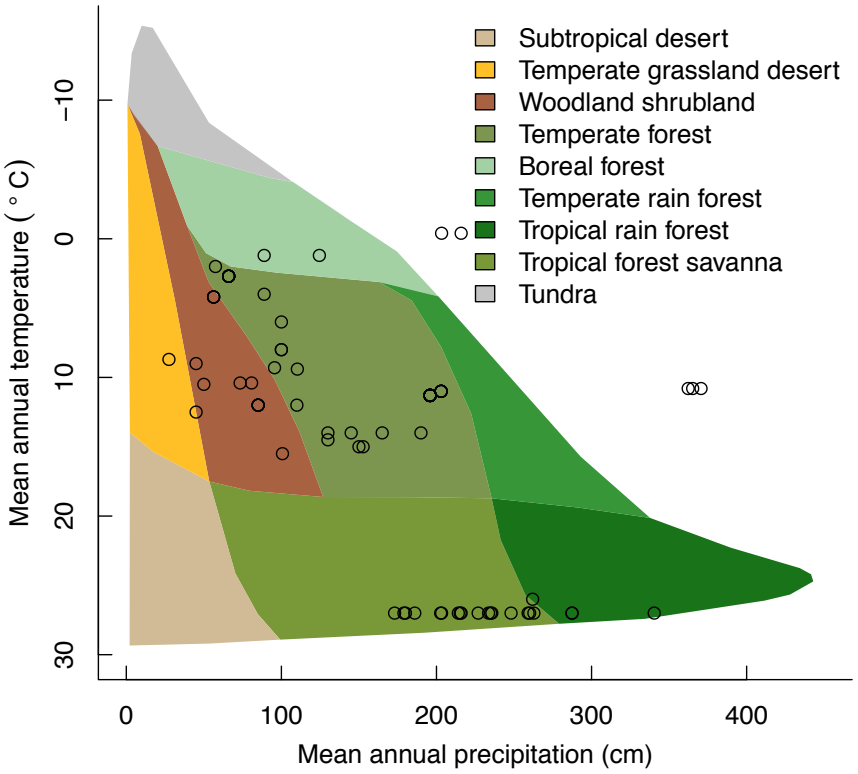

Figure S2. The Whittaker' diagram used to determine the main biomes comprised in our dataset.

Appendix S3 – Soil depth effect on soil P composition

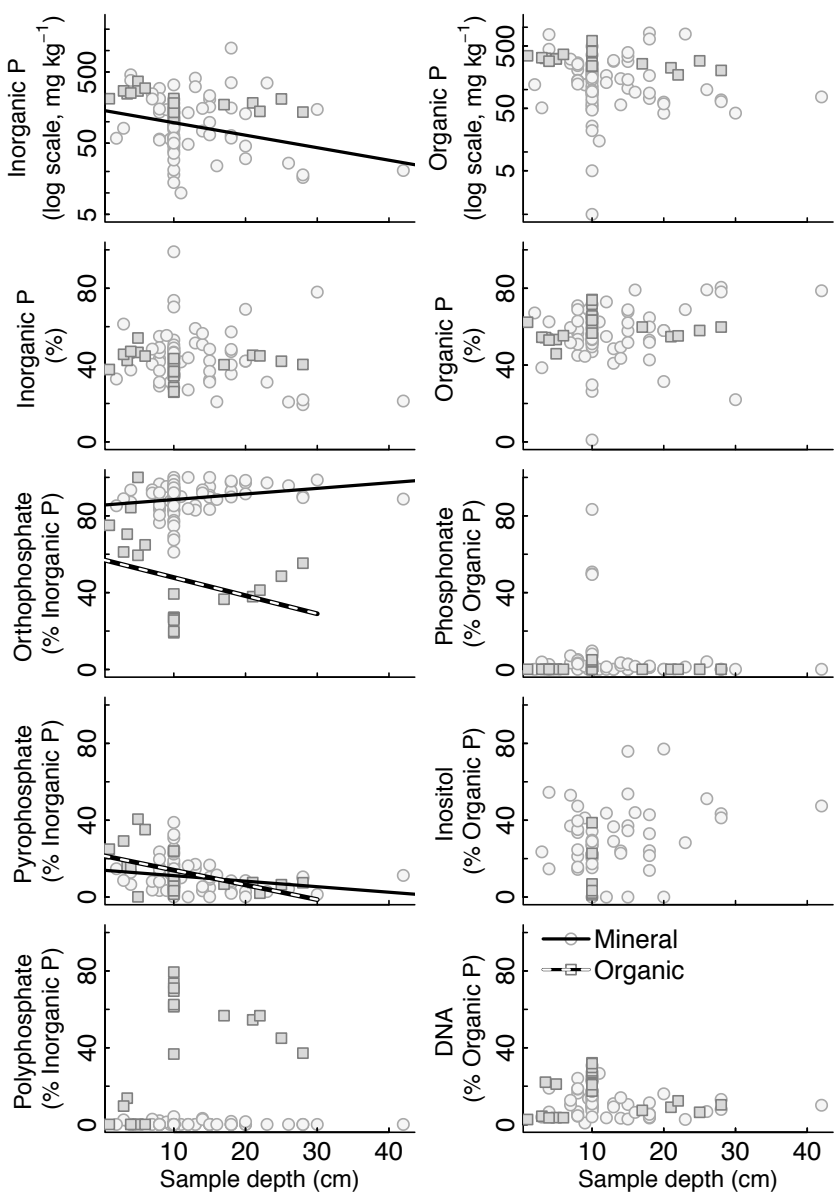

Figure S3. Soil inorganic and organic phosphorus (P) composition in NaOH EDTA extract as influenced by soil sampling depth on terrestrial natural ecosystems. Significant relationships are indicated with regression lines.

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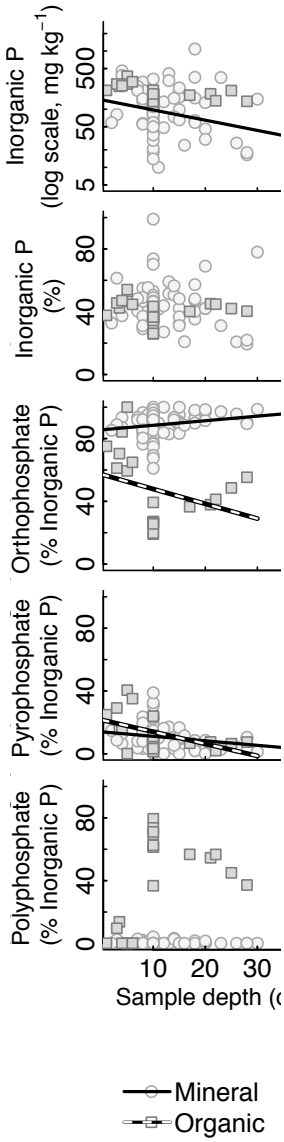

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Appendix S4 – Latitude effect on soil P composition

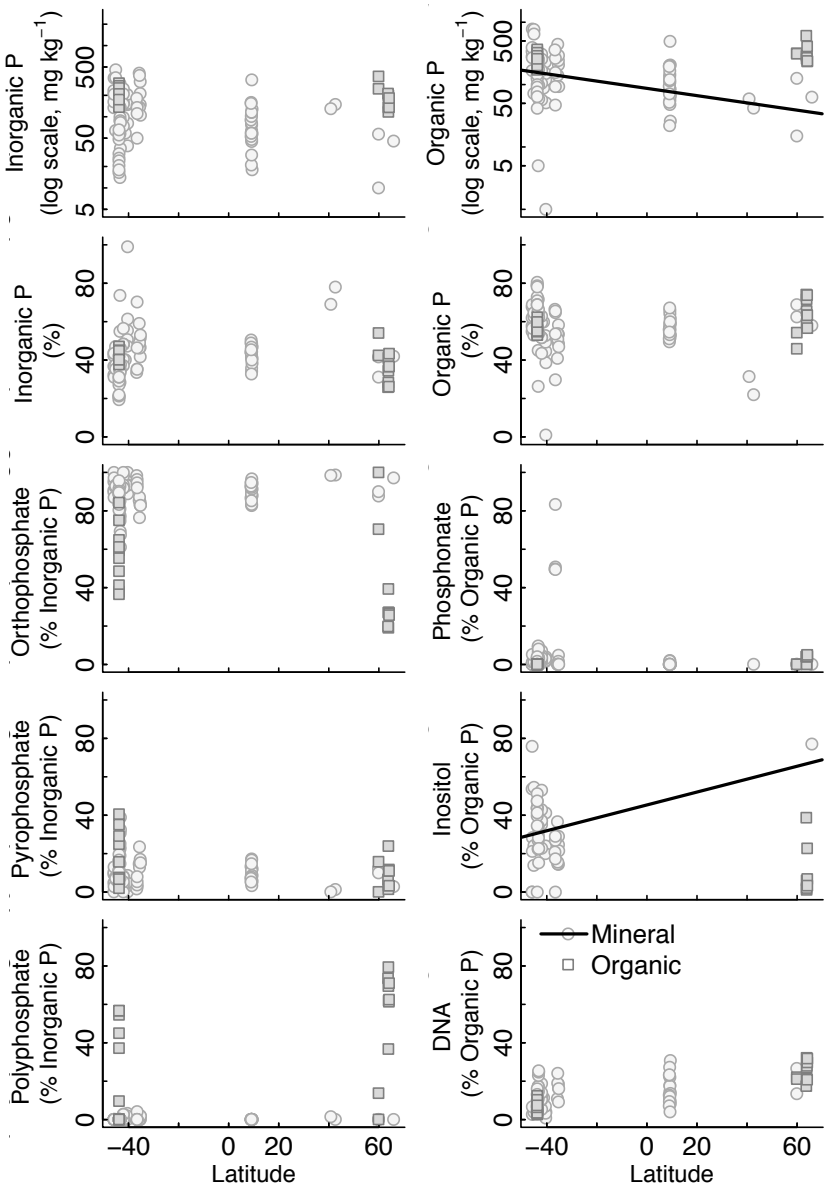

Figure S4. Soil inorganic and organic phosphorus (P) composition in NaOH EDTA extract as influenced by latitude on terrestrial natural ecosystems. Significant relationships are indicated with regression lines.

Appendix S5 – Percentage of P extracted with NaOH EDTA effect on soil P composition

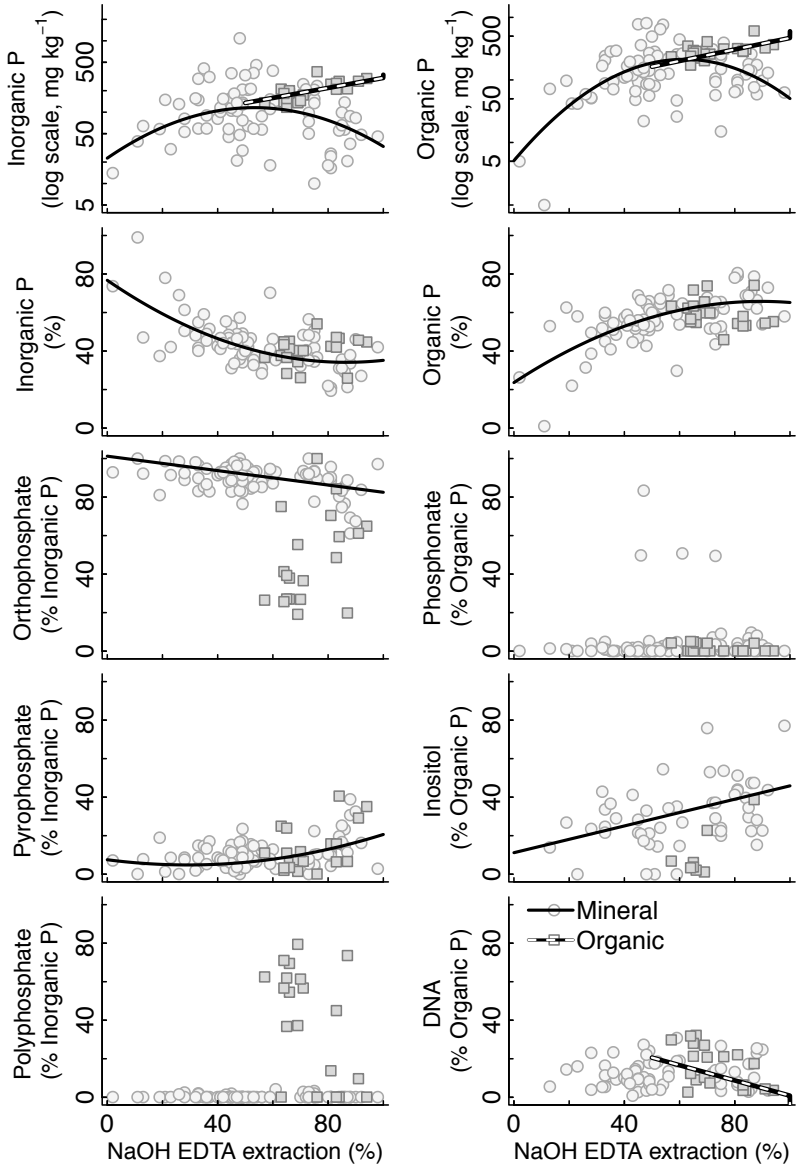

Figure S5. Soil inorganic and organic phosphorus (P) composition in NaOH EDTA extract as influenced by soil sampling depth on terrestrial natural ecosystems. Significant relationships are indicated with regression lines.

Appendix S6 – Soil properties and soil organic phosphonates

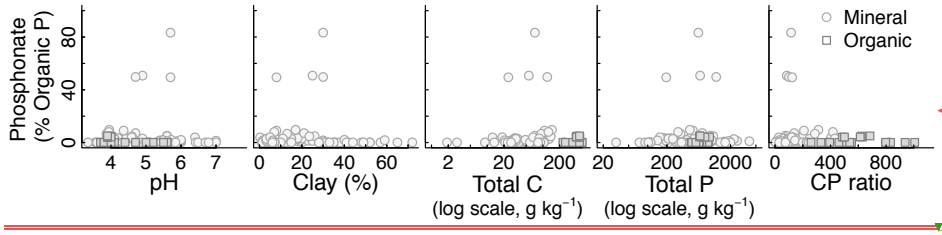

Figure S6. Relationship between edaphic properties and soil organic phosphonates in NaOH EDTA extract from soil mineral and organic layers on terrestrial natural ecosystems. No significant relationships were found.

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Appendix S7 – Climatic properties and soil inorganic phosphorus

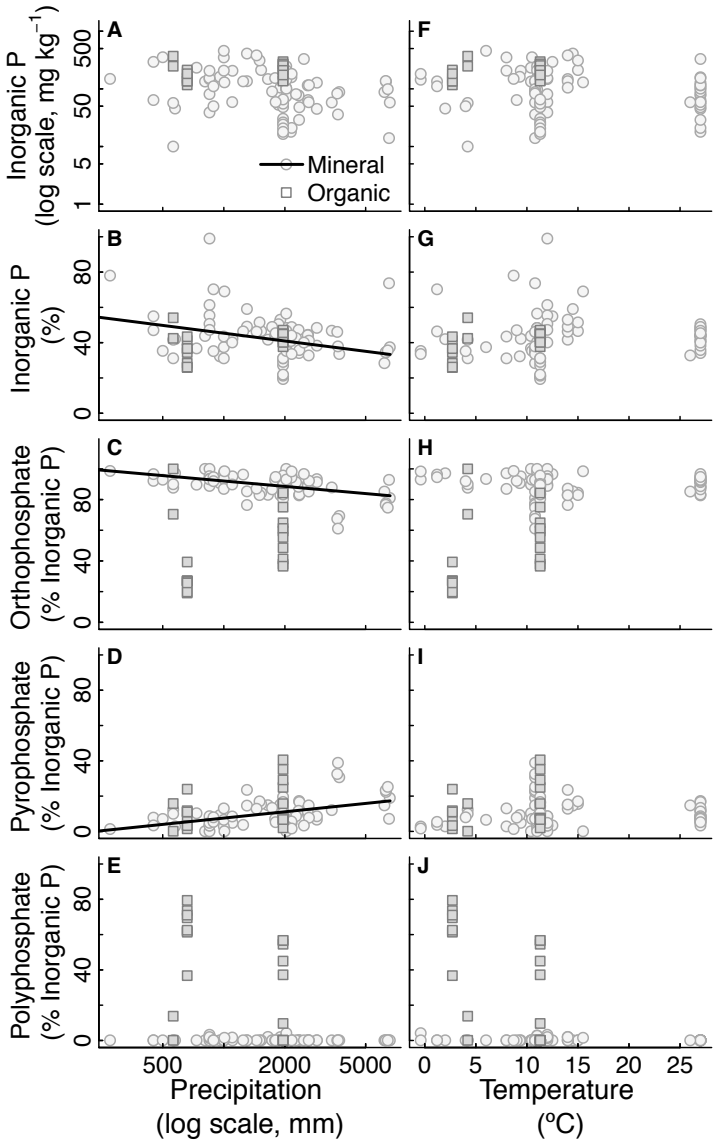

Figure S7. Relationship between climatic properties and soil inorganic phosphorus (P) composition in NaOH EDTA extract from soil mineral and organic layers on terrestrial natural ecosystems. Regression models (n = 80 mineral layer and n = 20 mineral layer): mineral layer, total P_i (%) = 89.4 – 14.7 log(precipitation), r^2 =

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0.08; mineral layer, orthophosphate = 127 – 11.7 log(precipitation), r^2 = 0.24; mineral layer, pyrophosphate = -28.1 + 11.9 log(precipitation), r^2 = 0.24.

Appendix S8 – Climatic properties and soil organic phosphorus

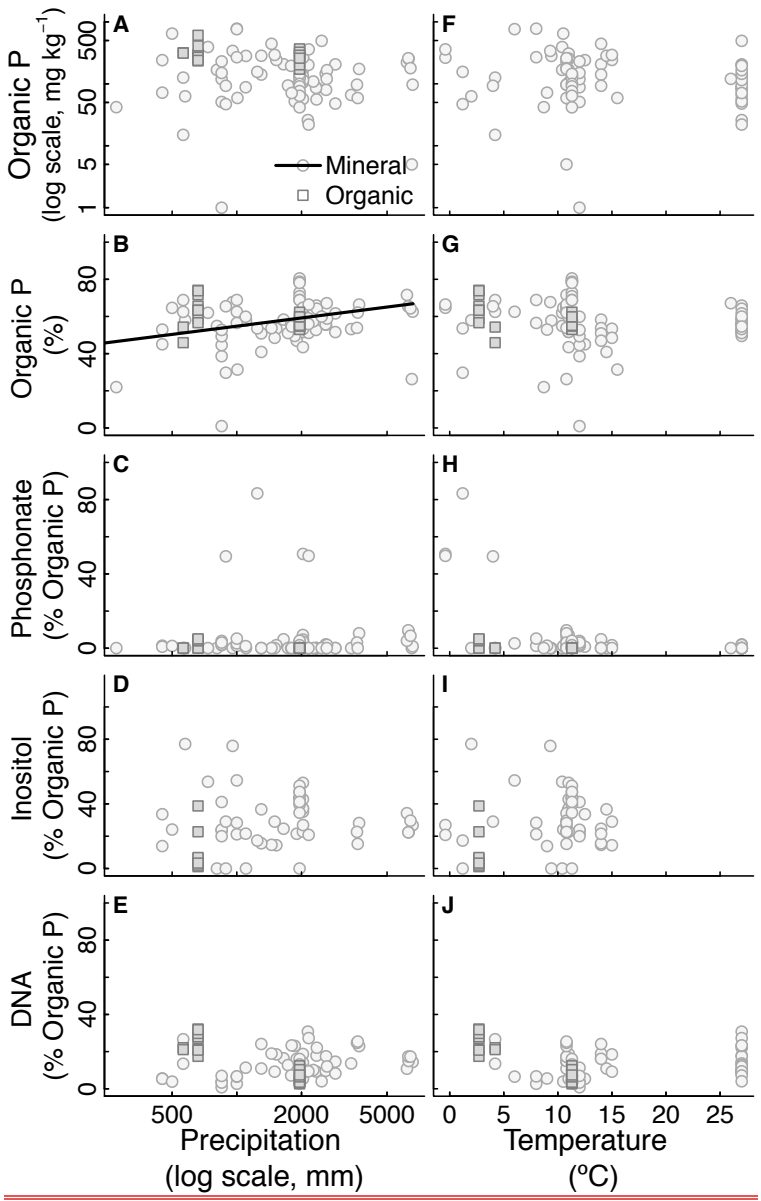

Figure S8. Relationship between climatic properties and soil organic phosphorus (P) composition in NaOH EDTA extract from soil mineral and organic layers on

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terrestrial natural ecosystems. Regression models: mineral layer, total P_o (%) = 10.6 + 14.7 log(precipitation), r^2 = 0.08 (n=80).

Appendix S9 – Soil weathering stages and poorly crystalline Al and Fe concentration

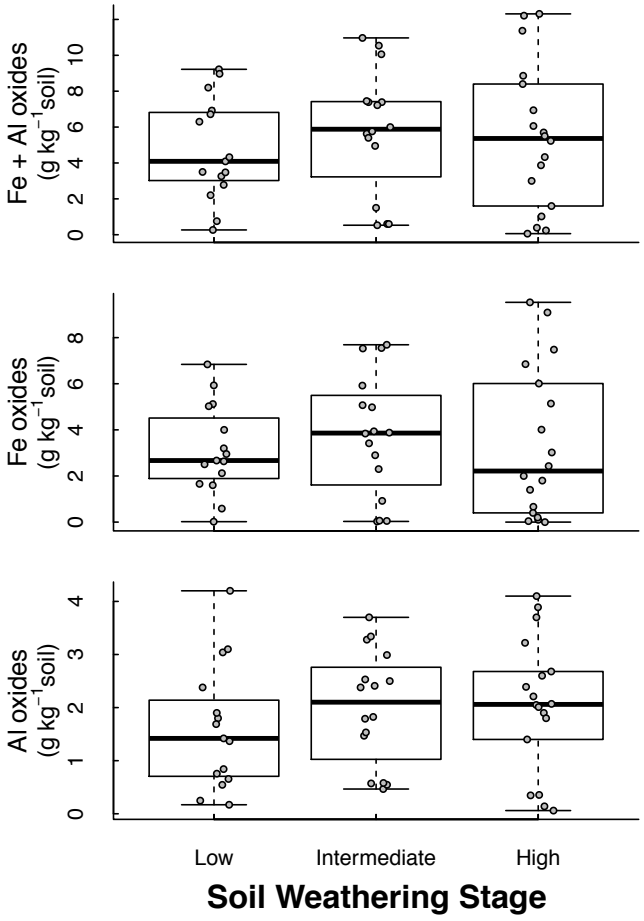

Figure S9. Soil weathering stage relationship with soil poorly crystalline Al and Fe (n = 49) on terrestrial natural ecosystems. For all three panels p>0.1.

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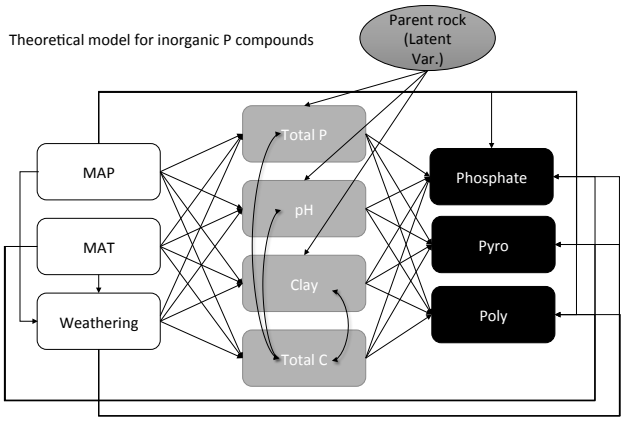
Appendix S10 – Models tested to explore the interdependences between edaphic and climatic variables (path analysis) as the main environmental predictors of soil inorganic and organic P compounds.

We expected some directionalities in the relationships, based on the literature (theoretical models on Figures S10.1 and S10.2, top panels).



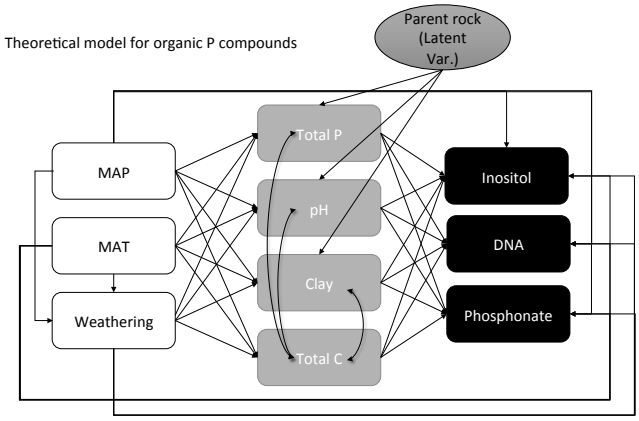


Figure S10.1. Theoretical models set to explore the interdependences between edaphic and climatic variables (path analysis) as the main environmental

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predictors of soil inorganic (upper panel) and organic (bottom panel) P compounds.

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Final model 1

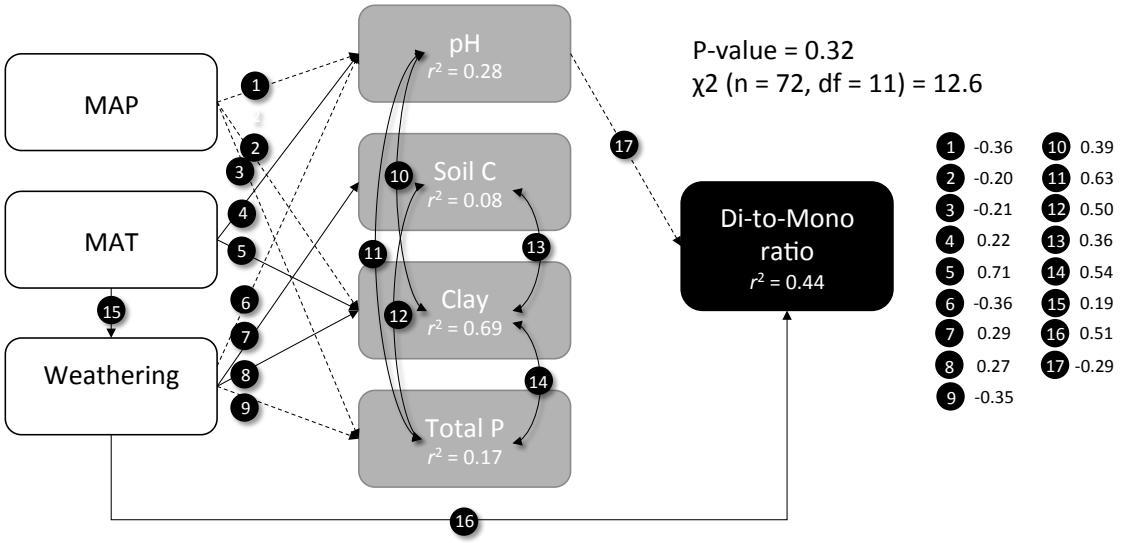

Figure S4.2 Final model 1 set to explore

climatic variables (path analysis).

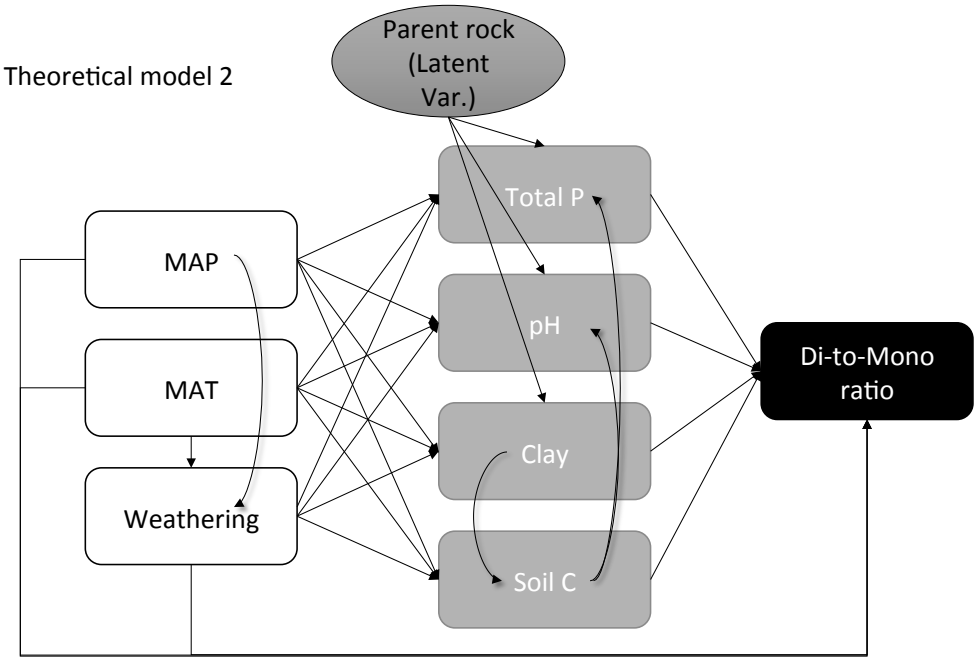

Figure S4.3 Theoretical model 2 set to explore the interdependences between edaphic and climatic variables (path analysis).

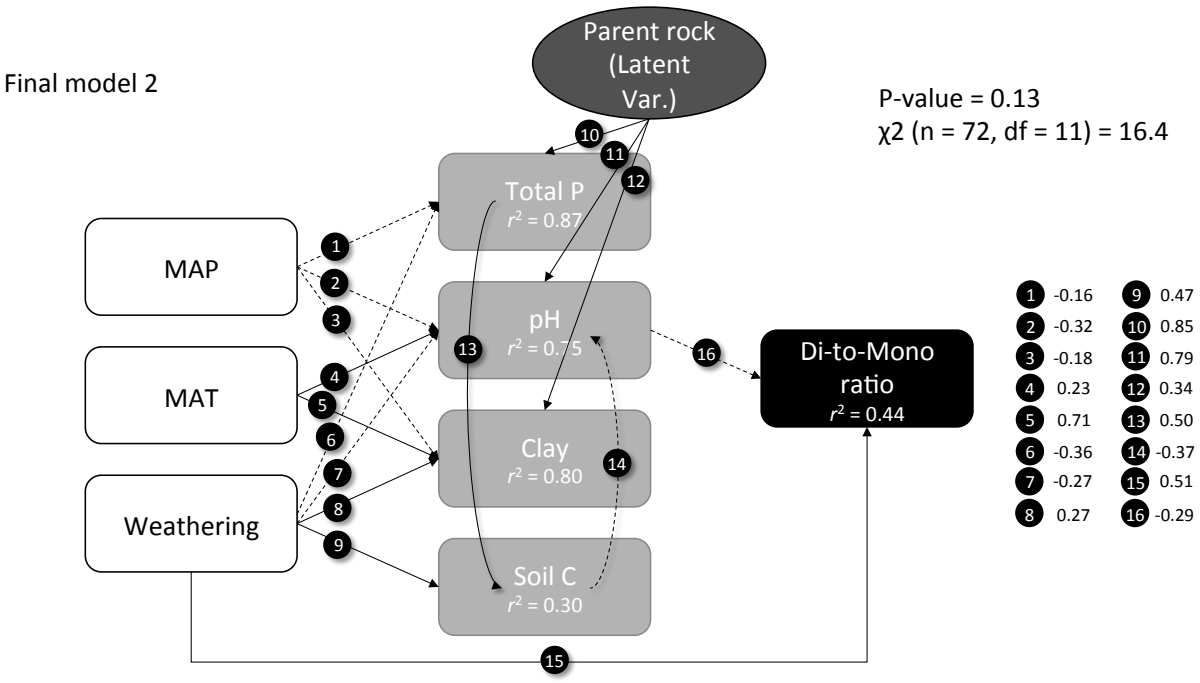

Figure S4.4 Final model 2 set to explore the interdependences between edaphic and climatic variables (path analysis).