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Dear Associate Editor Sönke Zaehle,

Thank you for your evaluation of the manuscript. We also thank the reviewers for the great effort they made in reviewing the manuscript, and for the significant improvements they promoted on it. We are sincerely grateful for the comprehensive review of our manuscript, and we valued each comment made. We added acknowledgments to reviewers recognizing their effort.

We responded to each comment and revised the manuscript. We made appropriate changes in the manuscript or have provided an explanation where we could not follow exactly what reviewers suggested. To the best of our understanding, no remaining issues are left in this corrected version.

The major changes made on the manuscript are briefly described in the following. A new title was introduced including a more specific description of the study. In the introduction, we added the rationale about the limitations of the 31P NMR method. For the Material and Methods, a complementation was made on how the structural equation modeling was developed. For the results, we added the average and range of P extracted by NaOH-EDTA (%) for both mineral and organic layers. Moreover, most changes were made on the discussion. These included adding discussion and several references on the effect of acidifying soils, which may increase the charge of some organic compounds, and thus increase sorption. Discussion on why inositol hexakisphosphates proportions increase as soils get more weathered in non-tropical environments, but no inositol hexakisphosphates have been found in very weathered soils of tropical environments was added. We also added discussion on how vegetation change during pedogenesis and how it affects soil P composition. Ultimately, we discussed future research priorities such as disentangling confounding effects among soil biotic and abiotic components, climate and vegetation; and coupling ^{31}P NMR with other important techniques to better understand P cycling and composition in terrestrial ecosystems. We excluded the terms "dynamics" and "terrestrial" throughout the manuscript. Finally, a native person with an affinity to the topic reviewed the whole manuscript again for precision and clarity of terminology.

Responses to the reviewers' comments are described below under "Response". Changes made on the manuscript were all based on these responses. To facilitate the evaluation, we left the reviewers comments before each response.

Responses made by the authors are in bold and indicated with the term "Response". After the responses to the reviewers comments one can find the manuscript with all changes marked.

REPORT 1

For the most part, the manuscript has been significantly improved compared to the original submission. In particular, the authors have constrained the studies used to avoid methodological differences, and have focused on specific P compounds to minimize differences among research groups (e.g. with respect to correcting for degradation), which could hamper the type of global comparisons they are trying to make. I still think the studies they have included are too heavily skewed to studies by a single author (Turner) and a single location (New Zealand). However, that is partially due to issues with analyses by some studies by other groups or from other locations. Hopefully, future studies will avoid these problems, allowing this type of analysis to be expanded in time. But for now, this is an interesting first attempt at this type of analysis.

There are still some problems that will need to be addressed before this is publishable. The majority of these fall more into minor or moderate revisions, as indicated below. However, the discussion will need some substantial revision, because many parts of it, as indicated below, are based merely on speculation, and are not grounded in fact or even in the authors' own results.

1. Writing quality: for the most part, the quality of English has improved. However, there are still some problems with some sections. These are specifically addressed below, but I still recommend that the authors have the revised version read by a native English speaker who is familiar with the research topic. There are still problems with the writing in that the authors make sweeping statements of fact, without citing any references to support these statements. This is particularly true in the introduction, and specific instances are noted below.

2. Title: The current title isn't very informative or very well written (e.g. "soil" by definition is "terrestrial", so including both is redundant). I suggest: "A meta-analysis of ecosystem properties influencing soil phosphorus dynamics in natural ecosystems".

Response: The "meta-analysis" term is being avoided because no meta-analytic methods were used to statistically analyze the data. These statistical methods were not used due to the lack of error measurements on the compiled data (e.g., variance or standard error). The title was changed to a more informative version, as suggested by the reviewer: "Environmental drivers of soil phosphorus composition in natural ecosystems". We used "compounds" instead of "dynamics" as suggested by the Reviewer #2.

3. Abstract: Although the main body of the paper appears to have been revised with the assistance of a native English speaker, the abstract appears to have been overlooked.

Line 7: “phosphorus (P) compounds can be modified by distinctive ecosystem properties” Using the term “modifies” implies that the ecosystem properties directly change P forms. However, most of the changes are indirect. For example, pH can change the soil environment to influence sorption of P forms, or it can affect the organisms that produce P forms, or that produce enzymes to mineralize P forms. But pH itself isn’t necessarily changing P forms, except potentially alter the charge of the P form. As such, I think it would be best to change this to “phosphorus (P) compounds can be influenced by distinctive ecosystem properties”.

Response: OK. “influenced by” was used instead of “modifies”.

Line 8: “soil P dynamics on terrestrial natural ecosystems, relating its organic “should be “soil P dynamics in natural ecosystems, relating organic “

Response: Changes were made.

Line 11: “determined the soil P composition” should be “determined soil P composition”

Response: Changes were made.

Lines 11-12: “nuclear magnetic resonance of soils extracted with NaOH EDTA” should be “nuclear magnetic resonance of NaOH-EDTA extracts of soils”

Response: Changes were made.

Line 12: “models were used to better understand the soil P” should be “models were used to better understand the factors influencing soil P”

Response: Changes were made. We used “environmental properties” instead of “factors”.

Line 13: “relationships, soil P compounds had similar overall behaviors on mineral and organic layers but with different slopes”. This analysis does not give any information about the behavior of soil P compounds; instead, it shows the relationship of soil P compounds with various factors. As such, this should be “relationships, trends for soil P compounds were similar for mineral and organic layers but with different slopes”.

Response: OK.

Line 16: “Soil, particularly” should be “Soil factors, particularly”; “ratio” should be “ratios”

Response: Changes were made. We used “properties” instead of “factors”.

Line 18: “soil P composition on terrestrial natural ecosystems” should be “soil P composition in natural ecosystems”

Response: Changes were made.

Lines 19-20: “and after altogether with plant and microbe coexistence” I do not understand what the authors are trying to say here; something seems to be missing.

Response: It was changed to “and after altogether through plant and microbe P cycling”.

4. Introduction

p. 2, line 4: “derives essentially” should be “derives predominantly”; “as an eolian deposit” should be “as eolian deposits”. However, I disagree with this statement, because it is only relevant for younger soils (e.g. Chadwick et al. 1999 Nature 397:491-497). And because this is a statement of fact, a reference needs to be cited here.

Response: We added the term “young” before “ecosystem” to fulfill the statement according to the Chadwick et al. (1999) findings and the reference was added: “Phosphorus input into a young ecosystem derives predominantly from the weathering of rocks, with little input as eolian deposits”.

p. 2, lines 5-6: “Phosphorus goes back to soil as organic materials (Noack et al. 2012” but not necessarily as organic P; Noack et al. showed that much of the P in plant residues was orthophosphate. I think the authors need to explicitly state that here, that “organic materials” are not primarily composed of organic P forms.

Response: The sentence was changed to “Phosphorus goes back to soil as organic materials, which are composed by organic and inorganic P compounds”.

p. 2, lines 7-9: “Each new cycle...primary minerals” Please cite a reference to support this statement of fact, because I am not sure that it is in fact true.

Response: McDowell et al. (2007) was added to support the following sentence “Each new cycle loop leads to more complex and less bioavailable P compounds”, and “Walker and Syers (1976) was used to “ultimately seriously limiting ecosystem productivity in the absence of ‘fresh’ P input as primary minerals.”

p. 2, line 10: Jenny (1941) described these as the five factors of soil formation, not ecosystem functioning. Please cite another reference to support using this term in this way.

Response: “soil formation” was used instead of “ecosystem functioning” in this sentence and across the manuscript. Therefore, Jenny (1941) was maintained for this statement.

p. 2, line 14:”main Po compounds are” should be “main Po compound categories are”, because monoesters, diesters, etc. are compound categories, not forms.

Response: Changes were made.

p. 2, lines 15-16: “orthophosphate monoester” should be “orthophosphate monoesters”, “orthophosphate diester” should be “orthophosphate diesters” and “phosphonate” should be “phosphonates”, because these broad compound classes contain a number of different P compounds.

Response: Changes were made across the manuscript.

p. 2, line 18: “used by” should be “directly taken up by”, because all P compounds can be used by plants and microbes after transformation, just not directly taken up by them.

Response: Changes were made.

p. 2, lines 17-19: “Specific phosphatase...and microbes” and “Obviously...optimum” Please cite references to support these statements of fact.

Response: Changes were made. Turner (2008a) was used for “Specific phosphatase...and microbes” and Frankenberger and Johanson (1982) was used for “Obviously...optimum”.

p. 2, lines 19-20: “Phosphomonoesterase is more active in acidic soils while phosphodiesterase is optimized in basic soils (Turner and Haygarth 2005)” NO! It simply is not possible to make such a broad, sweeping statement based on a single study of pasture soils from England. Please rewrite.

Response: The sentence was excluded to avoid the broadening of the statement, and also because the rationale was supported by the previous sentence.

p. 2, lines 28-29: “As soil ages...colimitation intermediate stage” Please cite a reference to support this statement of fact.

Response: Walker and Syers 1976 and Turner and Condron (2013) were used to supporting the statement.

p. 3, line 4: “precipitations” should be “precipitation”.

Response: Change was made.

p. 3, lines 9-14: I think “parent rock” should be changed to “parent material”, because soils can form on materials other than a specific type of rock (e.g. sand, glacial till).

Response: “parent material” was used instead of “parent rock” in all instances across the manuscript.

p. 3, line 16: “Soil P composition has been studied in soils from ecosystems worldwide” should be “Soil P composition has been studied in ecosystems worldwide”

Response: Changes were made.

p. 3, line 17: “(NMR) was a widely used method” should be (NMR) is a widely-used method”, because to the best of my knowledge it is still being used.

Response: Changes were made.

p. 3, line 19: “NaOH and chelating agent EDTA” should be “NaOH combined with the chelating agent EDTA”

Response: Changes were made.

p. 3, line 20 (and elsewhere throughout the manuscript): “NaOH EDTA” should be “NaOH-EDTA”.

Response: It was changed throughout the manuscript.

p. 3, lines 22-24: “The NaOH EDTA...(Turner and Blackwell 2013)” This sentence does not make any sense as written. How does this demonstrate that NaOH-EDTA is quantitative? Please rewrite and include more references.

Response: OK. The sentence was changed to “The NaOH-EDTA extraction method is recognized to quantitatively extract P compounds from the soil.”

p. 3, lines 26-27: “state factors of ecosystem functioning” see previous comment and rewrite.

Response: Changes were made. “soil formation” was used instead of “ecosystem functioning”.

p. 3, line 27: “because known responses were obtained from case-specific conditions”. I do not understand what the authors are trying to say here. Please rewrite.

Response: the sentence was rewritten to “how soil P composition is affected by different state factors of soil formation because most know responses were obtained from specific chronosequences.”

5. Methods

p. 4, line 5: “that namely estimated an adequate delay time” should be “that used an adequate time”

Response: Changes were made.

p. 4, line 15: “only control (unchanged) was used” should be “only control (unchanged) samples were used”

Response: Changes were made.

p. 4, lines 23-23: “organic P and its compound” should be “organic P and the compounds”

Response: Changes were made.

p. 4, line 24: “NaOH EDTA inorganic P, and its compounds” should be “and NaOH-EDTA inorganic P and the compounds”

Response: Changes were made.

p. 4, line 27: “climate characteristic” should be “climate characteristics”, because it refers to both precipitation and temperature.

Response: Changes were made.

p. 5, lines 29-30: “how the inorganic and organic P compounds variation” should be “how variations in inorganicand organic P compounds”

Response: Changes were made.

p. 6, line 4: “Differently” should be “In contrast”

Response: Changes were made.

6. Results:

p. 6, line 13: “McDowell and Steward” should be “McDowell and Stewart”; the name is spelled correctly in the References section.

Response: Changes were made.

p. 6, line 20: “And temperate rain forest” Don’t some of the New Zealand sites fall into temperate rain forest? The authors should check to confirm.

Response: According to Whittaker's diagram we found that some of those plots were classified as intermediates between the temperate rainforest and

boreal forest, and tropical rainforest and temperate rainforest. It is being described at the beginning of the third paragraph of the discussion section.

p. 6, line 25: “were absented” should be “were absent”

Response: Changes were made.

p. 6, lines 26-27: “All compiled results...polyphosphate” This sentence repeats information already given (p. 4, lines 22-24), and should be deleted in one of these locations in the text.

Response: OK. The given sentence was deleted.

p. 7, line 3: “The compounds proportions” should be “The proportioning of compounds”

Response: Changes were made.

p. 7, line 6: “Into the Pi pool” should be “In the Pi pool”; “layers, the orthophosphate” should be “layers, orthophosphate”

Response: OK. Both were corrected.

p. 7, lines 12-13: “There was no clay effect on both Pi and Po and their compounds proportions” should be “Clay had no effect on either Pi or Po, or on the proportions of compounds in these categories”

Response: Changes were made.

p. 7, line 18: “effect on both Pi and Po” should be “effect on either Pi or Po”

Response: Changes were made.

p. 7, line 24: “in both mineral and organic “ should be “in either mineral or organic”

Response: Changes were made.

p. 7, lines 30-31: “In is important to note...usually comprises the residual P not recovered by the NaOH EDTA extractant”. This sentence doesn’t make sense to me. Firstly, the x axes in figures 2 and 3 vary depending on the factor (e.g. %, % organic P, log scale mg/kg). Secondly, total P doesn’t just include residual P (which is what “comprises the residual P” means”), it includes both the extracted P and the residual P. This sentence needs to be rewritten.

Response: The sentence was rewritten as “It is important to note that the reported total P (x-axis on Figures 2 and 3) is the one obtained by digestion and it includes both the extracted P and the residual P.”

p. 8, line 2: “As the percentage” should be “As a percentage”

Response: Changes were made.

p. 8, line 12: “on the soil P composition on terrestrial natural ecosystems” should be “ in the soil P composition in natural ecosystems”

Response: Changes were made.

p. 8, line 13: “over total “ should be “on total”

Response: Changes were made.

p. 8, line 18 and line 19: “affected the soil age and CP ratio” should be “affected soil age and CP ratios”

Response: Changes were made.

p. 8, line 29: “of the ecosystem’s properties” should be “of ecosystem properties”

Response: Changes were made.

p. 8, line 31: “and its compound” should be “and its compounds”

Response: Changes were made.

p. 9, line 2: “poorly defined” should be “poorly-defined”

Response: Changes were made.

p. 9, line 3: “of the ecosystem’s properties” should be “of ecosystem properties”

Response: Changes were made.

p. 9, lines 7-8: “phosphonate had most of its variation explained by” should be “most of the variation in phosphonates was explained by”

Response: Changes were made.

p. 9, line 19: “by precipitation into the “ should be “by precipitation in the”

Response: Changes were made.

p. 9, line 21: “Into the” should be “in the”

Response: Changes were made.

p. 9, line 26: “Pyrophosphate had a positive influence of” should be “Pyrophosphate was positively influenced by”

Response: Changes were made.

p. 9, line 27: “inositol was negatively” should be “inositol phosphates were negatively”

Response: Changes were made.

p. 9, line 29: “Phosphonate was” should be “Phosphonates were”

Response: Changes were made.

7. Discussion: Much of this is very poorly done, and relies on speculation rather than data. Major revisions are needed in this part of the manuscript. p. 10, line 2: “compounds respond to” should be “compounds responded to”

Response: Changes were made.

p. 10, line 3: “at a wide” should be “on a wide”

Response: Changes were made.

p. 10, line 4: “terrestrial nature ecosystems” should be “natural ecosystems”

Response: Changes were made.

p. 10, line 7: “persistence on ecosystems” should be “persistence in ecosystems”

Response: Changes were made.

p. 10, lines 7-11. This is a very long, confusing sentence; as written, the point that the authors are trying to make with this sentence is not clear. It should be rewritten.

Response: The phrase has been rewritten in order to clarify it.

p. 10, line 14: “the decaying degree of C element is lower than the P” I do not understand what the authors are trying to say here.

Response: OK. The sentences were changed to “As time passes after the onset of pedogenesis, the ecosystem accumulates organic matter up to a maximum, and then starts to decline. Along with this decline, there are also changes in the chemical composition of organic matter, in which the decaying degree of C element is lower than the P, and concomitantly there is an increasingly acidic environment (Walker 1965; Turner and Condron 2013).”

p. 10, line 15: “resulting in the slowed decomposition of the older soil system” Again, I do not understand what the authors are trying to say here. As currently written, it indicates that the soil system itself is decomposing more slowly. However, soils do not decompose. Do they mean that decomposition is slower in older soil systems? If so, then decomposition of what? Plant material? Or do they mean mineralization of specific P forms? This needs to be rewritten for clarity.

Response: The sentence “resulting in the slowed decomposition of the older soil system” was excluded because it was not being relevant to the understanding of the manuscript. What we meant was that the organic matter decomposition is slower in older soil systems due to a higher CP ratio and a more acidic soil environment.

p. 10, lines 16-19: “more weathered soils are remote from the parent material”. Again, this is confusing as written. Do the authors mean physically remote? If so, wouldn’t that depend on the nature of the soil weathering processes, and even on the soils themselves? Or do they mean “substantially changed”? Please rewrite.

Response: We meant physically remote, but we understood and agreed with the point the reviewer is making. To fulfill that we changed “remote” to “substantially changed”.

p. 10, line 25: “occluded P increases at the expense of organic P”. This sentence implies that organic P compounds cannot be occluded, which isn’t the case – one of the reasons that inositol phosphates increase with weathering is that they become occluded in the same way that inorganic P compounds become occluded. The inaccuracy of this statement is due to citing a dated reference (Crews et al. 1995) that relied on Hedley fractionation. Please update this statement and cite something that uses P-NMR or other modern techniques.

Response: The sentence was modified to “In highly weathered soils, occluded P increases through the encapsulation of the organic and inorganic P compounds inside of Fe and Al minerals (McDowell et al., 2007; Turner et al. 2007).”

p. 10, line 31 and p. 11, line 2: “phosphorus” should be abbreviated as P.

Response: Changes were made.

p. 11, line 2: “with species maximum” should be “with species’ maximum”

Response: Changes were made.

p. 11, line 5: “shapes” should be “shaped”, because it is discussing a student that has been published.

Response: Changes were made.

p. 11, line 9: “As soil aged” should be “As soils aged”

Response: Changes were made.

p. 11, line 11: “weathering stages had a major role” should be “weathering stage had a major influence”

Response: Changes were made.

p. 11, line 11: “dynamic” should be “dynamics”

Response: Changes were made.

p. 11, lines 16-23: It is well-established that ectomycorrhizal fungi convert the orthophosphate that they take up from the soil into polyphosphates, and translocate the polyphosphate along fungal hyphae, sometimes a great distance from where the orthophosphate is taken up (e.g. Bücking and Heyser 1999 Mycol. Res. 103:31-39; Plassard and Dell 2010 Tree Physiol. 30:1129-1139). This needs to be mentioned, as well as the need to define plant and microbial communities in studies of P forms.

Response: The information about the ectomycorrhizal fungi was added and the need to define plant and microbial communities in studies of P forms was emphasized.

p. 11, line 28 to p.12, line 2: Polyphosphates can potentially degrade to pyrophosphates during extraction and analysis by P-NMR (e.g. Cade-Menun et al. 2006, Environ. Sci. Technol. 40:7874-7880), so pyrophosphate and polyphosphate shouldn’t be considered as fully distinct P forms, and the authors should use care when discussing hydrolysis of pyrophosphate separately from polyphosphate.

Response: Changes were made.

p. 12, lines 5-6: “Plant and microorganism breakdown diesters need” should be “Plant and microorganism breakdown of diesters needs”

Response: Changes were made.

p. 12, lines 8-9: “P limitation increased phosphoesterases synthesis as a way to increase the organic P breakdown to the bioavailable P”. As written, it doesn’t make sense. Do the authors want to say: “P limitation may stimulate increased phosphoesterase synthesis as a way to increase bioavailable P by the mineralization of organic P”?

Response: The sentence was changed to the proposed one.

p. 12, lines 12-16: “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”. These two sentences are baseless speculation, and should be deleted from the paper. The authors did not include any measures of phosphatase activities of any kind from any of the published studies, and thus are unqualified to say anything about how enzyme activity changes with soil factors. Their results in Fig. 3 show that the proportion of total organic P, as well as the proportions of IHP and DNA decrease with increasing pH. This is related to increased charge of these compounds with decreasing pH, and thus increased sorption. This is discussed in detail in Condron et al. 2005, among other papers.

Response: That part of the discussion was rewritten and our hypothesis deleted.

p. 12, lines 18-20: As noted for the introduction, it isn’t possible to take the results of one study into pasture soils from one country and extrapolate to a global model about enzyme activity, especially when the current study did not include any measures of enzyme activity. This needs to be deleted.

Response: Sentence and assumption about those results were excluded.

p. 12, lines 23-26: As noted above, the influence of pH on DNA sorption is well-established (Condron et al. 2005). The authors need to revise these lines of the discussion.

Response: The discussion was strongly improved following the note above.

p. 12, lines 28 to p. 13, line 12: The authors need to be very precise in their terminology. In this section, they discuss “inositol phosphates”. However, the data they include from various studies is for inositol hexaphosphates, which very specifically are inositols with 6 phosphate groups. These will behave very differently in soils from inositol phosphates with fewer phosphate groups. Please rewrite this section to be more precise. In addition, all of the processes discussed here to govern the behavior of “inositol phosphates” in soil will also apply to other P forms, especially DNA.

Response: the terms "inositol phosphates" were changed to "inositol hexakisphosphates" throughout the manuscript. Moreover, the rationale was changed to include both inositol hexakisphosphates and DNA.

p. 13, lines 9-10: This sentence discusses changes in the concentration of "inositol phosphate" with soil weathering. However, the authors only present changes in "inositol phosphates" as proportions of organic P, not as concentrations. As such, this is all speculation and should be revised or deleted.

Response: The text was revised. We think that the rationale is important in the manuscript to justify why inositol hexakisphosphates concentrations could decrease in more weathered soils. In fact, all of our tropical results had negligible inositol hexakisphosphates concentrations. To complement the rationale we added a text that describes our results in concentrations from figure 7.

p. 13, line 14: "there were no inositol phosphates on tropical, more weathered soils". Could this not also relate to either inputs from plants or production of phytases by plants and microbes? The authors did not include any factors that might influence the cycling of IHP in these types of soils. As such, this is merely speculation and should be deleted.

Response: The text was rewritten including the plant role on the inositol hexakisphosphates acquisition and more references were added. The motive for not excluding the rationale is described in the comment above.

p. 13, lines 20-23: "Recent investigations have contradicted the often-cited literature" please cite references here to support this statement of fact, both for the "often cited literature", and the "recent investigations".

Response: the phrase was modified excluding the "often cited literature" part and references for "recent investigations" were added across the present and next paragraphs. We thought it would be redundant citing the recent investigations references at the present sentence and again along the following sentences.

p. 14, lines 10-30: One thing missing from this section is the role of vegetation with respect to inputs of different P forms. It should also be noted that a greater soil wetness is not necessarily associated with leaching, if there is impeded drainage of the soils in some ways (e.g. from the formation of placic horizons). This section of the manuscript is really vague, and doesn't add anything to our knowledge of P

cycling in soils. The authors must do a far more thorough literature review to discuss their results properly.

Response: We agree that vegetation may have played a role on the results, but we were unsuccessful in finding studies that clearly explained the role of vegetation, without confounding effects of soil or climate, on specific soil P compounds. These issues were also recently pointed out by another paper that dealt with the same dataset (Huang et al. 2017). Therefore, we added a section to the end of the discussion, along with other suggestions made by the other reviewer, to point out future research priorities. These included, among others, a suggestion for studies aiming to disentangle confounding effects among vegetation, soil biotic and abiotic components, and climate. The terms "Soil wetness" were excluded and that part was rewritten using "precipitation (in soils with no impeded drainage)".

p. 14, line 32: "As the orthophosphate percentage decreased following precipitation, the pyrophosphate percentage increased". This does not necessarily mean that the concentration of orthophosphate decreased, or that of pyrophosphate increased, or imply a cause-and-effect relationship. This line suggests to me that the authors do not understand compositional analysis: as the percent of one thing decreases the percentage of another thing will increase because the total must add to 100%. In my opinion, reporting and discussing these as percentages is misleading, for this reason. It would be far better to use concentrations.

Response: We completely agreed with the reviewer's point so we added the concentration results from Figure 7, which followed the same behavior, to imply in the rationale a cause-and-effect relationship.

p. 15, lines 6, 7: "phosphorus" should be "P"

Response: Changes were made.

p. 15, lines 7-8: "greater organic P concentrations were associated with increasing biomass production".

Response: Changes were made.

Studies of biomass (e.g. Noack et al. 2012) show that the majority of P in plant biomass is as orthophosphate, not organic P compounds. As such, this sentence doesn't make sense to me, and isn't supported by the literature. Please revise.

Response: We acknowledge that most of P in crop biomass (e.g. Noack et al. 2012) and possibly native vegetation biomass is as orthophosphate. To clarify the rationale we added a note "However, it is important to note that the majority of P in plant biomass (e.g. Noack et al. 2012) is as orthophosphate and not as organic compounds. We believe that even with higher orthophosphate inputs through plant biomass, organic P concentrations in soils would increase altogether with inorganic P concentrations, and also at expense of soil orthophosphate due to the greater bioavailability to plants and organisms of that inorganic form."

8. Conclusions:
p. 15, lines 22-23: "after altogether with plant and microbe coexistence" as noted for the abstract, I do not understand what the authors are trying to say here. Something seems to be missing, or mistranslated.

Response: The sentence was rewritten to "and then altogether through plant and microbe P cycling".

9. Figures: In all figures, "phosphonate" should be "phosphonates", because this is a general compound category containing a number of specific P compounds. Also, "inositol" should be IHP or inositol hexakisphosphate.

Response: All figures were changed to correct "phosphonate" to "phosphonates", and "inositol" to "IHP". We also corrected the line 21 of Figure 7 to a dotted line.

10. References: there are many problems with this section of the manuscript. The authors need to carefully proof- read and correct this section.

. a) Many of the references are out of order alphabetically, including the entire "Y" section (which should come after W), Deiss et al. 2017 should come after Damon et al. 2014; Laliberté et al. 2017 should come before Legendre and Legendre 2102 and Li et al. 2015; Whittaker 1975 should come before Wilson et al. 2013.

Response: All references were ordered. Y is coming after X, Y after W. Deiss after Damon. Laliberté was placed before Legendre and Legendre. Whittaker was placed before Wilson.

. b) There are formatting differences among references. Cade-Menun et al. 2000 has the volume and page number listed as 30:1714-1725, while other references use a comma (e.g. Celi et al., 2013, 367, 121-134). Chen et al. 2004 does not include periods after abbreviations in the journal title (Aust J Soil Res); Cade- Menun and

Preston 1996, Kizewski et al. 2011, Turner et al. 2013 have all the words capitalized in the manuscript title.

Response: volume and page number were standardized across all references. Periods after abbreviations in the journal title were added for Chen. Capitalized letters of title name of Cade-Menun and Preston 1996, Kizewski et al. 2011, Turner et al. 2013 were turned to lowercase, and other occurrences were followed by the same rule.

. c) The journal volume number and/or page numbers are missing from Deiss et al. 2017, Doolette et al. 2016, Li et al. 2015, while George et al. 2017 is missing the name of the journal as well as the volume number.

Response: Deiss, Doolette, and Li were completed with volume number and/or page numbers, and George was completed with Journal Name. We did not find volume number for the latter.

. d) Rumpel et al. is 2015 in the References, but 2016 in the text; Bünemann is "Bunemann" in the text (p. 11, line 16)

Response: Rumpel was corrected to 2015. Bünemann was corrected in the text.

REPORT 2

Reviewer Comments
The manuscript presents some interesting results and the results are now quite clearly presented. While some aspects have been improved in the revision, other aspects remain problematic, including some issues already pointed out by earlier reviewers. The four main issues I see are:

1. Inadequate understanding / discussion of the limitations of the NMR method

2. Precision and clarity of terminology

3. A discussion section that does not go beyond what is already known

4. English language

Limitations of the NMR method.
Liquid 31P NMR is a powerful method to study the presence of certain P forms in soils, and it is of interest to combine results produced by different studies in a meta-analysis. However, it is absolutely necessary that the authors are aware and clearly and transparently communicate the limitations of liquid 31P NMR on NaOH-EDTA extracts. There are many other methods out there to study P cycling (Kruse et al. 2015), and only if results are correctly interpreted in light of the limitations of the methods can readers integrate knowledge gained from one method with knowledge gained from another method.

Response: We added the rationale for the introduction to clarify the limitations of the 31P NMR following the reviewer suggestions.

Firstly, as already pointed out by reviewer 1 (first round), NaOH-EDTA extraction never extracts all soil P and often only a small portion of total soil P. Please state the fraction of total P extracted by NaOH-EDTA for all soils used in your analysis.

Response: The fraction of total P extracted by NaOH-EDTA is being reported on the Figure S5, and was used in all models as a random factor, but to complement those, we added the average and range of % extracted by NaOH-EDTA for both mineral and organic layers to the results section (end of second paragraph of results section).

Secondly, 31P NMR is a tool for P speciation, providing important information on P stocks, it does not, however, provide information on dynamics. Turnover, exchange kinetics, mineralisation rates, etc, which is what I understand under “P dynamics”, can only be assessed using isotopic techniques (Frossard et al. 2011).

Response: The "dynamics" term was excluded from the manuscript when referring to soil P composition results.

Thirdly, liquid 31P NMR is not the preferred method for studying inorganic P species, since it cannot give information on predominant inorganic P species such as Fe-, Al- and Ca-phosphate. XANES is a more preferred method for looking at inorganic P species. This does not mean that the results on pyrophosphate, orthophosphate and polyphosphates concentrations are not useful, however, the paper reads as if these are the only inorganic P species of importance in soils. Please be more honest on the potentials and limitations of this approach.
These limitations need to be stated clearly and transparently in the introduction, since they provide the scope for the meta-analysis.

Response: As described earlier, we added this rationale on the introduction to clarify the limitations of the 31P NMR and other methods for studying soil P. Moreover, we included this topic about the orthophosphate speciation as a main "Research Priority" in the last, new section of discussion.

Precision and clarity of terminology. As mentioned above, the word "dynamics" is misleading for a study looking at P forms. I suggest changing the title to something more accurate, e.g. "The impact of soil, climatic, and temporal drivers on inorganic and organic P compounds".

Response: The "dynamics" term was excluded from the manuscript, and a new title was introduced.

An often-recurring term is "complex P compounds". From my understanding, "complex" is used to refer to high-molecular weight organic compounds of variable composition (McLaren et al. 2015). Please use a more precise term than "complex P compounds".

Response: The terms "complex P compounds" throughout the manuscript. Complexity, otherwise, was maintained in some instances to explain modifications in soil P composition in later stages of pedogenesis.

Discussion section
The discussion section remains the main weak spot of the article. As it stands, the discussion could be summarized by Fig 1 from the introduction. This is a problem because Fig 1 is a very generic and commonly used figure in the field. I suggest the authors follow Mensh and Kording (2017), who provide useful tips for structuring a discussion (Mensh and Kording 2017).

There follows a few ideas for rewriting the discussion that authors may use if they find them helpful. The first paragraph can more or less stay as is, it summarizes the results.

Response: We thank the reviewer for the suggestions about the paper to better structure the discussion. We followed it to improve the discussion presentation and organization. We maintained those initial paragraphs of the discussion.

The next two paragraphs could be dedicated to outlining the limitations of the analysis. Here it would
be worth pointing out that climate and weathering drive soil properties, so that it is not entirely appropriate to compare their influence on P forms, because much of the variation in soil properties can be explained by climate and weathering. Also, it might be worth discussing why variation in polyphosphates could not be explained by the models.

Response: We added the point about interacting effects between climate and weathering on soils along with the first paragraph of discussion ("While the soil P composition was mainly directly influenced by soil properties, the impact of climate and weathering stage occurred mainly through indirect paths and their influence on soil properties"), and we also included this topic in the discussion "Research Priorities" section. We added a possible explanation to the poor response of the polyphosphates at the end of the third paragraph of the item "4.1 Soil properties and the soil P composition". The other reviewer presented an interesting point and we believe that it had a role in our responses. So we added this specific discussion at that part.

The final two paragraphs could point out how this study adds to the literature. The focus of these paragraphs should not be comparisons to individual NMR studies, since they are anyways incorporated into the data, rather it would be interesting to weave the findings together with insights from studies using XANES, enzyme activities, or isotopes, to start painting a full picture.

Response: We added a new section in the discussion to point out future opportunities for studies we struggled to find responses for while exploring our results.

English language
Authors stated that a native English speaker revised the manuscript. However, I don't think that the quality of the writing has improved from the previous version. There are still many grammatical issues and awkward writing, which make the manuscript difficult to read at times.

Response: A different native person with an affinity for the topic revised the English again throughout the manuscript.

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Environmental drivers of soil phosphorus composition in natural ecosystems

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Abstract. Soil organic and inorganic phosphorus (P) compounds can be influenced by distinctive environmental properties. This study aims to analyze soil P composition in natural ecosystems, relating organic (inositol hexakisphosphate, DNA and phosphonates) and inorganic (orthophosphate, polyphosphate and pyrophosphate) compounds with major temporal (weathering), edaphic and climatic characteristics. A dataset including 88 sites was assembled from published papers that determined soil P composition using one-dimensional liquid state ^{31}P nuclear magnetic resonance of NaOH-EDTA extracts of soils. Bivariate and multivariate regression models were used to better understand the environmental properties influencing soil P.P. In bivariate relationships, trends for soil P compounds were similar for mineral and organic layers but with different slopes. Independent and combined effects of weathering, edaphic and climatic properties of ecosystems explained up to 78% (inositol hexakisphosphateshexakisphosphates) and 89% (orthophosphate) of organic and inorganic P compounds variations across the ecosystems, likely deriving from parent material differences. Soil properties, particularly pH, total carbon and carbon-to-phosphorus ratios, 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 in 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 withwith plant and microbe PP cycling, 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 evolves. Soil organic and inorganic compounds respond differently to combinations of environmental drivers, which likely indicates that each P compound has specific factors governing its presence in natural ecosystems.

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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 a youngyoung ecosystem derives predominantlypredominantly from the weathering of parent materialmaterial, with some systems receiving P input from eolian deposits (Chadwick et al. 1999). Once P has been dissolved from primary minerals, plants and microorganisms access it from the soil solution. This phosphorus is then recycled through soil as organic and inorganic P compounds (Noack et al. 2012; Damon et al. 2014), which are similarly subjected to a new cycle of physico-chemical and biological reactions. Each iteration of this cycle alters the form and bioavailability of the phosphorus, leading to decreasing levels of bioavailable P compounds (McDowell et al. 2007). In the absence of 'fresh' P inputs, this results in severe P) limitations to ecosystem productivity (Walker and Syers 1976). TheT five state factors of soilsoil formation (time, parent materialmaterial, climate, topography and biota) determine the rate at which the cycle is completed (Jenny 1941). Therefore, a better understanding of the role of the five state factors as drivers of soil P composition is crucial to quantifying the relative abundance and form of bothQuantifying organic and mineral P pools soilcomposition.

In soils, inorganic and organic P (P_i and P_o, respectively) pools are each composed by specific P compounds (species). The main P_o compound categories are: i) orthophosphate monoestersmonoesters (single ester linkage to orthophosphate) such as inositol hexakisphosphateshexakisphosphates, ii) orthophosphate diesters (two ester linkages to orthophosphate) such as ribonucleic acid, deoxyribonucleic acid, lipoteichoic acid, phospholipid fatty acids, and iii) phosphonates (Nash et al 2014). Inorganic P compounds include orthophosphate, polyphosphate and pyrophosphate (Cade-Menun and Preston 1996). Specific phosphatase enzymes are required to transform the different P_o and P_i forms into orthophosphate, which is the P compound directly taken upup by plants and microbes (Turner 2008a). AsAs most enzymes, the activity of soil P cycling enzymes are very sensitive to thethehydrogen potential (pH) with specific enzyme optimum (Frankenberger and Johanson, 1982)However, other O soil variables are also involved in regulating P_i and P_o transformations. For example, inositolInositol hexakisphosphates bind strongly to metal oxides and other soil components, which strongly limitsconstrained their bioavailability (Turner et al. 2007). Similarly, amino group protonation of adenine, guanine and cytosine bases in the DNA molecule can cause adsorption of positively charged DNA to the negatively charged clay surface (Yu et al. 2013). As a result, many soil properties regulate soil P composition but their relative importance across contexts is unclear.

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 soils undergo pedogenesis, ecosystem productivity progresses from nitrogen (N) to P limitation with ecosystem productivity peaking at the N-P colimitation intermediate stage of pedogenesis (Walker and Syers 1976; Turner and Condron, 2013). Parallel changes occur in soil properties including a decrease in total exchangeable bases, an increase inacidification, Al and Fe oxide concentration (Albrecht 1957; Walker 1965). As a result, some P_o and P_i

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compounds increasingly react with the mineral surface and progressively become occluded P (Yang and Post 2011). Subsequently, the complexity in in P_o and P_i composition increases during ecosystem development (McDowell et al. 2007, Figure 1, bottom panel). The degree of soil weathering is inherently linked to the state factor 'time'—as demonstrated along many chronosequences (e.g. Turner and Laliberté 2015)—but it can be altered through other state factors (Albrecht 1957), such as along climosequences (e.g. Feng et al. 2016) or toposequences (e.g. Agbenin and Tiessen 1995). Along a climosequence, precipitation increased both base cation leaching and the degree of soil weathering, whereas potential evapotranspiration decreased these processes (Feng et al. 2016). While this study evaluated the mineral-P associations described by the Hedley P fractions, rather than P speciation, it illustrates the opposing effects of various climatic factors on edaphic factors of interest.

As an overlooked state factor, parent material has distinct effects on soil properties, all all other state factors otherwise being equal. Some of these effects are direct effects, such as the total P concentration of the initial geologic materia. However, other factors may be more indirect. Parent materials can differ in total exchangeable base concentration and mineral composition. Variation in mineral composition can lead to differences in soil pH, soil texture, and Al and Fe oxides, all of which influence soil P cycling and P composition. For instance, soil P retention potential is influenced by differential absorption of P_i and P_o dto clays soluble Ca content, as well as and Al and Fe oxyhydroxides (Batjes 2011). As such, the parent material statefactor is an essential consideration in describing soil P cycling. Most importantly, we need to investigate the hierarchical nature of causal effects between state factors, soil weathering, soil properties, and P_o and P_i composition.

worldwide Nuclear magnetic resonance spectroscopy (NMR) is a widely-used method to study P_o and Pi compounds in ecosystems around the world (Kizewski et al. 2011). This technique can be used for both qualitative and quantitative estimates of P compounds in soil (Cade-Menun and Preston 1996). The most effective extractant for NMR analysis has been NaOH combined with the 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 widely recognized to quantitatively extract P compounds from the soil (Turner and Blackwell 2013, Cade-Menun and Liu 2014). However, there are drawbacks of using NaOH-EDTA extractant for ^{31}P NMR analysis. NaOH-EDTA does not extract all soil P and the highly alkaline environment can potentially degrade some P compounds (Cade-Menun et al. 2006; Cade-Menun and Liu 2014). Additionally, the NaOH-EDTA extraction does not separate Fe-, Al- and Ca-phosphate compounds (Kizewski et al. 2011). ThereNonetheless, there are many other methods available to study P dynamics (Frossard et al. 2011), and soil P composition (Kruse et al. 2015) in ecosystems. X-ray absorption near edge structure (XANES), is a more preferred method for looking at orthophosphate speciation (Hesterberg 2010). This does not mean that the results on pyrophosphate, polyphosphates and total orthophosphate concentrations are not useful, however, there are other inorganic P compounds of importance in soils.

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There is a lack of broader understanding of how soil P composition is affected by different state factors of soil formation. Using a large-scale comparative geographical approach, we aim to determine the causal paths through which climate, parent material and time influence soil properties, as well as and their impact on P_i and P_o pools and specific P compounds. CWombining the soil organic and inorganic P results obtained with ^{31}P NMR using NaOH-EDTA from different studies, allows usenable to describe the effect of state factors on soil P composition in natural ecosystems. We hypothesize that the compounds comprising soil P_i and P_o will be modified by distinctive edaphic and climatic properties due to different key ecological processes coupled with soil P cycling.

2 METHODS

2.1 Dataset

A database search was conducted until November 17, 2017, to identify published papers that accurately determined soil P compoundscompounds 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 as accurate the papers that used 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 followedpre-defined eligibility criteria to consider the papers, and then to select or reject them.m were.The first criteria was that Nonly native growth media were considered.manure, pot soil, soil leachate and sediment samples were excluded). In studies focusing on changing natural conditions, only the control (unchanged) samples were used,used(e.g., litter removal in Vincent et al. 2010 was excluded). Next we only considered studies where theThe one-dimensional liquid state ^{31}P NMR method was usedwith the following features: 1)NaOH-EDTA extractor without pretreatment (0.5 or 0.25 M NaOH and 0.1 or 0.05 M EDTA), 2) delay times > 2 s (i.e., quantitative data, see Cade-Menun and Liu 2014); 3) NMR features or explanations according to ^{31}P NMR principles (see Cade-Menun and Liu 2014); and 4) total NaOH-EDTA extracted P and total P. Both top mineral and organic layers were considered. From Fromselected we compiled: total P, total NaOH-EDTA P, andNaOH-EDTA organic P, as well as the compoundscompounds inositol hexakisphosphates (*myo*-, *scyllo*-, *neo*-, and D-*chiro*-IHP, when available), deoxyribonucleic acid (DNA), phosphonates, NaOH-EDTA inorganic P and the compounds orthophosphate, pyrophosphate and polyphosphate. No duplicity was found in the selected papers.

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2.2 Site environmentalS environmental properties

Soil texture, total C, total N and pH, short range ordered Al and Fe minerals (poorly crystalline) estimated with oxalate extraction, climate characteristics (mean annual precipitationand mean annual temperatures)) as well as soil age, when available, were also collected from the papers. WhenW the total C was unavailable, the organic C was assumed to be the total C. This assumption only occurred for non-calcareous soilsSome variables were unavailable for some results, and the number ofexperimental 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 inAppendix S1. We used the Whittaker's diagram (Whittaker 1975) and the "BIOMEplot" package (Kunstler 2014) to determine thebiomes ofof our sites (Appendix S2).

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-regression models including edaphic and climatic variables as continuous and 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 toto control for the spatial auto-correlation (MaestreMaestre et al 2005). 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 concentration concentration along the soil profile. The bivariate effects of latitude, percentage of P extracted and sampling depth on the soil P composition are presented in additional Appendices S3-S5.

We used variation partitioning and Venn diagrams (Legendre and Legendre 2012) (the '*vegan*' package) to partition the total variation explained uniquely by the matrix of either soil variables, climate variables or soil weathering stages as well as the variation explainedby 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.

Structural equation modeling (path analysis, the '*lavaan*' package) was used to explore how variations in inorganic and organic P compoundsare driven by both direct and indirect effects of key environmental drivers (soil, climate and parent

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material) material. We first established an *a-priori* model that is based on our knowledge and is presented in additional Appendix 10.Appendix 10. Then, we tested for the most parsimonious model among many alternative ones, i.e. the one that differed the least from the observations and presented thethe greater *P*-value. Parent material, which was unknown for our sites, was considered as a latent variable in the model to explain the remaining coordinated variations in total P, clay and pH variables that were not explained by climate and weathering. As mainly determined by the biota, total C was not considered as being constrained by the parent material.

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). In contrast, in both Venn diagrams (Figure 6) and structural equation modeling (Figure 7) soil P compounds were in mg kg^{-1}. In bivariate relationships, we used percentages was to compare the relative composition of P along environmental variables that are linked with the weathering of soils. In both the 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 (additional Appendix S1) (Backnäs et al. 2012, n=1; Celi et al. 2013, n=4; Doolette et al. 20177, n=5; Li et al. 2015, n=1; McDowell and Stewart 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; Vincent et al. 2013, n=8; Vincent 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's diagram (Whittaker 1975), except for 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 hexakisphosphates results (including all tropical regions), and 12% of DNA results were absent (including both non-tropical and tropical regions). The P extracted with NaOH-EDTA on mineral layers averaged 55% (2 to 98% range), and on organic layers averaged 73% (57 to 94% range) of total soil P.). The average sample depth was on 12.2 cm (2 to 42 cm range) for mineral layers), and 11.0 cm (1 to 28 cm range) for organic layers.).

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3.1 Edaphic properties

All surveyed 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, although this effect was constrained to the mineral layers. In mineral layers, the proportion of NaOH-EDTA P in the form inorganic P increased with pH (Figure 2B), whereas the proportion in the form of organic P decreased with pH (Figure 3B). NaOHNaOHNaOHHowever, there was no pH effect on both pools in the organic layers. The distribution of compounds in both P_i (Figure 2C-E) and P_o (Figure 3C-D) pools responded dynamically to the pH. In the P_i pool (% of P_i) of mineral layers, orthophosphate decreased, and pyrophosphate accounted for the remaining P_i as the pH decreased. The pH had no effect on 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 S3 shows the sampling depth effect over the soil P composition). In the P_o pool (% of P_o), both inositol hexakisphosphates (mineral layer) and DNA (mineral and organic layers) proportions increased as the pH decreased. Phosphonates response to edaphic properties (insignificant) is presented in 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 3E). Clay impactedimpacted on neither P_i and P_o, nor on the proportions of P compounds (% of NaOH-EDTA P) (Figures 2G-J and 3F-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 3I). 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 on either P_i and P_o proportions in organic layers (Figures 2L and 3J). 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 (phosphonates, inositol hexakisphosphates and DNA) in either mineral or organic layers (Figure 3K-L; additional Appendix S6).

Both total P_i and P_o concentrations inNaOH-EDTA extracts (mg kg^{-1} soil) from both mineral and organic layers increased as the total soil P concentration increased (Figures 2P and 3M). Only the DNA compound from the P_o pool (% of P_o) in the mineral layer was affected by the total soil P concentration (Figure 3P). As the total soil P concentration increased, the DNA proportion 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 it includes both the extracted P and the residual P.P The recovery of the total P by NaOH-EDTA extraction varies depending on soil characteristics and laboratory procedures (Cade-Menun and Liu 2014).

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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 3Q). As aa 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 3R). As proportions in the P_i pool (% of P_i) of mineral layers, orthophosphate decreased 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 orthophosphate decreased 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_o), the DNA proportion increased as the soil CP ratio increased, only in the mineral layer (Figure 3T).

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 additional Appendices S7 and S8. The mean annual temperature, ranging from -0.4 to 27 Cº, did not promote any change in the soil P composition in natural ecosystems. There was no effect of climatic variables on total P_i and P_o concentrations in NaOH-EDTA extracts (mg kg^{-1} soil) (additional Appendices S7A and S8A). As a fraction of the NaOH-EDTA extract (% of NaOH-EDTA P), P_i decreased and P_o increased as the precipitation increased (additional Appendices S7B and S8B). As the precipitation 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 soil age and CP ratios ratiosfollowing an expected effect of pedogenesis (Figure 4). 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 5). Total P_i and P_o inNaOH-EDTA extracts (mg kg^{-1} soil) were more concentrated in soils at moderate weathering stages when compared to low and high weathering stages (n=79, Figure 5A, F). As percentages in 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 5C-D). In the P_o pool (% of P_o), the DNA (n=64) proportion was greater in more weathered stages, and there was no effect of weathering stages on phosphonates (n=79) and inositol hexakisphosphates (n=52) proportions (Figure 5H-J). Using available data (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 S9).

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

The variation partitioning of ecosystem 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 6). For the total P_i concentration and its compounds orthophosphate and

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pyrophosphate, the total variation explained by models ranged from 46% to 89%, 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 ecosystem 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 6). The total P_o, inositol hexakisphosphates and DNA had their 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, most of the variation in phosphonates was explained by combined effects of climate and soil variables, followed by uniquely soil variables.

3.5 Interdependences between environmental variables and 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; additional Appendix S10). The parent material 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 up to 78% of P_o compounds variation and 89% of P_i compounds variation.

Following an expected effect of pedogenesis, the path analysis indicated that the parent material (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 in 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. In 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.

In the P_i model, orthophosphate was negatively related to precipitation, and it was positively influenced by soil total P and total C. Pyrophosphate was positively influenced by precipitation, soil total P and total C. Polyphosphate wasas negatively influenced by temperature, and it was positively related to soil pH. In the P_o model, inositol hexakisphosphates werewere 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. Phosphonates werere negatively affected by temperature and weathering, but positively affected by precipitation and soil total C.

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4 Discussion

Our results showed how soil P_i and P_o compounds responded to edaphic variables (Figures 2 and 3), climatic variables (additional Appendices S7 and S8), and soil weathering stages as a proxy for pedogenesis (Figures 5) onon a wide geographical scale, including a variety of natural ecosystems. While the soil P composition was primarily directly influenced by soil properties, properties the impact of climate and weathering stage occurred mainly through indirect paths and their influence on soil propertiesproperties (Figures 6 and 7). In addition, soils P_i and P_o compounds responded toto different combinations of explicative variables, which likely indicates that each P compound has specific factors governing its presence, transformation and persistence ini ecosystems. This could be due to many factors including: i) the source of P inputs, primarily by minerals, and then altogether with plant and microbe P cycling; cycling, ii) the)T presence of specific phosphatase enzymes that are required to transform P_i and P_o compounds into orthophosphate; iii) the soil)The s specific reactivity and P losses governed by physico-chemical properties (e.g., clay, short-range ordered oxides and pH); and A iv) the P persistence induced by the increasing complexity of P organic and inorganic compounds as pedogenesis evolves.

As time passes after the pedogenesis onset, the ecosystem accumulates organic matter up to a maximum, and then starts to decline. Along with this decline, there are also changes in the chemical composition of organic matter, in which the decaying degree of C element is lower than the P, and concomitantly there is an increasingly acidic soil environment (Walker 1965; Turner and Condron 2013). In addition, parent material supplies cations and orthophosphate to young soils, whereas more weathered soils are substantially changedchanged from the parent material. 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 included 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 increasesthrough the encapsulation of the organic and inorganic P compounds inside of Fe and Al minerals (McDowell et al. 2007; Turner et al. 2007). 2007.

Even though most results were from New Zealand and Panama, our dataset comprised several biomes according to the 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, quantitative data on the feedback between P compounds and biological communities during pedogenesis is still incipient to conclusions drawn 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 extent, soil

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food webs. In a global analysis, Maire et al. (2015) demonstrated that the soil available P is a key environmental dimension increasing leaf P content along withwith species' maximum photosynthetic rates and lower stomatal conductance. However, this trend is expected to gradually decline in more weathered soils due to a lower P availability. Conducted at a narrower scale, Laliberté et al.'s study (2017) showed that soil fertility (including P availability) strongly shapedd 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 soils aged, pyrophosphate and polyphosphate may 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 weathering stage had a major influence on the soil P_i pool composition. As the orthophosphate proportion decreased in more weathered, acidic, organic-rich, and P-limited soil environments (Figures 2C, M, W and 5C), pyrophosphate and polyphosphate proportion increased and dominated the P_i pool (Figures 2D, N, O, X and Y and 5D).

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). Bünemannü 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 proportion 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, acidified soil conditions (Wang and Qiu 2006). Ectomycorrhizal fungi convert the orthophosphate that they take up from the soil into polyphosphates, and translocate the polyphosphate along fungal hyphae, sometimes at a great distance from where the orthophosphate is taken up (Bücking and Heyser 1999; Plassard and Dell 2010). 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, but much information is still needed in regard to plant and microbial communities characterization in studies of P forms. These organisms could have helped to deplete and transform the bioavailable orthophosphate, turning it into more microbial biomass derived P compounds as pedogenesis progressed in these environments.

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 soil pH (Sutton and Larsen 1964), and this may have contributed to 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

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compared to orthophosphate and pyrophosphate.yrophosphate. 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 is produced. However, caution must be taken when interpreting pyrophosphate and polyphosphate hydrolyzation results from ^{31}P NMR analysis on NaOH-EDTA extracts. Polyphosphates can potentially degrade to pyrophosphates during extraction and NMR analysis of P (Cade-Menun et al. 2006), so they cannot be considered as fully distinct P forms. This potential degradation could be one explanation to why the polyphosphates results were poorly explained by the variation partitioning and structural equation models.

As time passes after the onset of pedogenesis, modifications in the soil P_o composition were possibly related to the acidifying environment in soils, which may increase the charge of some organic compounds, and thus increase sorption.sorption. Soil pH affect the sorption of inorganic and organic P compounds by soils, but different P compounds respond differently to pH changes (Shang et al. 1992). Shang et al. (1992) verified that sorption of both orthophosphate and inositol hexakisphosphate by Al and Fe precipitates generally decreased as pH increased, whereas there was little pH effect on the adsorption of glucose 6-phosphate by both precipitates. Sorption of inositol hexakisphosphate to minerals surface is often stronger than orthophosphate, but both tend to be less sorbed to those minerals in neutral to alkaline soils (Berg and Joern 2006; Xu et al. 2017). The presence of humic acids may affect the amount of inositol hexakisphosphate that sorbs on the minerals surface at lower pH values, but it cannot displace inositol hexakisphosphate from that surface (Ruyter-Hooley et al. 2016). Moreover, another study founds that inositol hexakisphosphate sorption in soils was unaffected by the presence of orthophosphate, β-D-glucose-6-phosphate or adenosine 5′-triphosphate (Berg and Joern 2006). Following a similar pattern, the amount of DNA bound on the clay minerals such as montmorillonite, kaolinite, hydroxyl aluminum species and variable charge soil colloids also increased by lowering pH of solution (Khanna and Stotzky, 1992; Cai et al. 2006; Cai et al. 2008; Wang et al. 2009; Saeki et al. 2010). At pH < 5, protonation of the amino groups of adenine, guanine and cytosine occurs and causes the increase of net positive charge of DNA and electrostatic attraction between negatively charged tetrahedral silica layer on the clay surface and DNA (Yu et al. 2013). Therefore, the increasingly acidic pH could have increased sorption, and therefore facilitated inositol hexakisphosphates (Figures 3C) and DNA (Figure 3D, Figure 7) accumulation in those soils.

In our study, we found that there was an increasing proportion of inositol hexakisphosphates and DNA in the P_o pool (% of NaOH-EDTA P) as pH decreased, and there was predominance of inositol hexakisphosphates in acidic, more weathered soils (Figures 3C-D, 5J). The hierarchy of investment for the P acquisition through enzymatic activity maymay also be a factor that contributed to modifications in the soil P_o composition as time passes since the onset of pedogenesis. According to Turner et al. (2018), Turner and Haygarth (2005) and Kunito et al. (2012), P limitation may stimulate increased phosphoesterase synthesis as a way to increase bioavailable P by the mineralization of organic P.PFungi are well 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

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high level of microdiversity (Zimmerman et al. 2013, George et al. 2017). Plant and microorganism that 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 (Turner 2008a). Although inositol hexakisphosphates are also classified as part of monoesters, they need a higher investment in organic P acquisition than other monoesters and diesters because they can 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 hexakisphosphates may accumulate more than DNA, and the latter more than other organic P compounds.

Nonetheless, inositol hexakisphosphates can persist as a main P_o compound up to a certain point of pedogenesis, and then decline in more weathered soils. SomeSome authors described that inositol hexakisphosphateshexakisphosphates declined to lower concentrations in older soils of non-tropical regions (Turner et al. 2014; Turner et al. 2007), and most tropical soils had negligible inositol hexakisphosphates contribution (e.g., Turner and Engelbrecht 2011). In our results, soil weathering had no effect in inositol hexakisphosphates concentrations in mg kg^{-1} in non-tropical environments (Figure 7), but in fact, all compiled results from tropical soils did not had inositol hexakisphosphates. Inositol hexakisphosphates have been found in very weathered soils (e.g., Oxisols), but under agricultural management that included well-known sources for that P compound such as plant seeds (Turner 2006; Smernik and Dougherty 2007; Deiss et al. 2016). These results suggest that inositol hexakisphosphates could occur in tropical soils under native vegetation, but they are either being rapidly turned into bioavailable compounds (by plants and microorganisms) or inputs of inositol hexakisphosphates, which are abundant in seeds and pollen (Raboy 2007), are lower in lowland tropical forests compared to temperate ecosystems (Turner and Engelbrecht 2011). In P limited soil environments, the acquisition of inositol hexakisphosphates may be strongly improved by root exudates, which may increase the solubility of these compounds in soil (Gerke 2015). 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 these environments. Finally we believe that otherother P compounds such as DNA (Figures 3D, 5J) and pyrophosphate (Figures 2D, 5D) will possibly prevail in more weathered systems from tropical regions because they are intrinsic components of the microbial biomass (Kornberg et al. 1999). Mature soils are known for having the microbial P as the main component of the P pool (Turner et al. 2013).).

Soil clay concentration affected both soil total P_o and P_i concentrations (Figures 2F and 3E), but had a minor association with soil P compounds. Recent investigations have foundfound thatorganic compounds stabilization may be mainly driven by factorsthat are typically minor constituentsconstituents of the clay-sized fraction by mass, buthighly reactive components. Vogel et al. (2014) showed that 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. 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 aggregates, most

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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 as observed in our study.

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 absorb and stabilize soil organic matter; however, we did not observe a significant overall soil weathering effect on Al and Fe oxide concentrations (additional Appendix S9), suggesting that it may depend on other factors such as the parent 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 others 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. 20155). Therefore, we could not confirm the role of Al and Fe oxides as influenced by soil weathering stages over the soil P composition.

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4.2 Climate and the soil P composition

Climatic variables exerted an important role on the soil P composition but to a lesser extent when compared to soil variables (Figure 6). 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 their compounds (additional Appendices S7 and S8). It was expected that the soil P_o concentration would decrease with increasing temperatures because higher temperatures are optimal for the breakdown of the soil P_o compounds by the microbial biomass through phosphatase enzymes release (Turner et al. 2002). Another investigation confirmed that greater maximum phosphatase activity occurred at incubation temperatures >25°C when compared to 20°C, but no differences were observed among temperatures greater than 25°C (Hui et al. 2013). Therefore, phosphatase 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 precipitation (in soils with no impeded drainage) reduced the inorganic concentration of P linked to Ca, corresponding to a marked decline in soil exchangeable Ca and suggesting an

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enhanced leaching of P along with weatherable cations. Moreover, greater soil 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 and concentration (additional Appendix 7C and Figure 7, respectively) decreased following greater precipitation, the pyrophosphate percentage and concentration (additional Appendix 7D and Figure 7, respectively) increased suggesting that this compound predominates underunder these environmental conditions. 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 (Figure 7, additional Appendix 7D) possibly mirrored the response of the total P_o (additional Appendix 8B) to climatic variables, which may have resulted from greater soil organic matter accumulation following greater productivity (i.e., plants and organisms) in these ecosystems with greater water availability. Evaluating the P budget of the whole ecosystem, Turner et al. (2013) demonstrated the dominance of microbial P 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. However, it is important to note that the majority of P in plant biomass is as orthophosphate (e.g. Noack et al. 2012) and not as organic compounds. Even though, we believe that with higher orthophosphate inputs through plant biomass, soil organic P concentrations would increase altogether with orthophosphate P concentrations, and also at expense of soil orthophosphate due to the greater bioavailability to plants and organisms of that latter P compound.

Changes in vegetation are expected to occur during pedogenesis, and climatic variables may govern magnitudes of these alterations along with soil changes. Vitousek et al. (1995) showed that as ecosystems develop, the pattern of P concentration in plants leaves follows a non-linear response to time, in which lower concentrations occur at either early or late stages of pedogenesis, and a maximum is reached at an intermediate stage of pedogenesis. In addition, precipitation can affect the magnitude of that maximum response (intermediate stage), where the P concentration in plant leaves is higher in mesic gradients when compared to more wet gradients (Vitousek et al. 1995). Moreover, as described earlier, the soil available P, along with other climatic variables, governs maximum photosynthetic rates, but a trend that is expected to gradually decline in more weathered soils, due to a lower P availability (Maire et al. 2015). Phosphorus limitation can become sufficiently intense in the late stages of ecosystem development (also known as the retrogressive phase) to cause a decline in forest biomass, and productivity (Wardle et al. 2004). The exception seems to be tropical forests (Turner et al. 2007), which exhibit very diverse tree communities on old, infertile soils (Losos and Leigh 2004). Moreover, Turner et al. (2018) showed that in lowland tropical ecosystems, P limitation affects individual species, but species-specific P limitation does not translate into a community-wide response, because some species grow rapidly on infertile soils despite extremely low P availability.

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4.3 Future research priorities

Many efforts have been made to explain soil P composition during pedogenesis; however, a clear picture on how specific plant species, plant functional traits, and their communities can influence the soil P composition is still lacking, especially with results obtained with ^{31}P NMR. For example, why inositol hexakisphosphates have not been found in tropical soils under native vegetation, i.e., is it because the rapid turnover promoted by plants and, or organisms (which one?), or exclusively due to lack of inputs from plants? Does the changes in forest biomass and plant species diversity as soil P turns scarcer contribute to soil P composition in non-tropical environments, either by P inputs or differing P compounds uptake, or the soil *per se* governs both the soil P composition and vegetation dynamics? Therefore, we point out that studies aiming to disentangle confounding effects among soil biotic and abiotic components, climate and vegetation are required to enable a better understanding of soil P composition in natural ecosystems.

Moreover, studies coupling ^{31}P NMR with other important techniques could contribute to a better understanding of P cycling and composition in terrestrial ecosystems. A clear understanding of how orthophosphate species such as Fe-, Al- and Ca-phosphate (Hesterberg 2010; Kizewski et al. 2011) respond to pedogenesis could be elucidated with XANES (see Prietzel et al. 2013; Hashimoto and Watanabe 2014). Studies that quantify specific-P-related enzymes activity (see Turner et al. 2018) in native vegetation soils could help understand if the hierarchy of investment for the P acquisition actually contributes to different degrees of accumulation of inositol hexakisphosphates and DNA as pedogenesis progresses in non-tropical environments, and if phosphatases are leading to a rapid turnover of inositol hexakisphosphates in tropical environments. This can be achieved through determining the presence and abundance of microorganisms and enzymes, and how these changes affect soil P composition. Turnover, exchange kinetics, mineralization rates could be assessed using isotopic techniques (see Frossard et al. 2011), and enable a separation between different sources of P compounds, and their dynamics in soils, organisms and plants.

Finally, we expect future research to provide results of as many soil P compoundscompounds as they can find rather than functional groups only (i.e., diesters and monoesters), even when compoundscompounds concentrations are low (and describe when main soil compoundscompounds are not detected), which may enable future analyses to avoid possible confounding effects of P compounds inside functional groups (e.g., inositol hexakisphosphates 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 analyses could use the different information provided by studies of different scopes and quality in a meta-analytical approach.

5 Conclusion

We conclude that edaphic and climatic properties are important factors in determining soil inorganic and organic pools as well as their compounds, since they regulate key ecological processes governing their presence, transformation and

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persistence in soils. These processes are related to the source of P inputs, primarily determined by parent material and then altogether through plant and microbe P cyclingcycling, 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 was the climatic variables that most influenced soil P composition. Soil P composition was more influenced by soil variables than either climatic variables or weathering stages. However, combined effects among these factors also contributed to explain considerable soil P variability in these ecosystems. Soil organic and inorganic P compounds responded differently to combinations of environmental drivers, which likely indicates that each P compound has specific factors governing its existence in natural ecosystems. Therefore, knowing how environmental drivers affect soil P composition enabled a comprehensive understanding of soil P innatural 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 S3 – 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 S9 – Soil weathering stages and poorly crystalline Al and Fe concentration.

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.

7 ACKNOWLEDGEMENTS

The authors gratefully acknowledge the Coordination for the Improvement of Higher Education Personnel (CAPES-Brazil) and the National Council for Scientific and Technological Development (CNPq-Brazil) for funding this research. Maire is funded by the NSERC-Discover (2016-05716) grant. We sincerely thank the anonymous reviewers for the comprehensive evaluations of our manuscript. We also acknowledge the effort made by all the authors from which studies we compiled our results. Our study would not be materialized without their work. We truly acknowledge the great effort made by Jordon Wade for his insights and language review.

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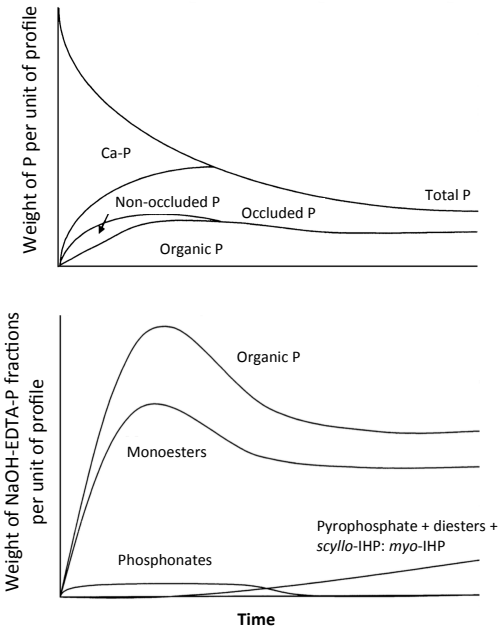

Figure 1: Conceptual diagrams of the changes in soil P fractions (above) and NaOH-EDTA extractable P compounds (excluding orthophosphate) (below) with time (redrawn from Walker and Syers, 1976, with permission from Elsevier, and McDowell et al. 2007, with permission from John Wiley and Sons, respectively).

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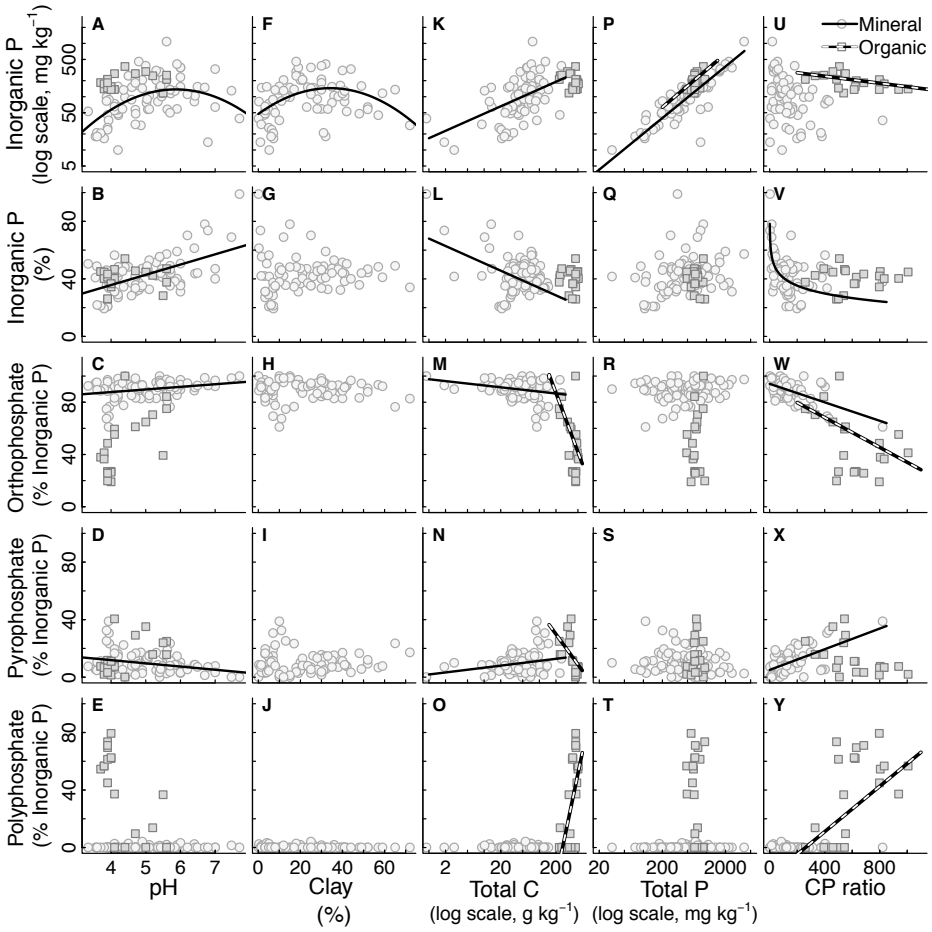

Figure 2: Relationship between edaphic properties and soil inorganic phosphorus (P) composition in NaOH-EDTA extract from soil mineral and organic layers on natural ecosystems. Note that the reported total P is the one obtained by digestion and usually comprise the residual P non-recovered by the NaOH-EDTA 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

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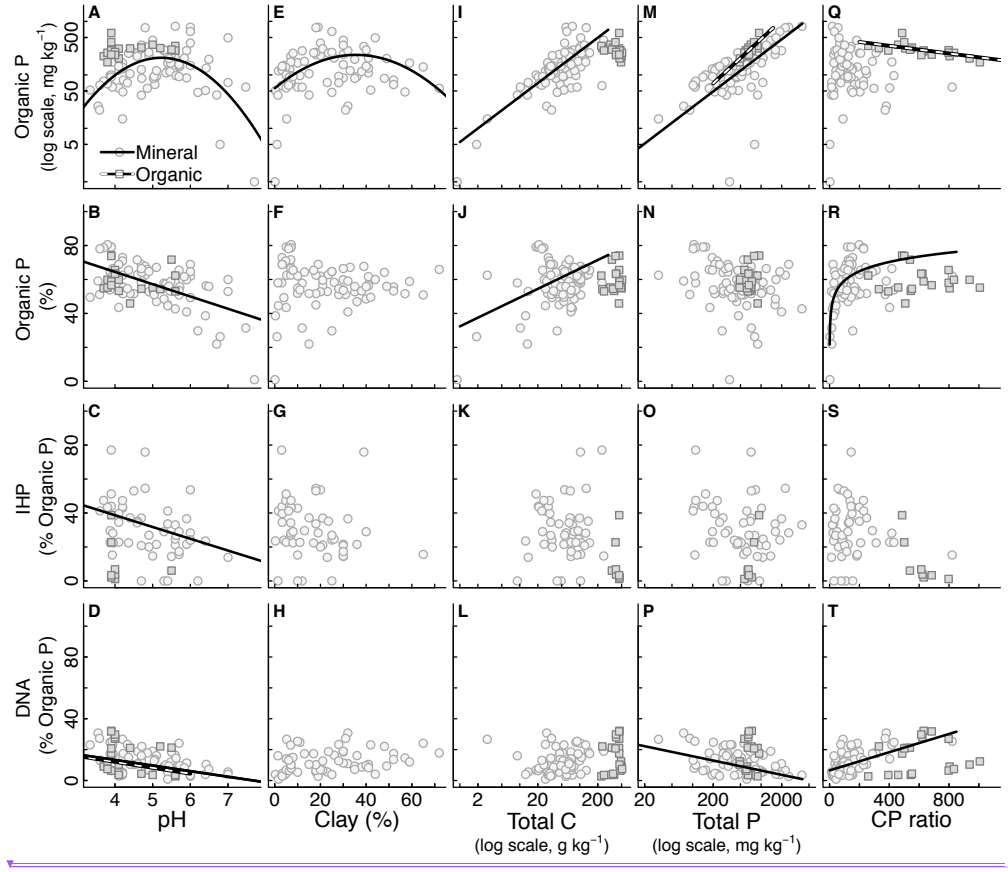

Figure 3: Relationship between edaphic properties and soil organic phosphorus (P) composition in NaOH-EDTA extract from soil mineral and organic layers on natural ecosystems. Note that the reported total P (x axis) is the one obtained by digestion and usually comprise the residual P non-recovered by 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 hexakisphosphate (IHP) =66.4 - 6.94 pH, r^2 = 0.16 (n=52); mineral layer, DNA = 27.8 – 3.63 pH, r^2 = 0.19 (n=64); organic layer, DNA = 27.4 – 3.83 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); mineral layer, 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); mineral layer, DNA = 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); mineral layer, DNA = 6.53 + 0.029 CP ratio, r^2 = 0.34 (n=64).

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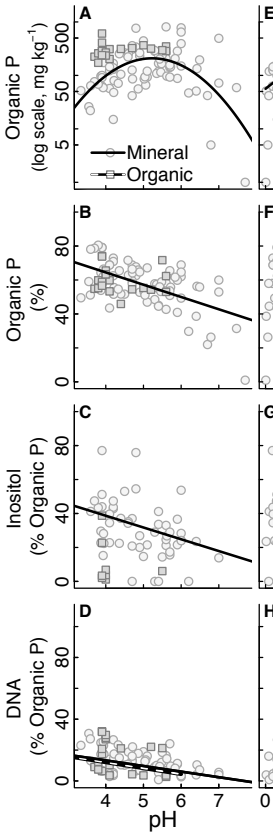

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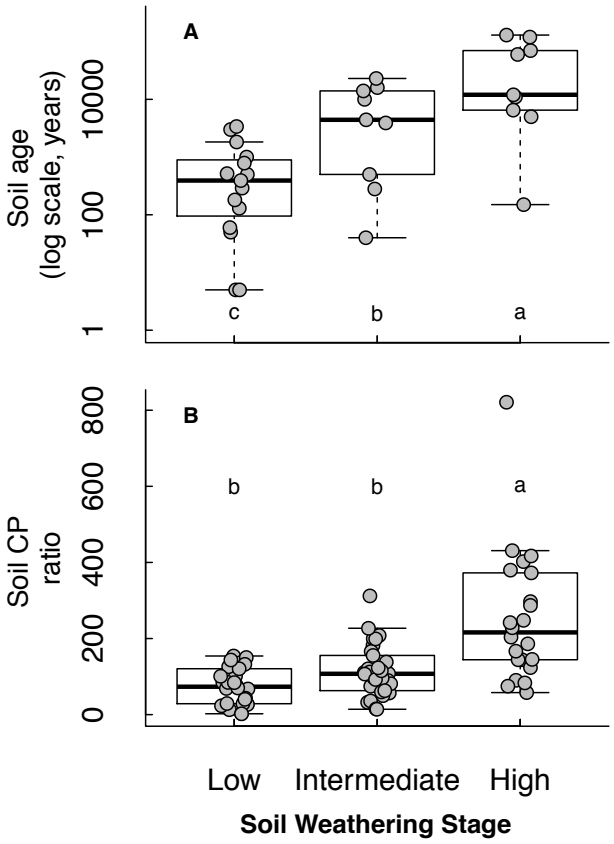

Figure 4: Soil weathering stage relationship with soil age (n = 33) and CP ratio (n = 78) on natural ecosystems.

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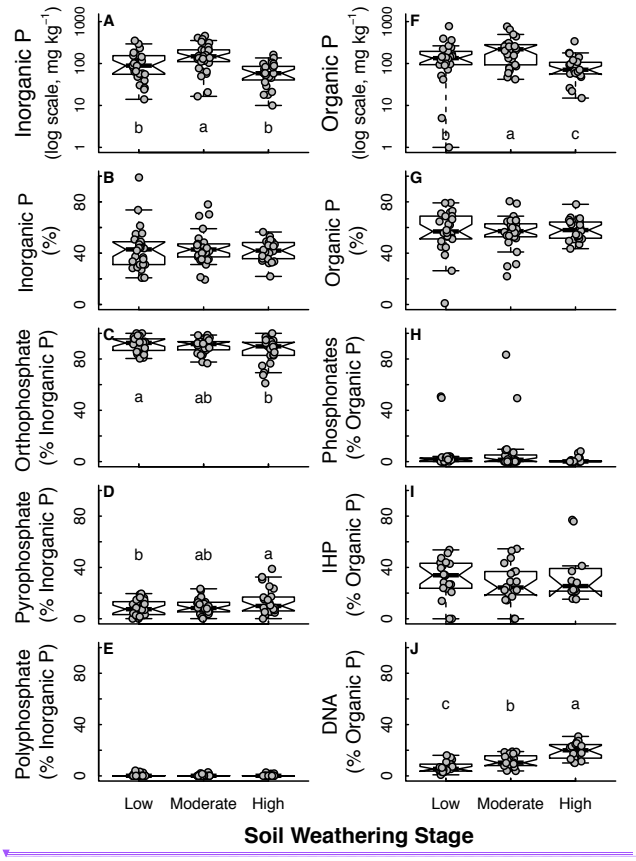

Figure 5: Soil inorganic and organic phosphorus (P) composition in NaOH-EDTA extract as influenced by weathering stages on natural ecosystems.

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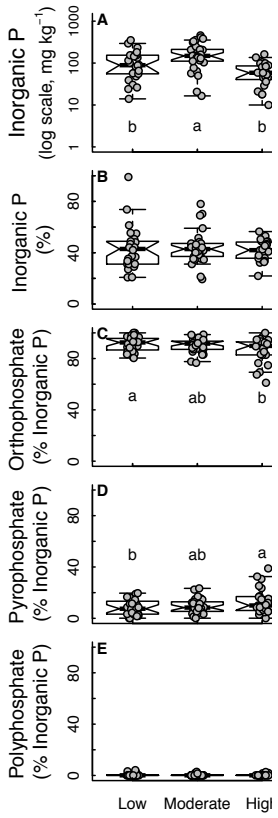

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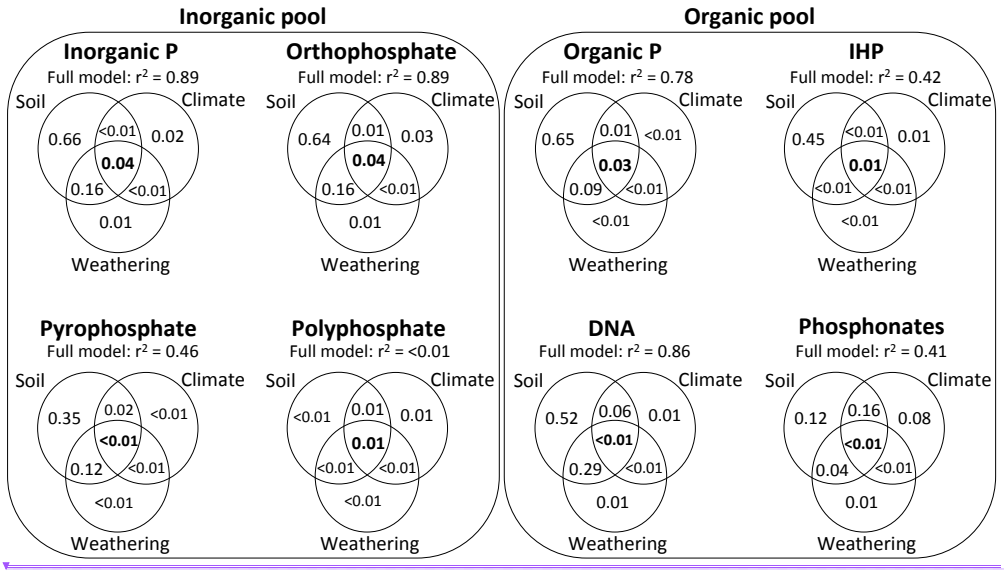
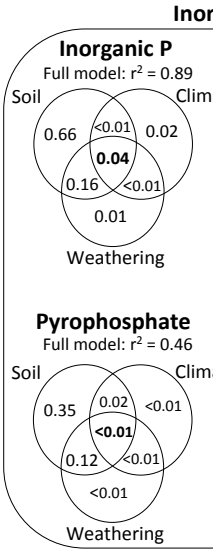


Figure 6: Variation partitioning among edaphic, climatic, and weathering stages on soil inorganic and organic P composition in NaOH-EDTA extract on natural ecosystems. Soil organic and inorganic P forms and compounds were in mg kg^{-1}, and the other variables followed units described on Figures 2 and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



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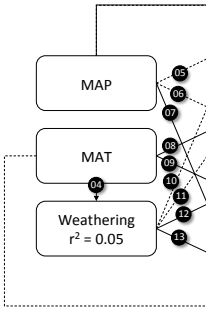

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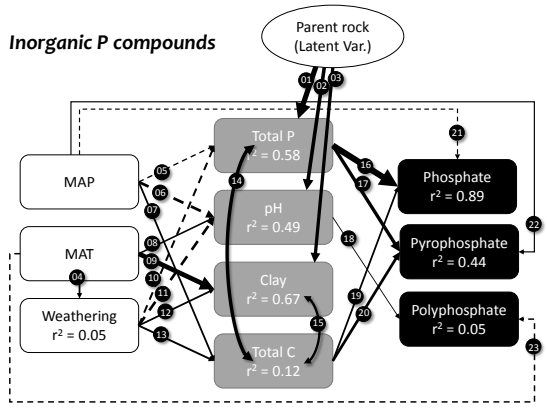

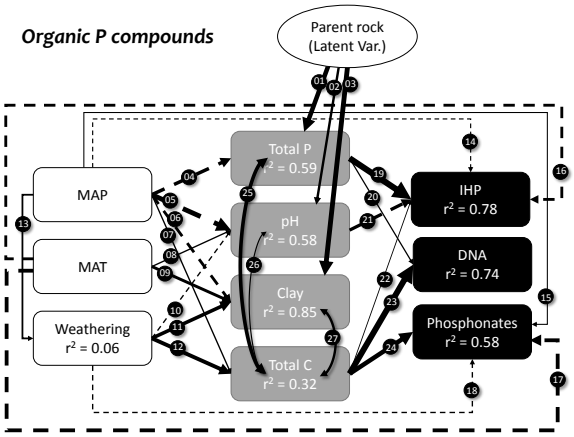

Figure 7: Path analysis describing the direct and indirect effects of the main environmental predictors of soil inorganic and organic P compounds (mg kg^{-1}) in NaOH-EDTA extract as influenced by edaphic and climatic drivers on natural ecosystems. Solid and dashed lines represent positive and negative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

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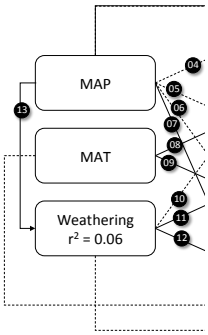

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Phosphomonoesterase is more active in acidic soils while phosphodiesterase is optimized in basic soils (Turner and Haygarth 2005).

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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 extractions.

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Most importantly, we need to investigate the hierarchical nature of causal effects between state factors, soil weathering, soil properties and Po and Pi composition.

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SomeHowever, there are drawbacks of using NaOH-EDTA extractant for ^{31}P NMR analysis. exists such as that NaOH-EDTA usuallyNaOH-EDTA does not extract all soil P and the highly alkaline environment, the extractant can potentially degrade some P compounds compounds due to the highly alkaline environment (Cade-Menun et al. 2006; Cade-Menun and Liu 2014). Additionally, the NaOH-EDTA extraction, and it does not separate Fe-, Al- and Ca-phosphate compounds (Kizewski et al. 2011).

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Some drawbacks of ^{31}P NMR exists such as that NaOH-EDTA usually does not extract all soil P, the extractant can potentially degrade some P compounds due to the highly alkaline environment (Cade-Menun et al. 2006; Cade-Menun and Liu 2014), and it does not separate Fe-, Al- and Ca-phosphate compounds (Kizewski et al. 2011).

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different state factors of ecosystem functioning because known responses were obtained from case-specific conditions.

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could help better understand how soil P dynamicscomposition is influenced by environmental properties. Therefore, w

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

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because of the potential biogeographical factors influencing soil P composition once knowing that soil, climate, fauna and flora can modify fluxes of soil P dynamics compounds

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because of the potential biogeographical factors influencing soil P composition once knowing that soil, climate, fauna and flora can modify fluxes of soil P dynamics compounds

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

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how the soil inorganic and organic P compounds variation

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Appendix

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Appendix

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For that, we used values as percentage.

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inositol phosphate

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inositol phosphate

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-EDTA P) increased (Figure 3B) as the pH decreased (from right to left), and

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and to a lesser degree, but not less importantly, by

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differently to these groups of factors

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Plant 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).

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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 proportions, 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.

Moreover, the 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).

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Our findings of pH influence on DNA (Figure 3D) are in keeping with Turner and Engelbrecht's observations (2011) for tropical forest soils, and with Turner and Blackwell's observations (2013) for temperate arable soils, where the most acidic soils contained an increasing proportion of P_o as diesters (mostly DNA).

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Our findings of pH influence on DNA (Figure 3D) are in keeping with Turner and Engelbrecht's observations (2011) for tropical forest soils, and with Turner and Blackwell's observations (2013) for temperate arable soils, where the most acidic soils contained an increasing proportion of P_o as diesters (mostly DNA).

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inositol phosphates

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inositol phosphates

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; however, some authors described that inositol phosphates declined to lower concentrations in older soils (Turner et al. 2014; Turner et al. 2007).

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In our study, we found that there was an increasing proportion of inositol hexakisphosphates and DNA in the P_o pool (% of NaOH-EDTA P) as pH decreased, and there was predominance of inositol hexakisphosphates in acidic, more weathered soils (Figures 3C-D, 5J).

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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, 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.

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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 these 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).

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Other P compounds such as DNA (Figure 3D) and pyrophosphate (Figure 2D) will prevail in more weathered systems.

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NaOH EDTA

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NaOH EDTA

Appendices - Soil phosphorus dynamics on terrestrial natural ecosystems

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 S3 – Soil depth effect on soil P composition (random factor).
Appendix S4 – Latitude effect on soil P composition (random factor).
Appendix S5 – Percentage of P extracted with NaOH EDTA effect on soil P composition (random factor).
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 S9 – Soil weathering stages and poorly crystalline Al and Fe concentration.
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.

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

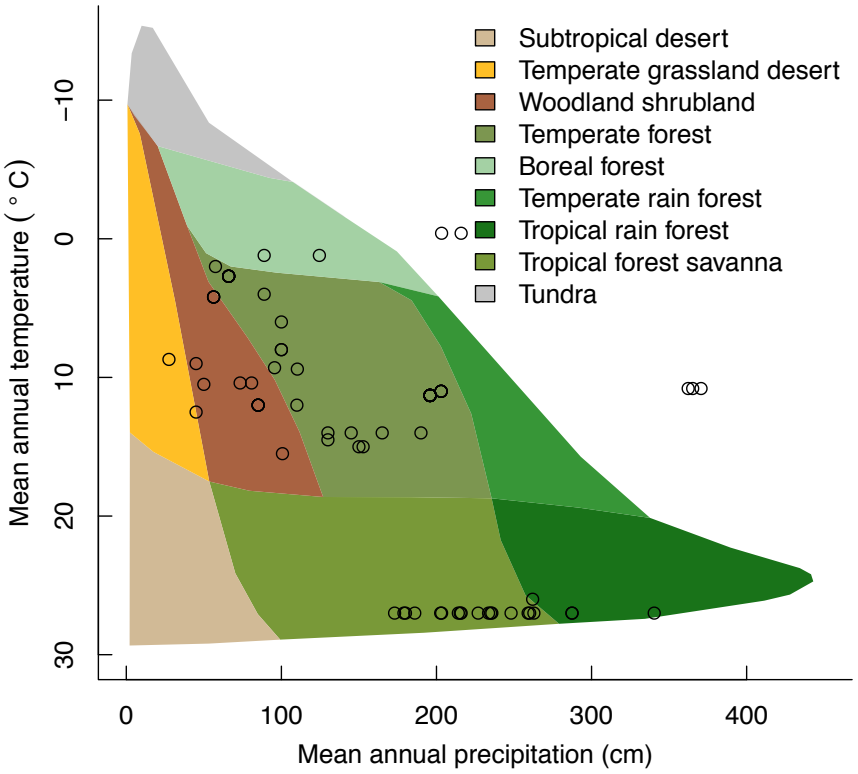

Figure S2. The Whittaker' diagram used to determine the main biomes comprised in our dataset.

Appendix S3 – Soil depth effect on soil P composition

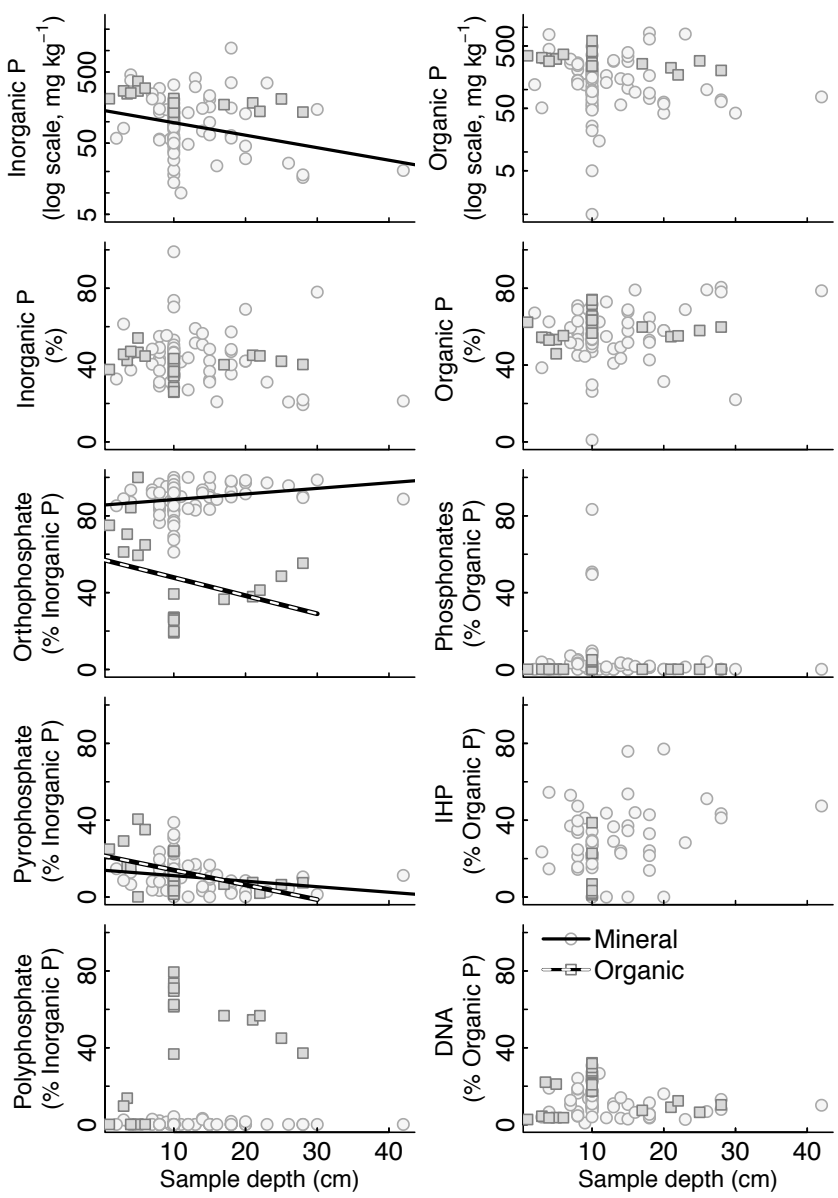

Figure S3. Soil inorganic and organic phosphorus (P) composition in NaOH EDTA extract as influenced by soil sampling depth on terrestrial natural ecosystems. Significant relationships are indicated with regression lines.

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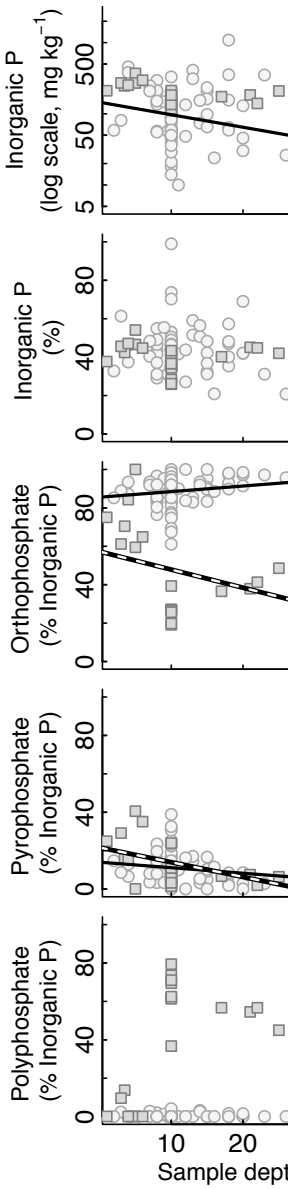

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Appendix S4 – Latitude effect on soil P composition

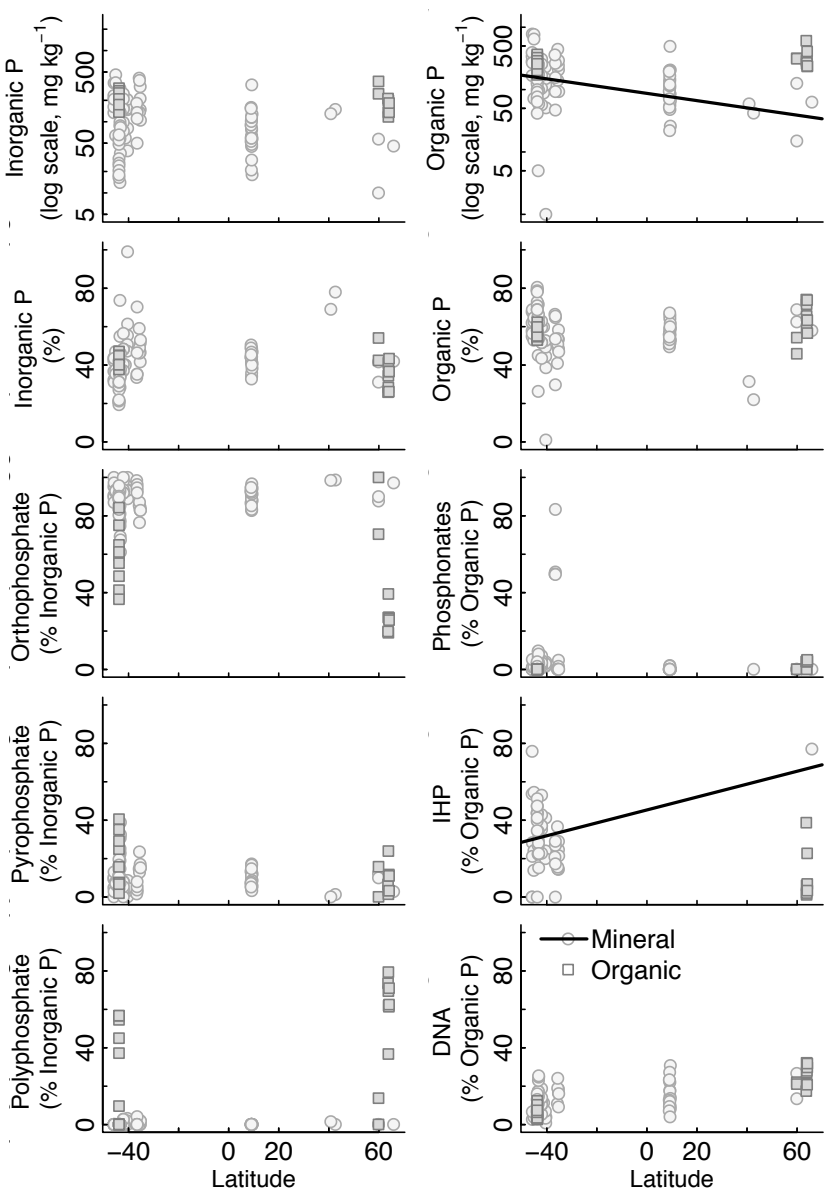

Figure S4. Soil inorganic and organic phosphorus (P) composition in NaOH EDTA extract as influenced by latitude on terrestrial natural ecosystems. Significant relationships are indicated with regression lines.

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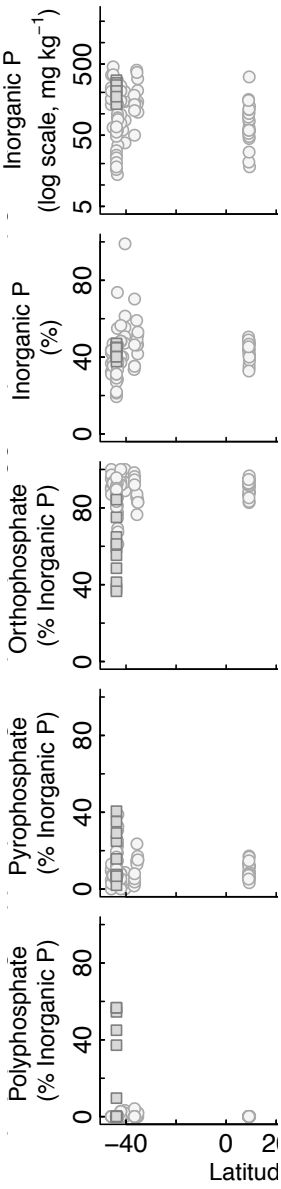

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Appendix S5 – Percentage of P extracted with NaOH EDTA effect on soil P composition

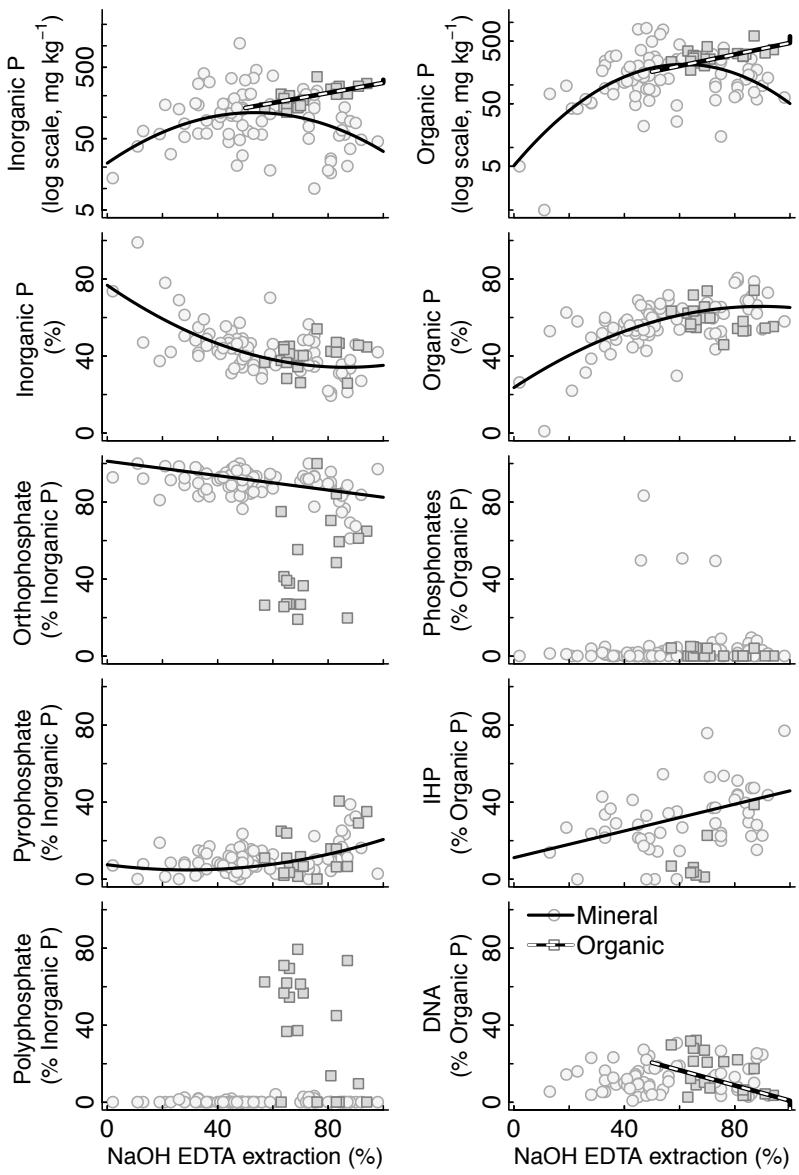

Figure S5. Soil inorganic and organic phosphorus (P) composition in NaOH EDTA extract as influenced by soil sampling depth on terrestrial natural ecosystems. Significant relationships are indicated with regression lines.

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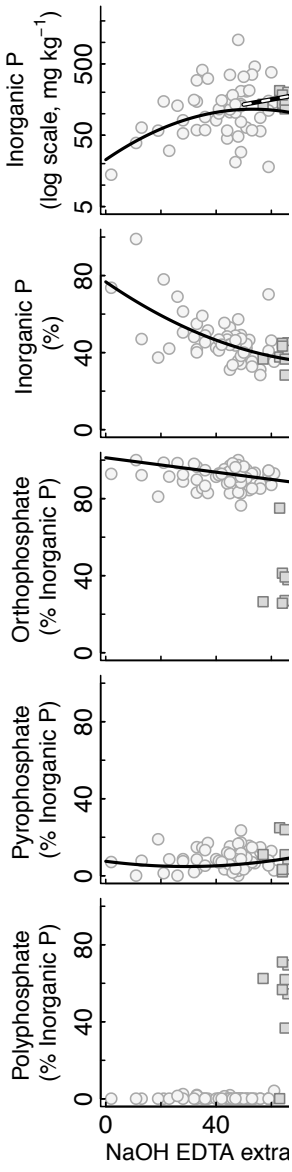

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Appendix S6 – Soil properties and soil organic phosphonates

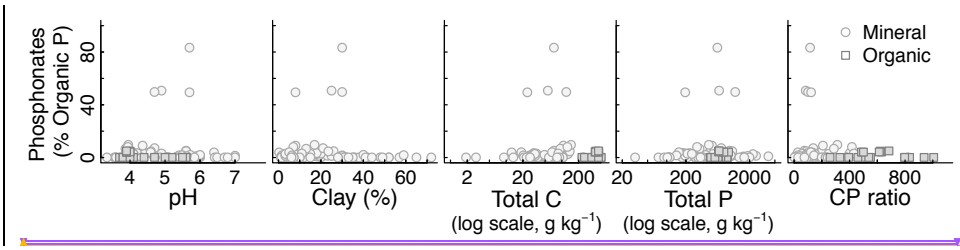

Figure S6. Relationship between edaphic properties and soil organic phosphonates in NaOH EDTA extract from soil mineral and organic layers on terrestrial natural ecosystems. No significant relationships were found.

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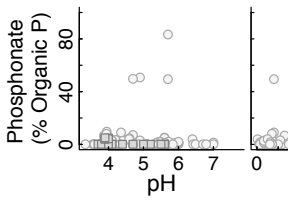

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Appendix S7 – Climatic properties and soil inorganic phosphorus

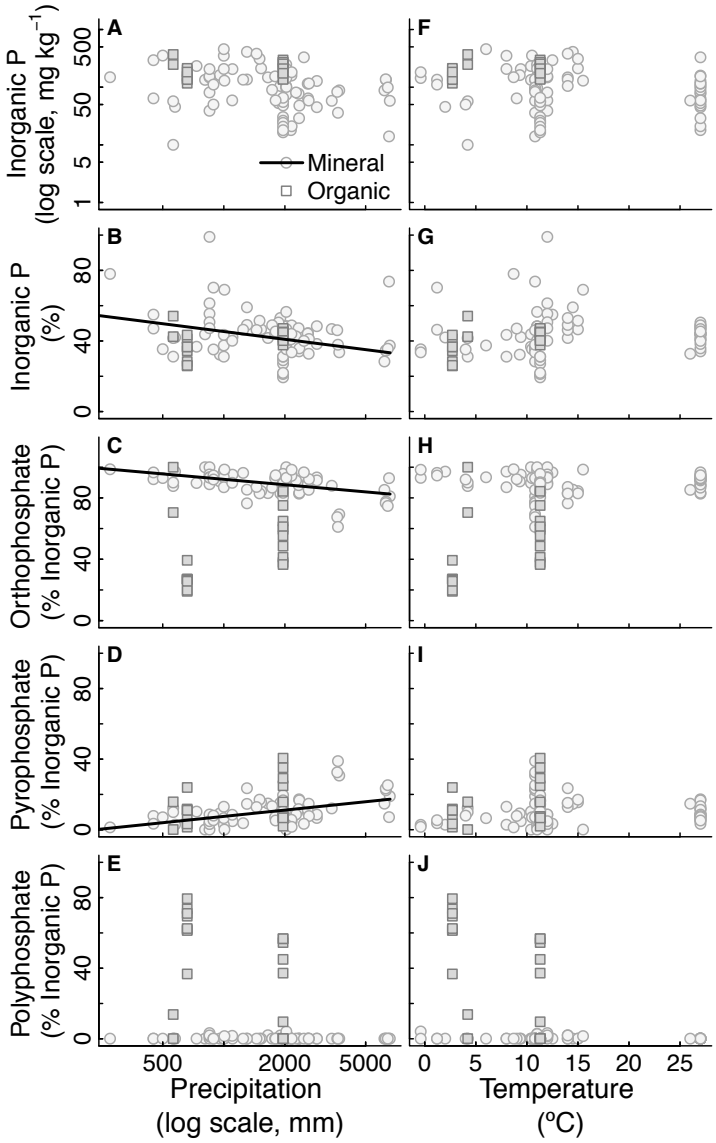

Figure S7. Relationship between climatic properties and soil inorganic phosphorus (P) composition in NaOH EDTA extract from soil mineral and organic layers on terrestrial natural ecosystems. Regression models (n = 80 mineral layer and n = 20 mineral layer): mineral layer, total P_i (%) = 89.4 – 14.7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

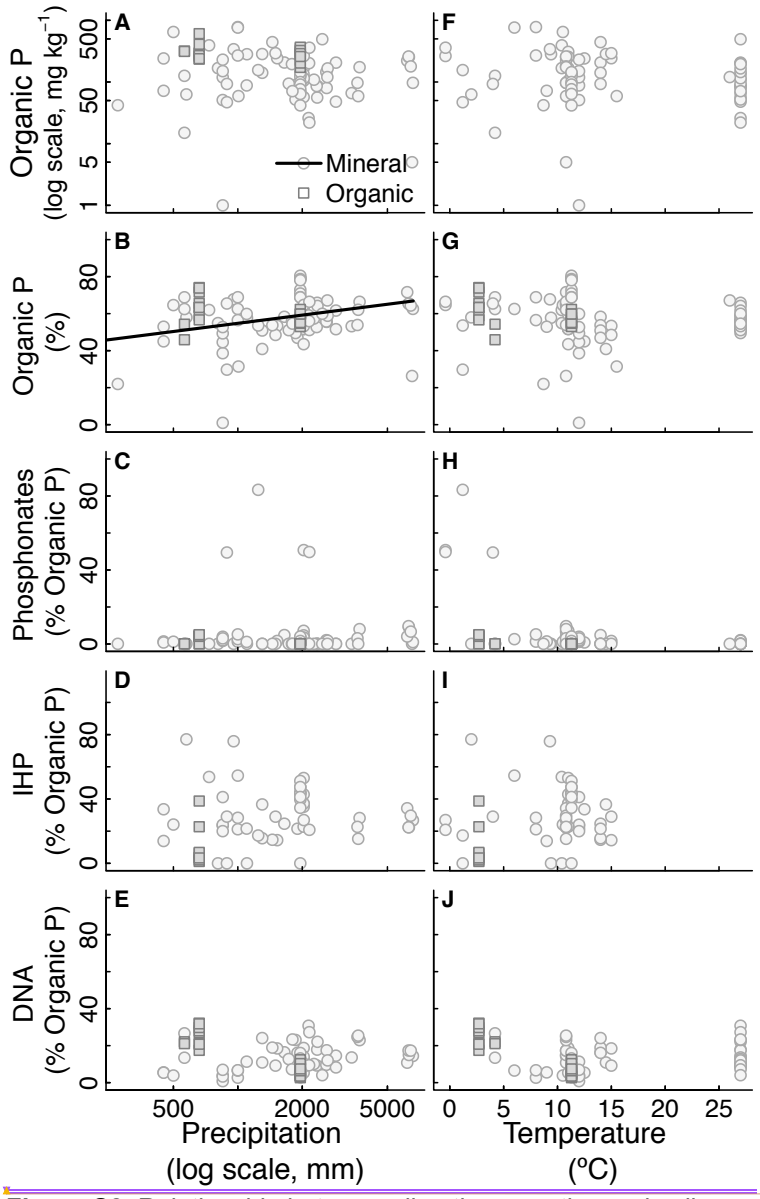

Figure S8. Relationship between climatic properties and soil organic phosphorus (P) composition in NaOH EDTA extract from soil mineral and organic layers on terrestrial natural ecosystems. Regression models: mineral layer, total P_o (%) = 10.6 + 14.7 log(precipitation), r^2 = 0.08 (n=80).

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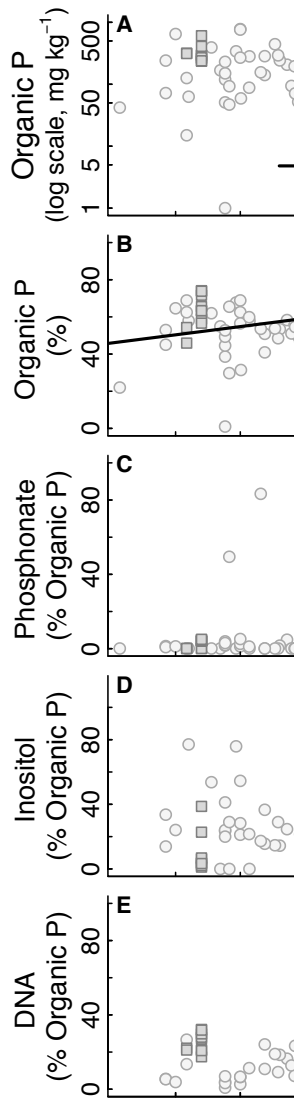

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Appendix S9 – Soil weathering stages and poorly crystalline Al and Fe concentration

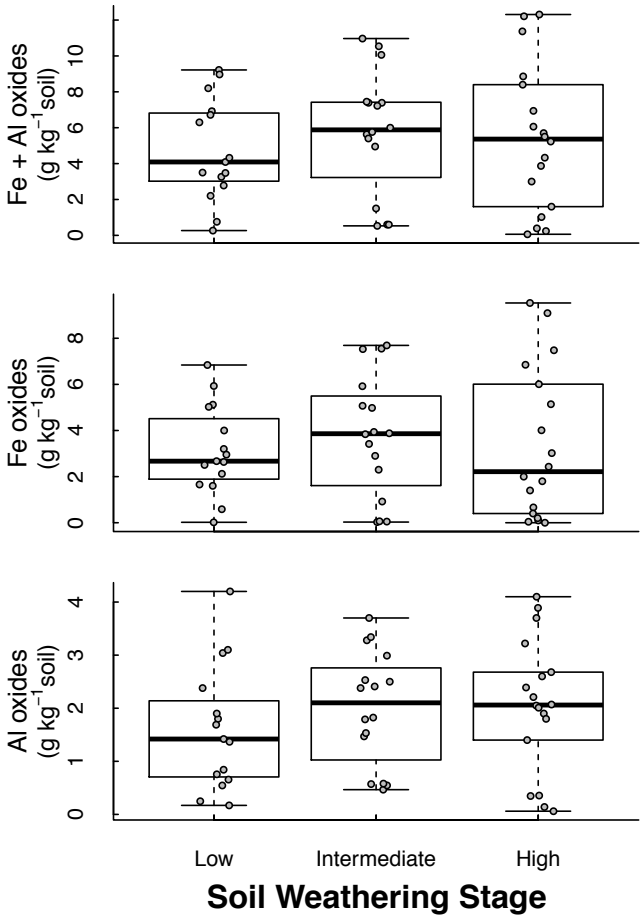

Figure S9. Soil weathering stage relationship with soil poorly crystalline Al and Fe (n = 49) on terrestrial natural ecosystems. For all three panels p>0.1.

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 S.10.1 and S10.2, top panels).

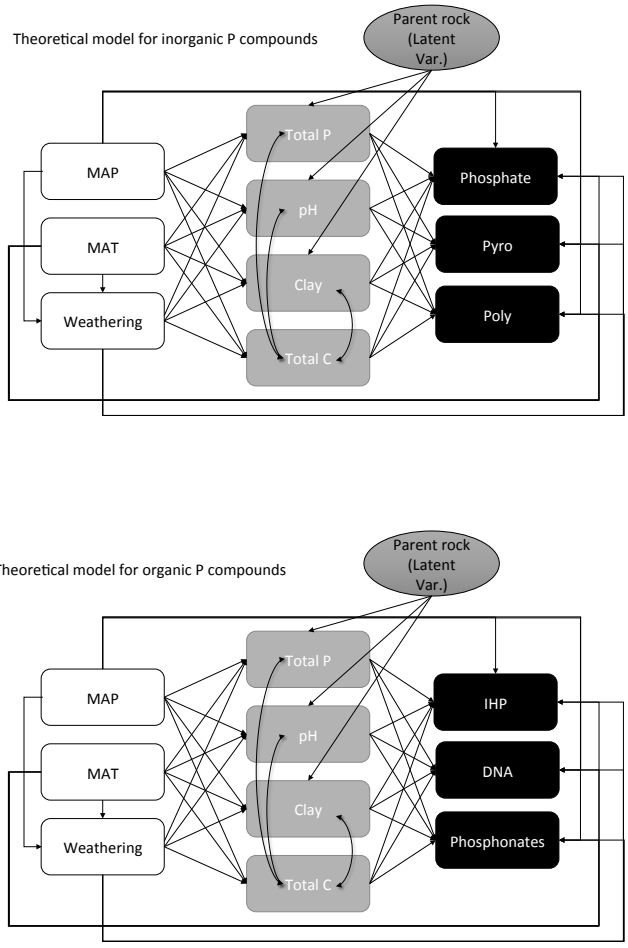

Figure S10.1. Theoretical models set to explore the interdependences between edaphic and climatic variables (path analysis) as the main environmental predictors of soil inorganic (upper panel) and organic (bottom panel) P compounds.

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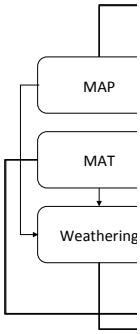
Theoretical model for



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