RC2.1 Petrash et al. present an interesting study on the spatial heterogeneity of soil solution in a central European high-elevation catchment which formerly received high loads of atmospheric pollutants. The topic is highly relevant for SOIL and the data gathered for the study are considered worth being published. However, with respect to the structure of the manuscript as well as the presentation of the methods and the data the manuscript seems to need major revisions.

We thank the constructive criticism of reviewer 2, Prof. Dr. Meesenburg, and have made every attempt to address each of his concerns and relevant suggestions for improvement.

RC2.2 The introduction reflects the history of atmospheric pollution in the "Black Triangle" The last paragraph of the introduction needs to be completely rewritten as it contains little information concerning the design of the study, but methodological issues, results and even concluding statements. No objectives neither hypotheses are given in the introduction. Please amend accordingly.

To address this concern of the reviewer, the introduction section has been significantly rewritten and expanded to now provide a brief review of background information and relevance, objectives and hypotheses of the study.

RC2.4 Otherwise, assessments are mentioned (i.e. soil moisture determination), which value for the study remains unclear.

Unused information such as unrelated, soil moisture measurements has been now removed from the revised methods.

RC2.5 According to Figure 1, sampling plots for soil solution and solid soil are up to more than 200 m apart. However, no information is given with respect to the comparability of the respective plots. Please give a rationale for this approach as the results from either solid soil or soil solution are related to the respective slope position.

The revised version of the MS page 5, rows 7-29 now clarifies this aspect: *"Five 0.5 m2 soil pits were excavated in July 2015 at some distance to the previously installed suction lysimeter nests to avoid disturbances to the zero tension soil solution collection systems (Fig. 1b) while preserving a soil profile equivalent to the one at the nearby nest and also the relative position within the catchment area."*

RC2.6 The contents of the results section are structured differently as the methods section.

The revised results section of our MS has been re-structured to address this flaw of the original submission kindly pointed out by the reviewer.

RC2.7 Some parameters, which are displayed in the tables and figures aren't mentioned in the text body of the results section.

Conductivity parameter has been removed from Table 2. The rest of the parameters listed are either mentioned/discussed in the main text, or needed for calculations (*e.g.,* $Al_{Ox,}$ Fe_{Ox})

RC2.8 For soil solution, different units are reported in the text and in table 2.

Now all in-text mentions to soil solution concentration are referred to Table 2, where concentrations units are expressed in ppb.

RC2.9 The last part of the results (i.e. page 8, row 6-11) seems to be more suitable for the discussion.

The misplaced text kindly highlighted by the reviewer was moved and integrated to the discussion section (page 12, rows 13-16).

RC2.10 The integration of the assessment of P availability into the study appears a little bit weak as no relation to either soil solution or runoff P concentrations is given. Also, a discussion on the role of P

availability for tree nutrition is missing. A convincing evidence for the “de-coupling” of P availability and organic carbon is missing. At least, an explanation should be given, why a “coupling” of P availability and organic carbon should be expected.

There is no further reference in the revised text to coupled/decoupled cycling of nutrients, but to the role of P in belowground C allocation and base cation imbalance. With regard to our P measurements, the revised text (page 12, rows 21-34) now reads: *“Soil P sorption saturation is often used as an environmental indicator of soil P availability to runoff. Phosphorus losses from soils not subjected to an augmented erosional process are generally small (see Heuck and Spohn, 2016). Our calculation of P sorbed by the soil particles, as determined by oxalate extraction (after Borovec and Jan, 2018), shows that between 22 and 29 mg of P per kg of soil was sorbed in the in the 40-80 cm depth at the time of sampling, with insignificant difference between hilltops, slopes and valley. It is possible that P limitation has developed because of the legacy of anthropogenic N deposition in this region. The homogenous pattern of low P availability contrasts with elevational differences in DOC concentrations in soil solutions (Fig. S1), which points to variable belowground leaching and allocation of C or could be reflective of variable inputs of C from regenerating vegetation in the N-saturated, P-limited forest ecosystem. Conifer tree species are generally more tolerant to P limitations, which in turn make them more susceptible to nutrient depletion following losses from harvesting and exacerbated rates of nutrient export (Hume et al., 2018). We attribute the variable belowground allocation of C in UDL to spatially variable Mg2+ and/or K+ deficiencies (e.g., Rosenstock et al., 2016) rather than to P imbalances within the catchment since we detected no spatially contrasting P deficiencies exerting influence over the contrasting patterns of nutrient limitation and subsoil nutrients leaching across the studied forest landscape.”*

RC2.11 The conclusion contains issues, that haven’t been discussed before (e.g. the role of drought and torrential rains, isotope investigations). This should be avoided.

The flaws on the original conclusions kindly pointed out by the reviewer are now avoided and the conclusion section have been streamlined, with no reference to undiscussed aspects.

RC2.12 The last three points of the conclusions resembles a collection of keywords more than elaborated findings.

The last three points indicated by the reviewer were removed from the text, and the section rewritten as follows:

The hydrochemical comparisons implemented here were aimed at evaluating spatial and temporal concentration patterns on the water chemistry among the subsoil compartment of the critical zone in temperate forest. Because of landscape and lithological simplicity, which facilitates discerning flow paths without variability effects introduced by differential bedrock weathering, it was possible discussing what factors in association to soil N-saturation affect the soil solution chemistries of a small mountainous catchment area reforested by Norway spruce after acidification-related die-back. By combining soil solution chemical measurements and establishing comparisons with published hydrochemical data this work provides evidence pointing to substrate variability, C but not P bioavailability as major controls over the flux of base metal leached into the subsoil level and across the elevation gradient. Soil solutions at the 50 cm depth were generally more diluted than stream waters due to lateral surface runoff of solutions originating in the litter and humus, enriched in SO42-, NO3-, K+, Na+, Ca2+ and Mg2+. Increased concentrations are linked to anthropogenic atmosphere-derived pollution affecting natural (bio)geochemical processes. Differences between chemistry of soil solution and runoff could have been also caused by a direct contribution of throughfall, which scavenged atmospheric chemicals of the canopy and leached nutrients from inside the foliage, or by polluted open-area precipitation. Soil solutions had lower pH in the valley than at upslope locations, being more diluted in the valley than on hilltops in the case of DOC, Ca2+ and Mg2+. Both NO3- and SO42- in soil solutions exhibited a clear seasonality that can affect base metal leaching, with maximum concentrations in the growing and dormant season, respectively.

The observed temporal trends amongst strong anion inputs and leaching of base metals and acid anions reflect that at the time of sampling nutrient imbalances in UDL were linked to groundwater carrying legacy pollutants. A complementary isotope modelling show that the responses of the studied mountain catchment to precipitation are fast, i.e., within the monthly sampling interval, with direct precipitation contributing 20 to 40% of the discharge, and the rest being the contribution of local groundwater. When evaluated with regard to stream water chemistries, and previously published input and fluxes data, this study provide insights into the localized controls and effects of acidification disturbances at a catchment-scale and offer a perspective of the spatially and temporarily variable nutrient concentrations in soil solutions that is relevant for more effectively designing stream water chemical analyses aimed at understanding the coupling of soil development processes and hydrology over variable time scales, and between deep and shallow weathering processes in mountain catchments and for evaluating soil recovery processes after atmospherically induced perturbations that affected the UDL catchment..

RC2.13 The chapter on $^{18}O/^{16}O$ modelling in the Appendix seems not very well integrated in the study.

We added the following lines to the rewritten introduction to address this concern of the reviewer (page 3, lines 16-17): *"In addition, the contribution of groundwater vs. runoff infiltration is further evaluated by mean of a supplementary isotopic runoff model, which provides evidence for a likely contribution of groundwater enriched in selected chemical species due to sufficiently long water-saprolite interactions."*

Also the revised discussion text (page 12, lines 1-6) now reads: *"On this note, by following the modeling approach implemented by Buzek et al. (1995, 2009), we determined that the mean residence time of soil solutions—calculated across all sampling locations—is approximately 8.3 months (Appendix A), which indicates that the volume of the entire mobile water at UDL is larger than the volume of soil solution transported the 50 cm subsoil levels and below (see Appendix A for details). In consequence, the diluted runoff water at UDL is a mixture of direct precipitation with older soil solution containing and even older shallow groundwater containing dissolved saprolite' weathering products. The supplementary isotopic modelling implemented here also shows that direct precipitation contributes between 20 and 40% of the discharge, with the rest being local soil pore and ground waters (Appendix A). The combination of all these three water types is called "mobile water", defined as the sum of all water pools and fluxes that respond to changing precipitation amounts. This effect is probably linked to transient increased soil solution saturation and concomitant increase in the hydrologic connectivity of soil pore waters to the stream, with a heterogeneous distribution of dissolved ions in soil solutions at the catchment-scale (Basu et al., 2010).*

RC2.14 The references are mostly relevant for the study and up-to-date. However, some citations aren't very specific to the referenced issues.

The reference have been revised as per reviewer indications (see below under Specific comments) .

RC2.15 The publication of the manuscript can only be recommend after a major revision considering the above mentioned concerns

We thank the reviewer for providing generous indications, guidance and sound revisions/ suggestions that significantly improved the original submission.

R2. Specific comments

RC2.16 page 2, row 1 "Blazkova et al., 1996" -> "Blazkova, 1996"

Done

RC2.17 page 2, row 3 "Blazkova et al., 1996" -> "Blazkova, 1996"

Done

RC2.18 page 2, row 6 “Fen et al. 1998” doesn’t appear in the references. Is “Fenn et al. 1998” meant? If so, this citation doesn’t support the before stated sentence, because it mainly reflects the situation in North America.

Done.

RC2.19 page 2, row 6 “Hruska et al. 2003” -> “Hruska and Kram, 2003”

Done.

RC2.20 page 2, row 8 “Gradowski et al., 2008” -> “Gradowski and Thomas, 2008”

Done.

RC2.21 page 2, row 9 “Matschullat et al., 1998” -> “Matschullat et al., 1992”. This citation doesn’t give relevant information on the bioavailability of phosphorus. Please replace accordingly.

Reference removed as per reviewer indication.

RC2.22 page 2, row 10-11 This sentence isn‘t relevant for the introduction. It may be shifted to section 2.1.

Text was removed as part of the editions implemented to the Introduction.

RC2.23 page 2, row 10-18 “For this aim, a nests of suction lysimeters was installed …” -> “For this aim, nests of suction lysimeters were installed …”

Done

RC2.24 page 2, row 30 Would this kind of orthogneiss with the reported composition really be termed “alkaline”?

Word “alkaline” removed. The term apply to granite protolith over the base of the observed gneiss composition.

RC2.25 page 3, row 1 “… m, UDL’s …” -> “… m a.s.l., UDL’s …”

Done

RC2.26 page 3, row 2 At page 2, row 30, the parent material was named “alkaline gneiss”. Is this the same as “porphyric granite”? Please clarify!

The first is the metamorphic product of the latter. We thank the reviewer for noticing the lack of clarity of the text. This has been now clarified (page 4, row 1-2): *“Low base status soils have developed at expense of the mineralogy of the porphyritic granite that served as protolith to the orthogneiss bedrock […]”*

RC2.27 How can Podzols develop on “alkaline gneisses”?

The unclear statement was corrected as per note above. In igneous petrology, the composition of the igneous-metamorphic bedrock falls within alkaline granite in a compositional diagram.

RC2.28 page 3, row 4 Are the given figures the runoff for one month? Please clarify!

We have clarified the text as per reviewer request (page 4, row 6): *“ […] the monthly highest stream water runoff flow is usually recorded (~162 ± 29 mm).”*

RC2.29 page 3, row 9 I wouldn’t consider saplings trees up to 40 yrs.

True. The revised text now refers these to as *“trees < 40 yrs”* (page 4, row 9)

RC2.30 page 3, row 12 Please delete “The.”

Deleted

RC2.31 page 3, row 12-13 Please give the depth interval for the pH figures.

The revised text now reads (page 7, row 23): *“The soil at the 40-80 cm depth was characterized by pHH2O < 5 (Table 2).”*

RC2.32 page 3, row 13 “… a pH increases in throughfall measurements …” -> “… a pH increase in throughfall …”.

The revised text now reads (page 4, row 25-26): *“[…] measured in water percolating through the canopy (i.e., throughfall) increased from 4.1 to 5.2 (Oulehle et al., 2017).”*

RC2.33 page 3, row 13-14 Two times the same citation in one sentence isn’t necessary.

The doubled citation referred by the reviewer is now removed from text.

RC2.34 page 3, row 14-15 A comparison of pH(H2O) and pH(KCl) doesn’t makes much sense.

Agreed. The offending text pointed out by the reviewer was removed.

RC2.35 page 3, row 15 “Hruska, 2000” -> “Hruska, pers. comm.” (unpublished data weren’t published in 2000).

Reference removed as per note above.

RC2.36 page 3, row 16-26 The content of this paragraph may be better placed in the results section. In addition figures presented should be given with standard deviation. Presented figures are a strange mixture of 13 or 14 catchments. Please revise consistently!

As suggested by the reviewer, a new section 2, titled *Study Site and Background Information*, now contains this information. There are 14 catchments in the Geomon network, this has been revised for consistency in page 4, line 32: “[...], which is far in excess of the atmospheric inputs observed in the remaining 13 GEOMON’s monitored catchments across the Czech Republic”.

RC2.37 page 3, row 27 Section “2.2.1” follows “2.1”. Please check for consistency.

Consistency of section and subsection numbering and general manuscript structure has been revised as per reviewer’s suggestion.

RC2.38 page 3, row 28 “… at a 50-cm depth below …” -> “… at 50 cm depth below …”. Please change consistently throughout the manuscript.

Done.

RC2.39 page 3, row 28 Were the lysimeters installed 50 cm below soil surface or below forest floor? Please clarify!

Done. The revised text now reads (page 5, row 6): *“were installed at 50 cm depth below soil surface”*

RC2.40 page 4, row 6 the sampled depth intervals aren’t horizons. Please specify!

Now these are referred to as soil levels (e.g., page 10, row 20; page 11, row 13).

RC2.41 page 4, row 8 the given citation (FAO 2006) isn’t about soil description. Please give correct citation!

FAO, 2006 removed from references.

RC2.42 page 4, row 19 Please specify the relation of soil to water for pH measurement.

Done. The revised text now reads (page 6, row 5-6): *"A Radiometer TTT-85 pH meter with a combination electrode was used to measure pH_{H2O} of soil (soil–water suspension ratio = 1 : 2.5)."*

RC2.43 page 4, row 19-20 Please specify the meaning of soil moisture determination for the study, if any.

Any reference to soil moisture in text is now removed.

RC2.44 page 4, row 22 "5 ° C" -> "5°C"

Done.

RC2.45 page 4, row 23-27 The sequence of the determination of exchangeable cations seems in an incorrect order. Where NO3 and SO4 determined in the BaCl2 extracts?

We thank the reviewer for kindly pointing out this flaw in our original methods description. The text has been revised as follow (page 6, row 9-11): *"[…] The concentrations of NO_3^- and SO_4^{2-} from the soil extracts described above were determined by ion chromatography (HPLC Knauer 1000), with limit of quantification (D.L.) of 0.1 and 0.3 mg L^{-1},[…]"*

RC2.46 page 4, row 28 Consider "For a phosphorus (P) release estimation, …" -> "For a estimation of phosphorus (P) availability, …"

Following the reviewer suggestion page 6, row 13 now reads: "For an estimation of phosphorus (P) release […]:

RC2.47 page 4, row 28-29 Please specify the soil:solution relation and the concentration of the solution.

Done. The revised text (page 6, row 13-18) now reads: *"[…], ammonium oxalate extractions were performed by following the protocol described in Schoumans (2000). In short, a reagent solution consisting of $(COONH_4)_2 \cdot H_2O$ and $(COOH)_2 \cdot 2H_2O$ was used to dissolve 1 g of the <2 mm soil fraction. Extractable phosphorus (P_{ox}), iron (Fe_{ox}), and aluminium (Al_{ox}) were determined by shaking for 2 h in the dark duplicate samples of soils with 30 mL of 0.5 M reagent in 50 mL centrifuge tubes. […]."*

RC2.48 page 5, row 1 "DPSox" in table 2.

Corrected as per Table 2.

RC2.49 page 5, row 3 "Beauchemin et al., 1999" -> "Beauchemin and Simard, 1999"

Done

RC2.50 page 5, row 4 "Borovec et al., 2018" -> "Borovec and Jan, 2018" page 5, row 11

Done

RC2.51 Please introduce the meaning of D.L. at the first appearance! If detection limit is meant, consider to use "limit of quantification" instead.

Done (page 6, row 10): *"with limit of quantification (D.L.)"*

RC2.52 page 5, row 14 Please explain the meaning of "non-parametric data".

Done (page 7, row 2): "The non-normally distributed (*i.e.,* non-parametric) data […]"

RC2.53 page 5, row 14-22 The description of the factor analysis resembles - in my view as a nonstatistician – a parametric method. Please specify the non-parametric component of the statistical analysis.

Done. Please see note above.

RC2.54 page 5, row 20 Please check the grammatical structure of the sentence.

The revised text now read (page 7, row 8-11): *"The variables can be then plotted in groups with correlation among them being determined by their position (e.g., proximity, distance, orthogonality,). The two-dimensional plane where the rotated, normalized data were plotted can be interpreted in terms of the main controls over the general variance (see Vega et al., 1998 for details)."*

RC2.55 page 5, row 24 The reporting of results for soil texture and pH in one subchapter appears strange to me. Why not report pH together with other soil chemical variables?

This flaw in our original structure t has been corrected in the revised version of the manuscript (see new subsection in *4.2 Soil Chemical Characterization"*

RC2.56 page 5, row 26-28 This sentence is more or less a repetition from section 2.2.1. Accordingly, it may be omitted.

Text omitted as per reviewer's suggestion.

RC2.57 page 6, row 1 Please give a definition of "pebbles" and "cobbles".

In the revised text (page 7, row 13) we now use "coarse soil particles (gravel and stones)"

RC2.58 page 6, row 4-7 Consider to shift this part to section 3.3.

Moved to equivalent section 4.2.1 in the restructured manuscript.

RC2.59 page 6, row 10 Is the mean given for all five sampling plots?

This revised text was clarified as follow (page 7, rows 28-29; 8, 1-5): *"In the eastern part of the catchment, the cation exchange capacity (CEC) of the mineral soil at 40-80 cm depth was up to 33 meq kg-1 on the slope and 58 meq kg-1 on the hilltop (Table 2). By contrast, in the western part, the CEC was 22 and 19 meq kg-1, which is lower than the mean CEC values measured at all of the plots at UDL, whilst CEC in the valley was 27 meq kg-1, which is within the mean CEC values measured at all of the plots at UDL: 32 ± 7 meq kg-1 (Table 2). The range of base saturation (BS) values in the soil varied between 6 and 13 %, with higher BS observed in the east (> 9 %) as compared to the west (< 8 %). The CEC in the studied soil depth at UDL was dominated by exchangeable Al. Consequently, the soil base saturation (BS) and soil pHH2O values were also low (Table 2)."*

RC2.60 page 6, row 16 Please check "twice larger" for correctness.

Done. Revised text (page 8 row 12) now reads: *"The BS at UDL is twofold higher than the BS determined at similar soil depths in the leucogranitic catchment LYS […]"*

RC2.61 page 6, row 17 "Hruska et al., 2001" isn't in the references. Please check.

Revised. It is Hruska et al., 2000 (page 8, row 10)

RC2.62 page 7, row 4-6 Water volumes haven't been mentioned before. This sentence may be omitted or the relevance of water volumes for the study should be emphasized.

This information was removed as is not further used in our results or discussions

RC2.63 page 7, row 9 "… to be higher at …" -> "… to be highest at …"

Done.

RC2.64 page 8, row 2 According to figure S2, explained variance is 24%

True. Corrected.

RC2.65 page 8, row 3 According to figure S2, explained variance is 18%

True. Corrected.

RC2.66 page 8, row 3 Is “Fig. S2” meant here?

Correct. Revised (now page 9, row 11)

RC2.67 page 8, row 6-11 This part seems rather to be dedicated to the discussion. However, logical and grammatical consistency should be checked.

The text moved as per reviewer suggestion to discussion and also revised for grammatical correctness (page 12, rows 14-19): *“Finally, given the complexity of the possible interrelations among the environmental variables considered here, there was an apparent generally poor correlation between solute concentrations measured in the soil in 2012-2013 and decadal runoff and atmospheric deposition data compiled in Table 1 (after Ouhlele et al., 2017). Such a result in turn points to a major control exerted by groundwater chemistry over soil solution chemistry, and also to soil organic and inorganic ligand properties also exerting a control over the residence time of each of the measured components.”*

RC2.68 page 8, row 7 Please explain the meaning of “apparent insignificant correlation”.

See note above: *i.e.,* poor correlation

RC2.70 page 8, row 23-24 “Manderscheid et al., 1995“ -> “Manderscheid and Matzner, 1995“

Done.

RC2.71 page 8, row 24 “Hruska et al., 2000“ appears twice in the references. Which one is meant?

Corrected as per RC2.61

RC2.72 page 8, row 24 “Armbruster et al., 2004“ -> “Armbruster and Feger, 2004“

Done.

RC2.73 page 9, row 9 “Meyer et al., 2001“ is missing in references.

Done.

RC2.74 page 9, row 12-14 Please check grammatical consistency of the sentence.

Done. Now page 11, row 14-16: *“The latter effect seems to be critical in the variability in soil solution chemistry at the hilltops, where the subsoil level contain significant amounts of coarse parental-rock material (Table 2).”*

RC2.75 page 9, row 20 Is “Novak et al., 2005“ meant here?

Right.

RC2.76 page 10, row 4 What is meant with “areal control” here?

Text edited as follow (page 11, row 17-18): *“For Na+ and K+ ions in soil solutions , our spatially resolved, time-series observations (Fig. 3) show that their concentrations defined patterns and trends largely derived from heterogeneity in soil granulometry (Table 2), with seasonality […]”*

RC2.77 page 10, row 4-6 I can’t follow this statement.

Corrected. Please see note above.

RC2.78 page 10, row 8-9 What is the rationale behind the comparison of K and Na outputs?

The revised text (page 11) clarify the rationale and provide further information.

RC2.79 page 10, row 14-15 To which other period is the spring season of 2013 compared here?

The request for clarification of the reviewer has been addressed as follow (page 11, row 28-29): *" […] a seemingly more rapid response of Na^+ leaching to soil solutions could result from strong anion belowground episodic accumulation (Fig. 3; e.g., Spring 2013)*

RC2.80 page 10, row 21 "Heuck et al., 2016" -> "Heuck and Spohn, 2016"

Done.

RC2.81 page 10, row 23 "… in the 50 cm-depth …" -> "… in the 40-80 cm depth …"?

Done.

RC2.82 page 12, row 5 "… these periods ware compared." -> "… these periods were compared."

Done.

RC2.83 page 12, row 11 Figure A1a isn't visible in the manuscript. If "Figure 2A is meant, it should be corrected throughout Appendix A. page 12, row 26 Consider "… in soil pore spaces …" -> "… in soil pores …"

Done.

RC2.84 page 12, row 28 What is meant with "direct precipitation"? If the "contribution of direct runoff (or "event water" as in eq. 2) to total runoff" is meant, a clear definition of "direct runoff" should be given.

The originally unclear text now reads "bulk precipitation" (page 15, line 1); "direct" removed

RC2.85 page 15, row 21 "Fenn, E.M.;" -> "Fenn, M.E.;"

Reference removed.

RC2.86 page 15, row 22 "Stottlemeye, R." -> "Stottlemyer, R."

Done.

RC2.87 page 15, row 24-25 "FAO: Guideline for soil description; Rome, Italy, 2006" should be cited here.

Reference removed given that soil profile description were done in previous works See also reply to RC2.4.

RC2.88 page 15, row 32-16/2 Please give correct title of the reference.

Done.

RC2.89 page 16, row 3-7 "Hruska et al., 2000" appears twice. Please indicate the respective citations with "a" and "b". page 16, row 13 This line should be deleted.

Done.

RC2.90 page 16, row 22 "Ma, L; Teng, F-Z.; Lin, L.:" -> "Ma, L; Teng, F.-Z.; Lin, L.; et al.:" page 18, row 2 "Soiling" -> " Solling"

Done.

RC2.91 page 19, Figure 1 (b) Please consider to shift the sentence starting with “in the studied UDL …” to the text body of section 2.1.

Caption edited as suggested.

RC2.92 page 23, Figure 2A The content of this figure relates to Appendix A and should be placed in the supplements accordingly?

Done.