RC1.1 In the paper, hydrological problems rather than soil properties are discussed.

We have confidence that the "hydrological problems" considered in our *SOILD* are relevant for soil systems scientists focused on studying atmospheric-derived soil pollution and the detrimental propagation effects to contiguous ecosystem. See also response to RC1.3 below.

RC1.2 Unfortunately, the paper is not generally well written, organized and balanced. The introduction is very short and quite approximate. A real state of the art is completely missing. Introduction should more concisely lead to the objectives of the work. Usually, a general rationale is needed. What is the purpose of the study?

We thank the reviewer for thoughtful criticism and kindly pointing out weakness of our original manuscript with regard to organization, balance, revision of pre-established concepts, and for carefully going through the text. A revised introduction addressing the reviewer concerns was made available to the SOILD forum on Monday 04.05.19, with only minor editions in the revised version of the manuscript. In addition, and as described below, by addressing each of the reviewer' concerns, requests for clarification and suggestions to other sections of the manuscript, we have made every attempt to improve our report and look forward to the opportunity to submit the revised version of the SOILD.

RC1.3 Methods, results and discussion of soil properties are poor. Authors indicated that soil profiles were described according to FAO guidelines, but no descriptions are included in the paper. They investigated Podzols, but soil properties are described and discussed referring only to a layer of a depth of 40-80 cm, without taking into account soil genetic horizons.

Descriptions of the soil profiles in UDL and their classification were published elsewhere (e.g., Novak et al. 2005; Oulehle et al. 2017). This fact has been made clearer in there revised version of the manuscript. Constituents of the layer referred by the reviewer are necessarily mentioned since their mineralogy, granulometry (particulate surface area), variable organic content and weathering paths are considered to affect (or be reflected by) the spatially variability of the chemical composition of subsoil pore waters collected via lysimeters in our small mountain catchment. With regard to soil texture, we have modified the manuscript as follow: page 3, lines 32-33; 4 lines 1-4 (Study Site: Background Information): *"A detailed description of the soils occurring on this watershed has been previously reported (Novak et al. 2005; Oulehle et al. 2017). Accordingly, the soil in the catchment are mostly acidic Podzols developed on orthogneiss to which only the Entic qualifier applies. Low base status soils have developed at expense of the mineralogy of the porphyritic granite that served as protolith to the orthogneiss bedrock, and given the lack of lithological discontinuities, pedodiversity is low across the catchment area, with Mor being the most common humus"*

page 7, lines 17-20 (Results): *"In the eastern part of the catchment, coarse soil particles (gravel and stones) accounted for 24 % in the hillslope and 62 % of total soil granulometry at the hilltop, whereas in the western part of the catchment the soil particles above 10 cm in size accounted only for ~12 % (Table 2). The soil texture is loamy sand, with presence of authigenic clays (7-15 %) as weathering-induced alteration products of the orthogneiss parental material. The groundwater table is shallow."*

RC1.4 In two different sites on each hilltop and slope only one profile was investigated, and they are compared with the properties of only one soil profile located in a valley. All together, five soil profiles were investigated, and soil properties were compared. Soils are very different in their properties, especially in a mountain areas, so the number of soil profiles was not sufficient to compare and to conclude on differences in such soil properties as carbon content, pH, cation exchangeable capacity, base saturation, etc. This, in addition to ignoring soil genetic horizons of Podzols investigated, is a reason of the weakness of data interpretation.

We respectfully disagree with the reviewer in the assertion that one soil profile for sampling area is insufficient for discussing the likely effect of soil textural features on the variability of the parameters listed

above *vs.* legacy pollutants and nutrients imbalances. Previous, more detailed soil profile analyses in UDL has consistently show low pedodiversity (see note above on this regard).

RC1.5 Soil properties, especially pH, changes in time through a year. It seems that soil samples were collected only once (in October 2010?).

We thank the reviewer for carefully going through the paper and for requesting sound clarifications. The soil samples were obtained in July 2015. This is now mentioned in page 5, line 17. On the other hand, we intended to compare pH values measured in stream water *vs.* pH of soil solutions. The confusing typo has been corrected as follow (page 4, line 24-26): *"[…] during the decade 1994-2014, the median pH in the stream water remained stable in the range 5.2 ± 0.4. Over the same period, median pH levels measured in water percolating through the canopy (throughfall) increased from 4.1 to 5.2 (Oulehle et al., 2017)".*

RC1.6 Why [soil] pH values are compared to pH values of water collected during a whole year (page 5 lines 25-26)?

In the revised text, the lines highlighted here now read (page 6, lines 13-16): *"Table 2 lists physical data for mineral soil and chemical data for soil extracts from the 40-80 cm depth layer and data for soil solutions collected by suction lysimeters (50 cm depth). As described above, the dataset is grouped according to sampling position within the catchment area (i.e., hilltops, slopes and valley; Fig. 1)."* **That is, for the pH parameter there is no reference to the table directly serving for comparison purposes.**

RC1.7 It is not clear what data were measured by authors and what data were cited from the literature (Oulehle et al. 2017).

To make clearer this key aspect highlighted by the reviewer, we have restructured the manuscript. Accordingly, the revised introduction now includes the following lines (page 3, lines 22-24): *"This paper addresses primarily soil solutions chemistry in the UDL catchment. Supporting data on the chemistry of spruce canopy throughfall and stream runoff—parameters which are used here for comparison purposes, are accessible in Oulehle et al. (2017)."*

Also, the first call to pre-existing data used for comparison purposes in the text is made in the new section 2 Study Site and Background Information (*i.e.,* Fig. 2 and Table 1)

RC1.8 It is not clear how authors define runoff and throughfall (amount of water versus chemistry of water) and how these parameters were measured. These parameters should be defined.

Following this important reviewer's request for clarification, the word "runoff", when originally and unclearly referring to runoff water samples, has been substituted for "stream water(s)" and a brief definition of throughfall water is now provided in page 4, lines 25-26: *"[…] measured in water percolating through the canopy (i.e., throughfall) […]"*

RC1.9 Conclusions summarize obtained results, but not contain real conclusions. Authors did not underline any innovative aspect that this article provides with respect to what is already present in literature.

The Conclusions section has been rewritten taking in consideration this concern of the reviewer and additional suggestions to improve the section provided by Reviewer 2.

RC1.10 Several sentences are hard to follow, thus English proofreading is necessary.

Proofreading have been conducted to avoid runoff sentences.

RC1.11 Summarizing, I do not consider this paper as relevant enough that deserves publication in the Soil journal.

We have made every attempt to address the reviewer's criticism and requests for clarification, clarified its importance to the SOIL journal readership in the Introduction and Discussion section, and, thus, hope that the revised version of our SOILD could be considered for publication in the EGU SOIL.

RC1.12 After a major revision it would be considered for publication in a journal dealing with hydrology.

Please see to the author's reply to RC1.1 and RC1.11.

Detailed comments

RC1.13 page 4, line 1: "A total of 15 replicates (3 per sampling location) were collected monthly". What replicates were collected? Does that mean water samples collected from 5 sites, each 3 times monthly?

This request of clarification of the reviewer has been addressed as follow (page 5, lines 7-8): *"[…] Each nest consist of 3 lysimeters, and thus produce equal number of monthly replicates per sampling location. […]"*

RC1.14 page 4, lines 6-7: soil material from the depth 40-80 cm was collected. What soil horizons corresponded to this depth

This request of clarification of the reviewer has been addressed as follow (page 5, lines 24-25): *"Only results from the 40-80 cm soil level are considered here. This level is in chemical equilibrium with waters collected by our 50 cm depth lysimeter nets and correspond to horizon Bs2 in all plots."*

RC1.15 page 4, line 16: "Runoff samples were collected monthly at the limnigraph location". How these samples were collected?

To address this reviewer's query, the revised text now reads (page 6, lines 1-2): *"Stream water samples and runoff flux estimations were collected monthly at a V-notch weir in the limnigraph location (Fig. 1b) according to methods outlined in Kram et al. (2003)."*

RC1.16 page 4, line 4: " After centrifugation and filtration through 0.45 um cellulose–acetate filters, the filtrates were analyzed for cations" - this is not clear. Do these data refer to exchangeable cations? If so, the method was described in a wrong way. If not, what these data were measured for? Where these data were presented in the paper?

To address this flaw of the methods kindly pointed out by the reviewer, the revised text has been streamlined as follow (page 6, lines 7-11): *"Exchangeable cations in soils were analyzed in 0.1 M $BaCl_2$ extracts by atomic absorption spectrophotometry (AAS, AAnalyst Perkin Elmer 200). Exchangeable acidity was determined by titration of the extracts. Cation exchange capacity (CEC) was calculated as the sum of exchangeable base cations (Ca^{2+}, Mg^{2+}, K^{+}, Na^{+}) and exchangeable acidity. Base saturation (BS) was determined as the fraction of CEC associated with base cations"*

RC1.17 page 5, lines 25-26: "Table 2 lists physical data for mineral soil and chemical data for soil extracts from the 40-80 cm depth layer and compares them with data for soil solutions collected by suction lysimeters (50-cm depth)" - what does mean "soil extracts"? Were they obtained as described on page 4, line 4?

Please see note above.

RC1.18 page 6, line 3: "characterized by acidic pH". Reaction can be acidic, but pH may be high or low.

The offending line highlighted by the reviewer now reads (page 7, line 23): *"The soil at the 40-80 cm depth was characterized by $pH_{H2O} < 5$"*

RC1.19 page 6, lines 4-5: "The mean pH of soil solutions ranged similarly between the first and the second year, except for the valley (pH valley of 4.1 in year 1, and 4.5 in year 2; Table 2). The two-year

averages of soil solutions were", - this part of the text belongs to the paragraph 3.1. (Soil texture and pH). Does this data refer to soil extracts (page 4 line 4)? It seems that soil samples were collected just once in 2010, so how was it possible to obtain two-years averages? If these data refer to soil solutions obtained from lysimeters (page 3, line 28), they should be included in paragraph 3.3. (Solute concentrations in soil waters).

The data refers to soil water solutions obtained from lysimeters, not to soil extracts. As per reviewer's suggestion the text has been moved to section "4.2.1 pH, CEC and BS" of the restructured manuscript

RC1.20 page 6, lines 9-10: how 33 meq and 58 meq can give mean 32 meq?

We thank the reviewer for highlighting the lack of clarity of the initial data description. The revised text now reads (page 8, lines 1-2): *"[…] which is within the mean CEC values measured at all of the plots at UDL: 32 ± 7 meq kg^{-1} (Table 2)."*

RC1.21 page 6, lines 10-17: usually cation exchange capacity differ significantly through Podzol profiles, from ectohumus through albic and spodic horizons. The same concerns base saturation. I have a doubt if these values may be compared without referring to given soil horizon.

To address this important observation of the reviewer. The following feature was added to our description of Soil Samples (page 5, line 25-26): *"Only results from the 40-80 cm soil level are reported in this work. This level is considered in chemical equilibrium with waters collected by our 50 cm depth lysimeter nets and corresponds to horizon **Bs2** in all plots."*

RC1.22 page 6, line 26: authors used different terms to characterize chemical properties of water, namely: soil water, soil solution, mineral soil solution. It is not clear to what data refer concentration in soil water? Are they data from lysimeters?

A unification of equivalent terms referring to soil water from lysimeters has been implemented in this revision, and are nowonly referred to as "soil solutions"

RC1.22 page 7, line 2: how was runoff measured?

Please refer to author's answer to RC1.15 above

RC1.23 Were data in table 1 obtained by authors?

Table 1 is comprised of background information and this has been now made clearer. (Please see author's response to RC1.7)

RC1.24 page 7, lines 10-11: "Our results for NO3- across the lysimeter network also show that this chemical species was readily bioavailable along mostly in the valley, where its concentrations were one order of magnitude higher than in the upslope soil solutions" - it is not clear how this was deduced.

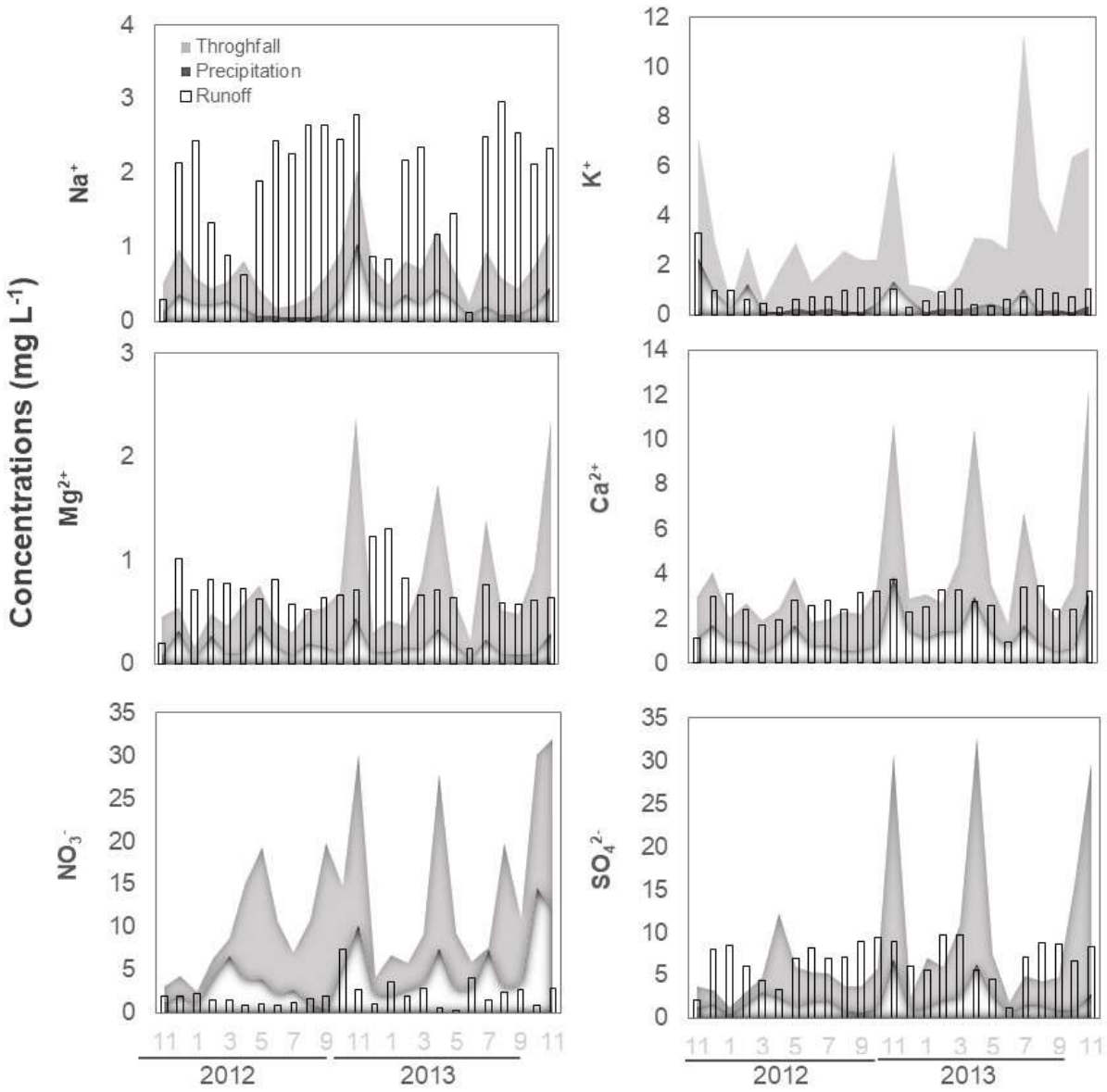
The revised text (page 8, 33 and 9, 1-2) now reads: *"Our results for NO_3^- across the lysimeter network also show that this chemical species was readily bioavailable along the study site but mostly in the valley, where its concentrations were one order of magnitude higher than in the upslope soil solutions (**Fig. S1**)."*

RC1.25 page 10, line 3: table 2 does not provide sufficient information on soil textures heterogeneity

Table 2 only presents the available relevant information on soil texture for comparison purposes in the studied low pedodiversity, UDL catchment area.

RC1.26 Figure 2 is unreadable due to unclear crosshatch. Does it present data obtained by authors?

As per reviewer request the crosshatch has been removed from the revised version of Figure 2 thus making it clearer. Figure 2 is background information and in consequence is first called in section 2, *Study Site and Background Information*.



RC1.27 Table 1. In Hydrology, throughfall is the process which describes how wet leaves shed excess water onto the ground surface. Was throughfall measured by authors? If so, how it was measured?

Generally accepted definition as used in our manuscript and related literature is the water percolating through the canopy (please see also author's response to RC1.8).

RC1.28 Table 2. Why (and how) soil particulate size (> 10 cm) were expressed in t/ha?

From the methods of estimation outlined in section 3.2. Soil Samples, where the weighted material from a given area. To address this concert, this parameter in Table 2 is now expressed in kg instead of ton.

RC1.29 Figure S2. What does mean the following: "hydrochemical data for runoff, atmospheric in lysimeters"?

We thank the reviewer for carefully going throughout the manuscript. The offending line caption now reads: […] *“Non-parametric multidimensional scaling ordination of time-series hydrochemical data for runoff, atmospheric and soil solutions collected in lysimeters.”*