nature portfolio

Peer Review File

Contaminated landscapes – Current Use Pesticides residues in soil and vegetation along altitudinal gradients in a European alpine valley

Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality

9th Aug 23

Dear Professor Brühl,

Your manuscript titled "Contaminated landscapes - Current Use Pesticides residue measurement along altitudinal gradients in a European alpine valley" has now been seen by 3 reviewers, and we include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript that meets the Editorial thresholds listed below before we make a final decision on publication.

Editorial thresholds:
• Substantially improve the modelling approach to ensure robustness, taking into account underlying uncertainties and limitations.
• Make sure that results and discussion are interpreted by taking into account the fact that the measurements were done during spray season.

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter), a tracked-changes version of the manuscript (as a PDF file) and the completed checklist:
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** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

Please do not hesitate to contact us if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

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Communications Earth & Environment

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Clare Davis, PhD
Senior Editor
Communications Earth & Environment

EDITORIAL POLICIES AND FORMATTING

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Editorial Policy: Policy requirements (Download the link to your computer as a PDF.)

Furthermore, please align your manuscript with our format requirements, which are summarized on the following checklist:
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and also in our style and formatting guide Communications Earth & Environment formatting guide .

*** DATA: Communications Earth & Environment endorses the principles of the Enabling FAIR data project (http://www.copdess.org/enabling-fair-data-project/). We ask authors to make the data that support their conclusions available in permanent, publically accessible data repositories. (Please contact the editor if you are unable to make your data available).

All Communications Earth & Environment manuscripts must include a section titled "Data Availability" at the end of the Methods section or main text (if no Methods). More information on this policy, is available at http://www.nature.com/authors/policies/data/data-availability-statements-data-citations.pdf.

In particular, the Data availability statement should include:
- Unique identifiers (such as DOIs and hyperlinks for datasets in public repositories)
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If a community resource is unavailable, data can be submitted to generalist repositories such as figshare or Dryad Digital Repository. Please provide a unique identifier for the data (for example a DOI or a permanent URL) in the data availability statement, if possible. If the repository does not provide identifiers, we encourage authors to supply the search terms that will return the data. For data that have been obtained from publically available sources, please provide a URL and the specific data product name in the data availability statement. Data with a DOI should be further cited in the methods reference section.

Please refer to our data policies at http://www.nature.com/authors/policies/availability.html.

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

The manuscript titled “Contaminated landscapes - Current Use Pesticides residue measurement along altitudinal gradients in a European alpine valley” presents the analyses of one single monitoring campaign of current use plant protection products (PPPs) measured in soil and vegetation samples, along altitudinal gradients, at different locations within a European alpine valley.
The objectives of the research are clearly stated. Methods are well written; for the monitoring, they are also scientifically sound. The field work have generated interesting data contributing to fill the scarce but growing literature on soil contamination monitoring. The paper provides strong evidence for the main conclusions. The implications would be interesting for environmental scientists and would flank the need for a EU soil framework directive as it is being discussed.
However, the conclusions are not necessarily novel. The Authors themselves refer to earlier literature reporting on high PPPs concentrations in the studied area. Importantly, other researches on soil contamination monitoring were not mentioned in this study. These studies would allow for a comparison of the contamination extent among different regions. Moreover, in my personal opinion, the data modelling is far-fetched and could be both validated and largely improved for robustness. On a last note, the Introduction reads as separated paragraphs and do not guide the reader into the study.

Below there are my detailed comments.
Abstract
L27 “originating mostly from applications in valley apple orchards.”. I see that apple orchards is the main land use in the Vinschgau valley. Still, in order to state that, the authors shall show at least that: a) the found PPPs were applied on apple orchards only (the analysis at L81-88 shall be extended), or b) the concentrations of PPPs that could be applied on other land uses except for apple orchards are significantly lower than the measured concentrations of PPPs that could applied on apple orchards too (L92-94 should be highlighted). Furthered in the results section.
L30 The Principal Component Regression (PCR) is not modelling, but it is a regression analysis technique. Please revise.

Introduction

L40, the authors mention that soil monitoring is limited. However, there exist relevant publications in this context which have not been mentioned (not my papers). The latter suggestion has a focus on apple orchards.

Pesticide residues in European agricultural soils – A hidden reality unfolded (https://doi.org/10.1016/j.scitotenv.2018.10.441)

Pesticide residues in agricultural soils in light of their on-farm application history (https://doi.org/10.1016/j.envpol.2023.121892)

Quantification of pesticide residues in the topsoil of Belgian fruit orchards: terrestrial environmental risk assessment (https://doi.org/10.1002/ps.5811)

L41 perhaps a comma is missing before “soil”.

L42, the authors claim for the need of long-term monitoring, although their research captures a snapshot of the spraying season. Please revise accordingly. For example, farmers in South Tyrol already participate in continuous soil monitoring, but the data need to be requested to the analysis laboratories. In the following paper, other soil parameters rather than PPPs were used, but the latter information may be available.

Farmers as data sources: Cooperative framework for mapping soil properties for permanent crops in South Tyrol (Northern Italy)

https://doi.org/10.1016/j.geoderma.2019.02.010

2.2. Soil sample data sourcing

In South Tyrol, viticulturists do not follow specific guidelines for soil management, while apple farmers conduct integrated farming and follow precise guidelines and regulations that prescribe soil sampling every 5 years (AGRIOS, 2018). Notwithstanding, about 70% of all the farmers in South Tyrol send soil samples to the Public Research Centre of Agriculture and Forestry, Laimburg; the other 30% send them to smaller local laboratories, and thus these soil data may be difficult to access. Data used in this study is publically available upon request and registration at the Research Centre for Agriculture and Forestry, Laimburg, Bolzano/Bozen, Italy. The license agreement prevents commercial use, while consent use of the data for research, educational and dissemination purposes. Derivative and aggregated products can be published if they do not contain cadastral parcel code in order to keep farmers data anonymous. Data refer to soil properties for apple orchards and vineyards located in the Venosta/Vinschgau and Adige/Etsch valleys and analysed by the Laimburg laboratory during the period 2006–2013

L44-53 Here I feel that the authors are not making their case for apple orchards. The jump from crops in this paragraph to orchards in the following one. I would suggest to include literature on apple orchards.

L69 Which part of the vegetation was sampled has a big impact on the measured concentrations. For example, apple orchards, as other plants, can transport and store different PPPs in different part of the tree (Development and application of a numerical dynamic model for pesticide residues in apple orchards, https://doi.org/10.1002/ps.6897).

L81 Here I would insert the measured concentrations, which is your result.

L81-94 currently this paragraph is an analysis that support your results. I feel like the reader forgets all the numbers before arriving to the concentrations.

L91 Clothianidin is also a major TP of thiamethoxam. But also thiamethoxam should be banned, right? Were there any emergency use of these banned PPPs in Italy? Some countries obtained it for sugar beets.

L95 “sum concentrations” but concentrations have not been shown yet. And here the authors are already looking at correlations with the land use (in particular, apple orchards). This analysis should

come after concentrations are presented.
L102 perhaps «a» to be removed?
L106 I cannot see the measured concentration for a single active ingredient at a single location. Is there a reason why only mean sum max min are reported?
In addition, am I right to understand that when calculating min and mean the concentration values below the level of quantification (LOQ) were disregarded (Table A2)? This would introduce a big bias in your statistics. Values below LOQ can be inferred to be 0, LOQ/2, etc.
Finally, the reported concentrations for "vegetation" samples, are in line with PPPs concentrations measured in leaves of riparian trees next to agricultural fields (Systematic Underestimation of Pesticide Burden for Invertebrates under Field Conditions: Comparing the Influence of Dietary Uptake and Aquatic Exposure Dynamics in the Supporting Information; https://doi.org/10.1021/acsenvironau.1c00023). Are the measured PPPs systemic or not (i.e., can they be taken up by the roots or is it dry and wet deposition)?
L139 Figure 2 is very interesting, it is a good job. If I understand correctly, it could be enhanced even more. Basically, the "0" could be on a fixed row, and transect rising on the left (hydrological sense – watching downstream) of the valley will be on one side and the transects rising on the right of the valley will be on the other side. In this way, we would see the continuity along the transects up-bottom-up. Just an idea.
L176 is there any permitted use in forestry for the measured PPPs in remote areas?
L232 The PCR is far fetched in my opinion. As found in the paragraph staring from L166, there were no relationships between landscape features and concentrations. For example, again in "Farmers as data sources: Cooperative framework for mapping soil properties for permanent crops in South Tyrol (Northern Italy) (https://doi.org/10.1016/j.geoderma.2019.02.010)" more than 18,000 georeferenced points were used to develop spatial maps using the state-of-the-art random forest model. I think the authors really need weather data to perform in-depth air trajectory analysis given the largely small number of points to use in geospatial mapping. There are also Bioproducers in the Vinschgau valley (https://www.vip.coop/en/biography/farmers/27-0.html). Can they be accounted for should the authors maintain the regression/modelling?
Rather than modelling, I would personally look into whether single PPPs cluster regionally? In fact, food supply chains may recommend which PPP to use in order to avoid the multi-residue risk in food and food commodities.
Continuous high-frequency pesticide monitoring in a small tile-drained agricultural stream to reveal diel concentration fluctuations in dry periods
10.3389/frwa.2022.1062198
The indication of a widespread and extended use all over the year of fluopyram in the catchment comes from application data (shared by farmers with the cantonal environmental office who kept the data confidential). The identified reasons for this widespread use are driven by the fungicide's low toxicity against tested organisms under the current regulatory framework and its diverse use on crops grown in the catchment. The food supply chain also recommended to their consortium farmers to use the same PPP, which in this case was fluopyram, in order to avoid the multi-residue risk of PPPs in food and food commodities. And pip fruit growers did treat their apples before harvest (in autumn) to mitigate the risk of fruit rotting during storage, and therefore, secure their income up to the point of sale to any array of buyers. Because the choices for widespread fluopyram use grounded on local factors, such as expert recommendations, personal preferences and agricultural practices, it was the responsibility of the whole food value chain to undertake actions to reduce the use of PPPs in a more general context (as described in Möhring et al., 2020).
L236 how was the PCR performance calculated? How about performing a k-fold cross-validation?
L238 why did the authors predicted the sum of the concentrations for all PPPs in soil and

vegetation? Is it not an extreme overestimation?
As the authors also mention, the map they produced is relative to the measured concentrations, and given that the sampling campaign was done during the spraying season, it is possible that the concentrations may be lower.
L245 Figure 3, the legend should not be continuous, the number of current used pesticides (CUP) is an integer. Why the left side of the map is cut off? Transects 9 and 10 have no values.
L337 perhaps “s” to be deleted?

Additional questions
Are the measured concentration values within calibration range? Or do the authors expect linearity also outside the calibration range. Or if not, would it be better to state when the measured concentrations are semi-quantitative?
L74A How did the authors account for possibly matrix effect? Did the authors use isotope labelled standard?

I hope my comments are helpful in improving the clarity and robustness of the manuscript.

Best regards
Daniele la Cecilia

Reviewer #2 (Remarks to the Author):

The manuscript entitled “Contaminated landscapes - Current Use Pesticides residue measurement along altitudinal gradients in a European alpine valley” presents interesting results deserving publication. The manuscript is well written and aim to study the contamination of soil and vegetation by current used pesticides in altitudinal transects located in a valley of intensive apple production. If the purpose of the study and the results are really interesting there is a lack of real presentation of the results with detailed information on the substances detected, their contribution to the total concentrations and the range of concentrations found. The authors need to describe their results more and also to be more critical especially concerning the modelling of concentrations using PCA and PCR-model given the low number of sites and the absence of data on soil properties in the analysis. To me this attempt of modelling is too ambitious considering the amount of data available and leads to a prediction highly overestimating pesticide concentration which is never pointed out. The model and the associated results should be taken with caution and the many limits should be highlighted. In addition, the sampling was done during spay season yet this is mentioned only at the end when it does highly impact the results! I think the authors can go deeper in discussing the results and integrating parameters such as DT50 or Koc in their interpretation. Degradation rates are displayed but never mentioned yet some of the substances found should be degraded in a few days.
I really believe in the potential of this study but many changes are required in the results and discussion part to better use the results and make a solid interpretation of those findings before this manuscript is suitable for publication. I detailed some other comments in the following paragraphs.

Comments

Introduction
L41: Pelosi et al. 2021 also looked at earthworms and not only soils
L62: Since you are looking at alpine gradient, why selecting only meadows and not forests soil that might also be exposed due to the effect of the canopy as a trap for deriving pesticides.

Material and methods
L308: what type of soil can be found in this region? Soil characteristics might influence the fate of pesticides in soils.
L312: you mention the sampling period in May, is it close to the main pesticide application in apple fields? Was it the objective to collect samples just after CUP application?
L313: Again, why focusing on grasslands and not forest also? Since they cover a large part of the valley we could expect the contamination of forest, perhaps with different substances…
L315: Please be more specific about the vegetation (mainly grass, did you keep roots? Etc.), and did you collect vegetation only in the 5m x 5m area where you sampled soil?
L315: Did you remove humus and vegetation before sampling soil?
L318: How was made the selection of the 97 CUP? Only those used in apple fields?
L323: Did you measure soil characteristics as well? Carbon, clay content etc. Some differences in pesticide content between soils could be explained by soil properties. As said in the supp mat line 43, some soils seem to have a high carbon content!
L330: As said before, why not include soil characteristics in the PCA? And also pesticide characteristics such as Koc that could explain the persistence in soils.

Supplementary
L4: A table with physico-chemical properties of soil samples might be included.
L34: If I understand correctly, a mixture or isotopically labelled standard with only imidacloprid was added to the reference soil sample known to have no pesticides residues? How about other substances that could be influenced by matrix effects during extraction?
L66: It could be interesting to have the list of the molecules included in the standard mix solution.
L71: Table A4 you mean?
L122: Physico-chemical properties of soils?
Figure 3A: this figure is of poor resolution making it difficult to read. Please provide a better-quality plot.
L169: I think we should have the results of PCA analysis (cos2 and factor loadings etc.) with both log.conc and the number of substances in a detailed table. Also, the PCR model (coefficient, residuals etc.) and the results should be included, not only the map. To allow the reader to have a critical view of the modelling and the PCA results.
L161: There is a big issue I think in using PCR to predict pesticides in forest soils covering a large part of the study region when the monitoring was conducted only in meadows. Forests should be excluded of the mapping process to keep only land cover related to the monitoring.

Results and discussion
L86: Do you have information on the amount of use of individual substances? To see if the most concentrated or frequent substances are also the most applied?
L99: This is the first time you mention the period of sampling: if I understand well, this period is not considered as the highest pesticide application period? Do you know what type of pesticides are used first in the season? This would definitely impact your results if the orchards treatments begin in march/April with fungicides and continue in May/June with insecticides…

L102 to 128: I miss a small part in the main text describing the main substances found in the soils and vegetation and the detection frequency by substances. The main cocktail of pesticides found in soil and vegetation would be interesting as well as the contribution of individual substances to the cumulative concentration.
L143: When was conducted the air sampling of the cited study?
L157: what about the meteorological parameters before the study? To identify the main wind direction for example? The absence of soil parameters makes the interpretation of the results highly questionable especially considering the attempt of predicting the distribution in the valley without any data on forest soils contamination nor any soil characteristics.
L169: Where can we find this information in the SI? I do not see any table reporting by transect point the concentration by substance, this would be interesting, also the sum concentration by type (fungicide, insecticide and herbicide).
L185: The canopy could act as a trap of substances brought by the wind, therefore high concentrations in soils near to forest areas is not surprising.
L188: You did not present the substances with the highest concentrations in soils…
L193: Do you have any information on the amount of those insecticides used? If one is not that volatile but widely used it is quite expected that we will find it everywhere…
L202: this is very important indeed!! To identify those substances that should not drift in the atmosphere!
L210: What about the Koc to predict their persistence in soil? And in the atmosphere linked to the particles?
L219: You never discuss the relationship with expected degradation time. Trifloxystrobin has a DT50 of 1 to 3 days and is reported in the highest transects, this is never pointed out in the discussion…Same thing for fluzinam, 21 to 34 days of DT50.
L232 to 239: Many limitations question the real interest in doing PCA + PCR: first the absence of soil parameters, and the selection of only soils in meadows when a lot of forests are covering the valley as I mentioned before. Also, the wide differences between the groups of transects which are not disposed regularly. Specifically, between T5 and T7 separated by ~ 15km, the cumulative concentration of pesticides is multiplied by 4.5! With such differences, the final prediction will be influenced by the one very high concentration of almost 3000 µg/kg… This leads to predicted pesticides concentrations of 7000 µg/kg in the valley (Fig4A) when the real measurements of the 6 transect points inside this highly contaminated area do not go beyond 3000 µg/kg for the maximum and then get down to 700 and 800 µg/kg. The overestimation of the concentrations is quite obvious here and probably due to the PCA and PCR based on log concentration.
L248 to 260: You never question your model based on few points, predicting concentration in the middle of the valley where there are only five points! You need to be a bit critical especially when mentioning the concentration of 7000 µg/kg predicted when you found half of this concentration in the real samples!
L257: Finally, you mention the spay season which may completely impacts your results! This has to be discussed with more hypothesis of why finding some substances might be related to the use in orchards just before your sampling…

Conclusion
L278: Yes, but you did not discuss the Koc parameter that could be an indicator of the affinity to particles…

Reviewer #3 (Remarks to the Author):

Review of:
Contaminated landscapes – Current Use Pesticides residue measurement along altitudinal gradients in a European alpine valley.

General review and subject summary
I think this is an interesting and important paper. The authors present a well-structured manuscript on the assessment of landscape contamination with currently used pesticides (CUP) by analyzing soil and vegetation samples from different altitudes.
The aim of this research was to better understand the fate of CUPs, their distribution and the magnitude of residues along altitudinal gradients. I believe this work provides new and important information towards this goal.
The study design, the selection of two different sampling matrices, the selection of sampling points and transects, the sampling itself and its duration, the analytical methods as well as the statistical methods applied are appropriate and innovative.
In my opinion, the selection of this well studied area is not a weakness of the study, but rather a strength, as the new data on pesticide residues along elevation transects complements previous research in this field and within this geographical area.

To my knowledge, the submitted work is the first to present a more landscape-oriented approach, whereas in the past studies have mainly focused on specific sites such as sensitive areas. (playgrounds, official places, landscape conservation areas, high altitude places).
Since even glaciers are contaminated with CUPs and old residues, this work again supports the hypothesis and findings of previous research that pesticide use is a landscape-wide problem, including the exposure of remote high alpine habitats and protected areas.

With these results that the number and concentration of CUPs decreased with altitude and distance from agricultural application sites, the researchers confirmed previous findings that most of the detected CUPs originate from apple production and that distance to source is a major factor in CUP contamination.
The results are not entirely new; however, it is very important to confirm previous findings, generate new monitoring data and combine them.

This research closes the gap in pesticide residue monitoring between the lower valley and high-altitude measurements such as glaciers.

I like the prediction method with PCR, but there are some methodological issues that could lead to too homogeneous overestimation at the valley bottom and underestimation along the elevation transects of the prediction model.
The PCR is appropriate, but can be improved in its application and reporting. However, the current manuscript needs some minor revisions regarding the statistical analysis and the presentation/description of the analysis. It is not clear to me why soil and vegetation samples are combined for prediction, as this work demonstrates that they differ significantly in the number and concentration of CUPs.

The authors present very interesting and new aspects for the field of CUP monitoring, therefore I

recommend the acceptance of the paper after some revisions.

Remarks
- Recommendation for other or additional references:

Line 41: Why did the authors not mention measurements of terrestrial matrix vegetation/grass samples from previous studies conducted in the area? Unless there are arguments against it, I suggest mentioning at least one of the studies on vegetation samples:
Cech, R. et al. Pesticide drift mitigation measures appear to reduce contamination of non-agricultural areas, but hazards to humans and the environment remain. Sci Total Environ 158814 (2022) doi:10.1016/j.scitotenv.2022.158814.
Linhart, C. et al. Year-round pesticide contamination of public sites near intensively managed agricultural areas in South Tyrol. Environmental Sciences Europe 33, 1 (2021).
Linhart, C. et al. Pesticide contamination and associated risk factors at public playgrounds near intensively managed apple and wine orchards. Environmental Sciences Europe 31, 28 (2019).

Line 43: The need for long-term and comprehensive monitoring was mentioned earlier than 2021, already in Linhart et al. (2019 and 2021).

Line 49: CUPs were detected in glacial streams near and in South Tyrol. In particular, glaciers north and south of the studied transects were investigated:
Forni Glacier in the Ortles-Cevedale group and Giogo Alto Glacier in the Palla Bianca-Similaun group in Italy. I propose to include this reference and to compare the results:
Rizzi, C., Finizio, A., Maggi, V. & Villa, S. Spatial-temporal analysis and risk characterization of pesticides in Alpine glacial streams. Environmental Pollution 248, 659–666 (2019).

Line 84: ISTAT data for 2019, recent data are not available?

Line 122: MRL exceedance was found in this area for the valley bottom (Linhart 2021).
Linhart, C. et al. Year-round pesticide contamination of public sites near intensively managed agricultural areas in South Tyrol. Environmental Sciences Europe 33, 1 (2021).

- Structure of the article
The main article presents the most important information about the study design and results. I like the short and straightforward format and the detailed information in the Supplementary Information for interested readers. However, for better readability, I recommend splitting the Results/Discussion section into at least two subheadings:
e.g. 1) CUP Residues and Identification of Related Factors, 2) CUP Prediction

- Tables
All tables are clear and present the important information very well. Good presentation of LOD and LOQ.

- Results and discussion:
Line 95-97: The sentence refers to the significant correlation of number and sum concentrations of CUPs with the proportion of apple orchards. However, the mentioned table (Table A1) does not show the results of the correlation. Please also refer to the results of the correlation.

Lines 102-114: I suggest a better order for the sentences.
Line 102-106: The maximum number of residues detected per site is discussed. The next sentence jumps to the differences between soil and vegetation samples - which I think is a separate, very important result. Then, at line 112, the sentence jumps back to the multi-residue problem.
The paragraph comparing soil and vegetation interrupts the discussion of multi-residue contamination. Please reorder and use a paragraph to separate these two findings (multi-residue results, comparison of soil and vegetation matrix) for better readability.
The significant difference in the number and concentration of residues in soil and vegetation samples deserves its own section, in my opinion, as it is a very important result.

Line 130
The term "statistically" is not necessary. Delete.

Line 137: Figure 2
"Y-Axis": Continue the axis to 6, for transect #6. I suggest also adding an arrow to indicate the increase in elevation (0 m to 3000 m asl, or the highest sampling point).
It may help to improve the figure to indicate the "x-axis" with a west to east arrow or valley downstream arrow.
I assume the color scaling of the number of pesticides in soil and vegetation is the same? If so, it would be helpful to show the same numbering and number formatting on the color scale. Either digits for both scales or no digits at all.
I understand that the log scales for soil and vegetation should be different.

Line 151 - 158
Line 151: The authors state that increasing altitude is not the only factor leading to a decrease in CUPs at higher altitudes. This is quite obvious for a dependent variable under different environmental conditions. It is not always a linear relationship; even linear statistical methods are used. Try to reformulate the first sentence in terms of the other environmental factors. Later, you will present a multivariate statistical method. This sentence weakens the later use of PCA and PCR.

Line 152: Seasonal factors: A previous study conducted in this valley identified seasonality as a factor in CUP contamination of non-target areas, please cite: Linhart, C. et al. Year-round pesticide contamination of public sites near intensively managed agricultural areas in South Tyrol. Environmental Sciences Europe 33, 1 (2021).

Line 154: A previous study identified the most important environmental factors on CUPs for the same valley (bottom only): CUPS decrease with increasing irradiance and distance. At higher altitudes, irradiance is also higher. The work identified rainfall, percentage of apple orchards and wind (from source to sample site) in combination with increasing distance as factors increasing CUPs on non-target areas. I strongly suggest that the results of this work be included in this paragraph. Compare Fig. 2 and Fig. 3 in Linhart, C. et al. Pesticide contamination and associated risk factors at public playgrounds near intensively managed apple and wine orchards. Environmental Sciences Europe 31, 28 (2019).

Line 157: Meteorological data for different points within the studied valley are available retrospectively from the regional hydrographic service: https://wetter.provinz.bz.it/download-messdaten.asp

I know it is a lot of work to incorporate different meteorological data into a model. Also, the choice of data resolution can be crucial. However, I strongly recommend including these data, as the quality of the work would be improved enormously. The PCR model would be much more realistic and valid. In its current state, the PCR model is, in my opinion, quite rough and overestimated by landscape use variables. The inclusion of temperature data, irradiance, wind and precipitation would improve the validity of the model. These factors have already been shown to be significant factors for CUP contamination.
At this stage, the model depends mainly on distance, altitude and different land use variables, which, in my opinion, is not sufficient. The PCR with additional factors previously shown to be significant would result in a similar model, but with less even distribution of CUPs along the valley and elevation transects, reflecting a more realistic situation. CUPs could be even higher or lower than predicted by the actual PCR. So far, the model is most likely overestimated for the bottom valley and underestimated for the slope sites.

I agree that the CUPs are likely to be homogeneously distributed in the atmosphere, but they do not precipitate or fall homogeneously, or are homogeneously displaced over non-target areas.
If it is not possible to include the meteorological data during the revision of the paper, I suggest to state again that the model is likely to be over/underestimated and dominated by land use factors, since the former have been shown to be significant factors and are not included.

Line 166: Compare comment for appendix and "wind" section.

Line 185: Very interesting that a site with closed canopy cover had a relatively high concentration of CUPs. Are there any additional findings on this? If so, please provide literature.

Line 238: Why are CUPs in soil and vegetation not separated for prediction by PCR analysis?

Line 244: Figure 3: Why are transects T9 and T10 not covered by the predicted CUP residues and concentration using PCR?

Questions and recommendations about PCA and PCR:
The text of the main article and Figure 3 indicate that prediction for soil and vegetation samples are not analyzed separately. Why are these two variables, which are clearly described to differ significantly in number and concentration of CPUs, not distinguished for prediction analysis?
Figure A3: PCA Biplot: log CUP sum concentration of soil or vegetation or the sum of both?
Please clarify.
I recommend separating the analysis for soil and vegetation.
I recommend restricting the prediction of CPUs by PCR to vegetation samples, as they have a significantly higher contamination rate and therefore less zero inflation of the dependent variable, which would strengthen the PCR. A cluster description of the four clusters of grouped sites would also improve the PCA results and the selection of principal components would be clearer.

The application of PCA regression analysis allowed the prediction of CUP contamination from the valley floor up to nearly 3000 m elevation.
I like the PCA and PCR approach and don't see anything fundamentally wrong with the methodology. However, there are neglected uncertainties in the form of missing variables and cofactors that probably lead to a homogeneous overestimation of the predicted contamination of the valley bottom. The factors related to contamination of the valley’s bottom vegetation are clearly presented

and described in Linhart et al. 2019. The factors distance, number of apple orchards, rain, wind and irradiance were shown to contribute significantly to the amount of CUP contamination.
I am missing at least actual weather data; data summarized over three to five days prior to sampling would significantly improve the PCR model and better reflect particle-bound transport.

Also the use of PCA is not new, it is a widely used and quite "old" method in ecology for dimension and factor reduction; nowadays criticized in ecology & environmental science because the selection of variables/predictors and later the selection of principal components does not depend on the response variable. PLS regression would be a better alternative.
See: https://doi.org/10.4209/aaqr.2020.01.0001

The use of PCR and prediction modeling of possible contamination is a very useful tool. However, there are three issues:
Neglect of available cofactors (meteo data), unclear separation or merging of soil and vegetation data, number and concentration of CUPs could be combined by weighting the concentration by the number of CUPs. (cf. Linhart 2019).

Suggestions for improvement
Highlight differences between soil and vegetation results.
May highlight and discuss the differences between soil and vegetation samples. Why are the results sign. different?
For example: Co-factors of soil samples are more complex than vegetation samples. Soil pH, soil microbiome, no irradiance, stable temperature,…
Vegetation samples may be more in line with the CUP concept, as this matrix reflects current use, whereas soil or even sediment samples may also show the presence of previously used pesticides.

Is PCR better at predicting soil or vegetation CUP contamination? Clarify what was done with the data, combination of CUPs in soil and vegetation?
The PCR prediction model could be fitted and is a very good starting point for future research in this area. The inclusion of open-source meteorological data is recommended.
The PCR prediction is very interesting at this point, but contamination is not a static process, it is a very dynamic process, strongly influenced by environmental factors.
I recommend using only the results of CUPs detected in vegetation samples for PCR. For two reasons:
- Vegetation samples reflect CUPs better
- Vegetation samples correlate better with elevation and distance
Or at least distinguish between soil and vegetation, as these matrices reflect different habitats and different levels of contamination.

The discrepancies between the results of the CUPs along the altitude transect and the EFSA peer review conclusion on volatility and air transport are a very interesting and important finding. I think this could be highlighted more.

- Appendix
It is confusing that the appendix is referred to as Appendix A. Are there other documents? Appendix B? C? D?

Line 259

The section "Effect of Wind on Pesticide Distribution" is somewhat misleading and confusing. First, there is no mention of wind data being collected or presented. Second, the term "foehn" is mentioned, but the analysis described focuses on differences in pesticide residues as a function of slope orientation. Differences in pesticide residues on north- and south-facing slopes are possible due to this warm upwind, but the statistical test analyzes orientation and does not include wind data. This is somewhat confusing because the reader is expecting wind data. I suggest using the term "effect of slope orientation" instead of "effect of wind". The "foehn" wind could be an obvious influence, but since it is not measured, it should be mentioned afterwards as a probable cause in the interpretation.
Data on wind speed or wind hours defined as "foehn" may lead to different results than presented. I suggest changing the terms (orientation instead of wind).

Line 256
The authors refer to Tab. D13. However, Tab. D13 does not include weather in the main article or in the appendix. Please clarify.

Line 283
Expanded Discussion of Butterfly and CUP Presence
I think this is a very interesting part of the study and provides hypotheses for further research.
Is there also species richness data from vegetation samples? If so, this data would complete the link between butterfly biodiversity - vegetation diversity - CUP residues.

Author Responses: first round

	Reviewer 1 Daniele la Cecilia	*Response*	Adressed (yes / not applicable / no)	Changed to:
1	The manuscript titled “Contaminated landscapes - Current Use Pesticides residue measurement along altitudinal gradients in a European alpine valley” presents the analyses of one single monitoring campaign of current use plant protection products (PPPs) measured in soil and vegetation samples, along altitudinal gradients, at different locations within a European alpine valley. The objectives of the research are clearly stated. Methods are well written; for the monitoring, they are also scientifically sound. The field work have generated interesting data contributing to fill the scarce but growing literature on soil contamination monitoring. The paper provides strong evidence for the main conclusions. The implications would be interesting for environmental scientists and would flank the need for a EU soil framework directive as it is being discussed. However, the conclusions are not necessarily novel. The Authors themselves refer to earlier literature reporting on high PPPs concentrations in the studied area. Importantly, other researches on soil contamination monitoring were not mentioned in this study. These studies would allow for a comparison of the contamination extent among different regions. Moreover, in my personal opinion, the data modelling is far-fetched and could be both validated and largely improved for robustness. On a last note, the Introduction reads as separated paragraphs and do not guide the reader into the study.	*Thank you for your time and constructive comments. We want to highlight that our data set does not only include soil daat but also vegetation data which is - at least currently and to our knowledge - unique , when collecetd along altitudinal transects. Therefore the data are novel. Also soil samples were only in one study colelcted systematically along multiple transects, so also here we present a very novel data set. We agreethat CUP non-target contamination is recently reported in studies, but most of those focus onagricultral regions in the lowlands and at least to our knowledge there are no systemically cllected landscape data avaialabe . Here we present 11 transects along 80 km with a width of approximately 15 km. We also want to highlight that we avoid the bias of sampling at different times by collecting our data within only 4 days. Regarding the regression analysis we also refer to Reviewer 3 who agrees that the approach is valid indeed. We respond to comments in detail below. Regarding the introduction we changed some sentences and hope to now provide the necessary background for an easy understanding of our approach.*	yes	
2	L27 “originating mostly from applications in valley apple orchards.”. I see that apple orchards is the main land use in the Vinschgau valley. Still, in order to state that, the authors shall show at least that: a) the found PPPs were applied on apple orchards only (the analysis at L81-88 shall be extended), or b) the concentrations of PPPs that could be applied on other land uses except for apple orchards are significantly lower than the measured concentrations of PPPs that could applied on apple orchards too (L92-94 should be highlighted). Furthered in the results section.	*We shifted the text accordingly and hope that we now clearly state which of the detected pesticides were recommended for apple cultivation.*	yes	
3	L30 The Principal Component Regression (PCR) is not modelling, but it is a regression analysis technique. Please revise.	*We changed the wording according to the reviewer suggestion. Changes made: "Modelling" was substituted by "Regression analysis"*	yes	
4	L40, the authors mention that soil monitoring is limited. However, there exist relevant publications in this context which have not been mentioned (not my papers). The latter suggestion has a focus on apple orchards:	*Thank you. We are aware of a few more papers and did not mention them but the review by Sabzevari and Hofman, 2021. We added the additional references and added e.g. for clarification.*	yes	
5	Pesticide residues in European agricultural soils – A hidden reality unfolded (https://doi.org/10.1016/j.scitotenv.2018.10.441)	*We refer to the review by Sabzevari and Hofman, 2021 that includes Silva et al.*	yes	
6	Pesticide residues in agricultural soils in light of their on-farm application history (https://doi.org/10.1016/j.envpol.2023.121892)	*added*	yes	
7	Quantification of pesticide residues in the topsoil of Belgian fruit orchards: terrestrial environmental risk assessment (https://doi.org/10.1002/ps.5811)	*added*	yes	
8	L41 perhaps a comma is missing before “soil”.	*added*	yes	
9	L42, the authors claim for the need of long-term monitoring, although their research captures a snapshot of the spraying season. Please revise accordingly.	*we shifted the sentence to the conclusions as a better context to our measurement during the beginning of the application period*	yes	

10	For example, farmers in South Tyrol already participate in continuous soil monitoring, but the data need to be requested to the analysis laboratories. In the following paper, other soil parameters rather than PPPs were used, but the latter information may be available. Farmers as data sources: Cooperative framework for mapping soil properties for permanent crops in South Tyrol (Northern Italy): https://doi.org/10.1016/j.geoderma.2019.02.010 2.2. Soil sample data sourcing: In South Tyrol, viticulturists do not follow specific guidelines for soil management, while apple farmers conduct integrated farming and follow precise guidelines and regulations that prescribe soil sampling every 5 years (AGRIOS, 2018). Notwithstanding, about 70% of all the farmers in South Tyrol send soil samples to the Public Research Centre of Agriculture and Forestry, Laimburg; the other 30% send them to smaller local laboratories, and thus these soil data may be difficult to access. Data used in this study is publically available upon request and registration at the Research Centre for Agriculture and Forestry, Laimburg, Bolzano/Bozen, Italy. The license agreement prevents commercial use, while consent use of the data for research, educational and dissemination purposes. Derivative and aggregated products can be published if they do not contain cadastral parcel code in order to keep farmers data anonymous. Data refer to soil properties for apple orchards and vineyards located in the Venosta/Vinschgau and Adige/Etsch valleys and analysed by the Laimburg laboratory during the period 2006–2013	*Thank you for this information. In our paper however we aim to understand the landscape scale contamination with pesticides in the off-crop habitat, not within the apple orchards. However the information would be interesting. Following the suggestion we contacted the researchers at Laimburg for data on 1st of September and received an answer on 21st of September. We were informed that the process to obtain data is very complex, the location of Vinschgau cannot be selected and that residue data for soil are not available at all. It seems that unfortunately no data relevant to this study are available at this location.*	not applicable	
11	L44-53 Here I feel that the authors are not making their case for apple orchards. The jump from crops in this paragraph to orchards in the following one. I would suggest to include literature on apple orchards.	*In this paragraph we want to highlight that crops are not only produced in lowlands but alos in mountain valleys adn that there are only two studies world wide that evaluated pesticide distribution along transects. We refer to the crop - apples - in the next paragraph.*	yes	
12	L69 Which part of the vegetation was sampled has a big impact on the measured concentrations. For example, apple orchards, as other plants, can transport and store different PPPs in different part of the tree (Development and application of a numerical dynamic model for pesticide residues in apple orchards, https://doi.org/10.1002/ps.6897).	*Thank you. We added soil and vegetation and added off-crop habitat to clarify what we sampled and where.*	yes	
13	L81 Here I would insert the measured concentrations, which is your result.	*Thank you for your comments on structure. We restructured this results part and started with detections and added also more observations adn then continue with the concentrations.*	yes	
14	L81-94 currently this paragraph is an analysis that support your results. I feel like the reader forgets all the numbers before arriving to the concentrations.	*See above, we restructured this part and added the discussion after the result of detection and then approached the concentrations.*	yes	
15	L91 Clothianidin is also a major TP of thiamethoxam. But also thiamethoxam should be banned, right? Were there any emergency use of these banned PPPs in Italy? Some countries obtained it for sugar beets.	*Correct also Thiamtehoxam is banned since 2018. To our knowledge there was no emergency use in Italy, In Germany Thiamethoxam was used as seed treatment in sugar beet. To our knowledge there is no sugar beet cultivation in the Vinschagu and we are not aware of any other use.*	not applicable	
16	L95 “sum concentrations” but concentrations have not been shown yet. And here the authors are already looking at correlations with the land use (in particular, apple orchards). This analysis should come after concentrations are presented.	*Thank you for this suggestion. We now present teh detections first and then the sum concentrations and also added some more text regarding the quantitative results.*	yes	
17	L102 perhaps «a» to be removed?	*Thanks for spotting this. Removed.*	yes	
18	L106 I cannot see the measured concentration for a single active ingredient at a single location. Is there a reason why only mean sum max min are reported?	*Tab. A2 gives only an overview of detected substances, i.e. mean, sum, max, min etc. For concentration of CUPs at single locations we will incldue the raw data in a repository or make them available on request*	yes	
19	In addition, am I right to understand that when calculating min and mean the concentration values below the level of quantification (LOQ) were disregarded (Table A2)? This would introduce a big bias in your statistics. Values below LOQ can be inferred to be 0, LOQ/2, etc.	It is correct that min and mean values in Table A2 refer to values above the LOQ as indicated in the table heading. However all calculations/statistics were performed including all values below LOQ (set to the respective LOD). We added some text.	yes	Text added in methods : For pesticides detected at a level below LOQ but above LOD the respective LOD was used as concentration value for subsequent calculations as minimum residue that can be assumed with certainty to be present in the sample.
20	Finally, the reported concentrations for “vegetation” samples, are in line with PPPs concentrations measured in leaves of riparian trees next to agricultural fields (Systematic Underestimation of Pesticide Burden for Invertebrates under Field Conditions: Comparing the Influence of Dietary Uptake and Aquatic Exposure Dynamics in the Supporting Information; https://doi.org/10.1021/acsenvironau.1c00023). Are the measured PPPs systemic or not (i.e., can they be taken up by the roots or is it dry and wet deposition)?	*We added information on the systemic properties in the Si table and also incldued it in the text.*	yes	Thirteen of the eighteen detected CUPs in vegetation had systemic properties (SI Tab. A2 & A3) and their uptake from soil is possible, however when detected at the same sampling site concentrations in vegetation were in most cases orders of magnitude higher than in soil (e.g. Acteamiprid 0,31 µg kg^{-1} soil, 5,30 µg kg^{-1} vegetation at T6_0, Methoxyfenozide 7,01 µg kg^{-1} soil 1370,85 µg kg^{-1} vegetation, Penconazole 3,32 µg kg^{-1} soil 59,55 µg kg^{-1} vegetation at T7_1) and therefore dry and / or wet deposition is more likely.

21	L139 Figure 2 is very interesting, it is a good job. If I understand correctly, it could be enhanced even more. Basically, the “0” could be on a fixed row, and transect rising on the left (hydrological sense – watching downstream) of the valley will be on one side and the transects rising on the right of the valley will be on the other side. In this way, we would see the continuity along the transects up-bottom-up. Just an idea.	*Tjank you we reviesed the figure and added it, however some suggestions could not be incorporated. We hope the figure is now better to understand.*	yes	
22	L176 is there any permitted use in forestry for the measured PPPs in remote areas?	*Not to our knowledge.The only application is Btk a biocide that is applied locally to control lepidoptera.Btk concentrations can not be detected by our or any other method.*	yes	
23	L232 The PCR is far fetched in my opinion. As found in the paragraph staring from L166, there were no relationships between landscape features and concentrations. For example, again in “Farmers as data sources: Cooperative framework for mapping soil properties for permanent crops in South Tyrol (Northern Italy) (https://doi.org/10.1016/j.geoderma.2019.02.010)” more than 18,000 georeferenced points were used to develop spatial maps using the state-of-the-art random forest model. I think the authors really need weather data to perform in-depth air trajectory analysis given the largely small number of points to use in geospatial mapping. There are also Bioproducers in the Vinschgau valley (https://www.vip.coop/en/biography/farmers/27-0.html). Can they be accounted for should the authors maintain the regression/modelling?	*We agree with with reviewer feedback and want to give some context regarding the choice of a PCR as opposed to more advanced approaches such as ensemble learning methods (e.g. random forest models, RFE):* *Random forest models were used in preliminary analyses but due to the restricted number of data points the generalization performance of RFEs was not adequate, amounting to only 15 or 5 data points for model testing (as opposed to the very large dataset mentioned by the reviewer), i.e. following training and hyperparameter tuning in a 70-30 or 70-20-10 split, respectively. Thus, a PCR approach was chosen, albeit it sacrificing some explanatory power compared to RFEs. Furthermore, with a PCA/PCR approach the underlying drivers can be more readily identified and interpreted, which was a prime goal of this first exploratory analysis.* ***However, we now highlighted this fact prominently in the conclusion, suggesting that future investigations - specifically those analyzing pesticide occurence over time - will leverage more advanced approaches as soon as more abundant termporal data becomes available.*** *Regarding the use of additional weather data: The available meteorological data was insufficient for matching definite weather attributes with individual transects absed on preliminary research. In the study region, only four weather stations exist that were assigned to the 11 transects. However, this rough classification does not allow for a detailed analysis of weather conditions that may contribute to the distribution, deposition and degradation of pesticides in mountains, e.g. precipitation rates at slopes (Daly et al., 2007b, Tremolada et al., 2008), temperature differences between altitudes (Daly et al., 2007a, Feltracco et al., 2022) and incoming solar radiation (Daly and Wania, 2005, Rani and Sud, 2005).* ***However, the suggestion by the Reviewer (as well as Reviewer 3) has now been added to the end of the discussion, highlighting that future work will benefit from the inclusion of geological and meteorological data- if better measurement data will become available -, as well as, highlighting the current limitations of the PCR modelling approach (L 324ff).***	yes	
24	Rather than modelling, I would personally look into whether single PPPs cluster regionally? In fact, food supply chains may recommend which PPP to use in order to avoid the multi-residue risk in food and food commodities. Continuous high-frequency pesticide monitoring in a small tile-drained agricultural stream to reveal diel concentration fluctuations in dry periods 10.3389/frwa.2022.1062198	*Thak you for this suggestion. We carried out a cluster analysis and added the figure to the SI. As expected from differences in apple phenology there is clustering of Lower and Upper Vinschagu with Mid Vinschagu sites falling inbetween, but not forming distinct clusters. WE added some additional text.*	yes	Text added: CUP residues formed specific clusters for Upper and Lower Vinschgau but not for Mid Vinschgau. This can be explained by the phenology of apple trees at the time of sampling: Apple trees where starting fruiting in the Lower Vinschgau but started to flower in the Upper Vinschgau, whereas trees in Mid Vinschgau were falling inbetween. Tree phenology influences the application of CUPs, as for example insecticides against the apple codling moth (Cydia pomonella) are applied with the flowering, so the majority of orchards was already treated in the LV but not in the UV.
25	The indication of a widespread and extended use all over the year of fluopyram in the catchment comes from application data (shared by farmers with the cantonal environmental office who kept the data confidential). The identified reasons for this widespread use are driven by the fungicide’s low toxicity against tested organisms under the current regulatory framework and its diverse use on crops grown in the catchment. The food supply chain also recommended to their consortium farmers to use the same PPP, which in this case was fluopyram, in order to avoid the multi-residue risk of PPPs in food and food commodities. And pip fruit growers did treat their apples before harvest (in autumn) to mitigate the risk of fruit rotting during storage, and therefore, secure their income up to the point of sale to any array of buyers. Because the choices for widespread fluopyram use grounded on local factors, such as expert recommendations, personal preferences and agricultural practices, it was the responsibility of the whole food value chain to undertake actions to reduce the use of PPPs in a more general context (as described in Möhring et al., 2020).	*Thank you very much for this information. Fluopyram is a widspread fungicide also for example in wine growing. Interestingly it is also marketed as a Nematicide in tropical crops, as there is supposedly also action against other organisms. Interestingly although detected a few times only it was not recommended for integrated production. We added this in the text*	yes	
26	L236 how was the PCR performance calculated? How about performing a k-fold cross-validation?	*Thjank you. You are correct and PCR validation was already performed using cross validation (k = 10 line segments) as suggested, with the RMSEP being used as a the main validation metric and the final model accurracy being expressed with the R-squared for the ease of interpretability. The information was missing but is now added to the SI in L-163ff.*	yes	
27	L238 why did the authors predicted the sum of the concentrations for all PPPs in soil and vegetation? Is it not an extreme overestimation?	*Thank you for this comment. aimed at descibing the contamination of both enviroenmental matrices. Insects for example develop in soil as larvae but feed on vegetation as adults.Conce ntration sin soil were generally lower and did not affect the pattern much. We additionally now produced a figure with the concentrations in plants only, so the interested reader can get an impression on vegetataion onyl sum concentrations.*	yes	

28	As the authors also mention, the map they produced is relative to the measured concentrations, and given that the sampling campaign was done during the spraying season, it is possible that the concentrations may be lower.	*Yes, we expressed this in the sentence: It has to be noted that the model is based on the residues detected at the beginning of the spray season in apple orchards in May 2022. Thus, landscape contamination patterns are certain to change throughout the year, depending on CUP applications and weather conditions affecting their distribution. We added more details on the spraying scheme , although only data for 2017 are available. Nevertheless it gives an indication that applications continue until end of September, potentially resulting in even higher resiudes.*	yes	The only availbale evaluation of farmers spraying records in the Vinschgau is for 2017 and shows most applications in spring between mid March and End of May, however there is no day between beginning of March and end of September without pesticide applications (Baumert et al. 2017). In 2017 insecticide applications peaked in early April and fungicides at the beginning of May. Application patterns are weather dependend and vary from year to year but our sampling period falls within the time of high pesticide applications. With the continuous applications until the end of September, accumulation of pesticidies, resulting in even higher amounts than recorded here is possible.
29	L245 Figure 3, the legend should not be continuous, the number of current used pesticides (CUP) is an integer. Why the left side of the map is cut off? Transects 9 and 10 have no values.	*Thank you for spotting this! We have extended the predicted area so that T9 and T10 are also included. Figure 3 was produced again, following the reviewer suggestions. The corrected Figure 3 is now added to the manuscript*	yes	
30	L337 perhaps “s” to be deleted?	*Thanks - deleted.*	yes	
31	Are the measured concentration values within calibration range? Or do the authors expect linearity also outside the calibration range. Or if not, would it be better to state when the measured concentrations are semi-quantitative?	*Yes , they are within the calibration range. Concentrations < LOQ are included in calculations as respective LOD. see above*	yes	
32	L74A How did the authors account for possibly matrix effect? Did the authors use isotope labelled standard?	*Thank you. We added some information for clarification in the SI.*	yes	info added in SI methods : A matrix induced signal suppression or enhancement was excluded for the relevant analytes by comparison of analytical response of spiked blank samples to standards.
	Reviewer 2			
34	L41: Pelosi et al. 2021 also looked at earthworms and not only soils	*Thanks we added the earthworms*	yes	added: earthworms (Pelosi et al. 2021),
35	L62: Since you are looking at alpine gradient, why selecting only meadows and not forests soil that might also be exposed due to the effect of the canopy as a trap for deriving pesticides.	*There are three reasons: As sampling is limited we aimed for comparability and chose similar sites. If we would also sample below tree canopy that data were already affected by another factor - open versus sourrounded by trees. Additionally forest are often private and sampling, especially from tree canopies would be difficult, logistically and also regarding permissions. Meadows fall within the allmende and sampling permission was granted by the local authorities. A third factor is safety for tree sampling and authorities are very strikt in this reagard. It would have been exremely difficult to get the encessary permission.*	yes	
36	L308: what type of soil can be found in this region? Soil characteristics might influence the fate of pesticides in soils.	*No soil characteristics were assessed but we are aware that soil charcteristics affect the disspation of pesticides.. We mention this in the text: " soil characteristics at the different sites were not assessed. Hence, possible differences among these factors along the altitudinal gradients that contribute to the observed differences in pesticides therefore require further investigations."*	yes	
37	L312: you mention the sampling period in May, is it close to the main pesticide application in apple fields? Was it the objective to collect samples just after CUP application?	*We mention" It has to be noted that the model is based on the residues detected at the beginning of the spray season in apple orchards in May 2022. " May is the start of the application period in the Upper Vinschgau. Apllications continue before harvesting in August. additionaly we added some more information on the pesticide application period in the Vinschagu - see other comments*	yes	Text addedd to clarify the pesticide application period.
38	L313: Again, why focusing on grasslands and not forest also? Since they cover a large part of the valley we could expect the contamination of forest, perhaps with different substances…	*To sample forest it would be necessary to sample the upper canopy, lower canopy and ground vegetataion below. We agree that this would be interesting, however this would be logistically (sampling along 11 transects in 4 days) extremely difficult and would require a totally differnt budget. Additionally safetly issues would complicate sampling permission in forests (see below). We also focussed on open vegetataion and meadows for comparability - see above.*	yes	
39	L315: Please be more specific about the vegetation (mainly grass, did you keep roots? Etc.), and did you collect vegetation only in the 5m x 5m area where you sampled soil?	*We added more information on the vegetation sampling in the text*	yes	Likewise, diverse, above-ground vegetation consisting of differing grasses and herbacous plants was hand collected randomly in an area of approximately 30 m_2 around each transect point using laboratory gloves, filling approximately half of a 1 L Ziploc freezer bag
40	L315: Did you remove humus and vegetation before sampling soil?	*Yes, we added the information in the text*	yes	Composite soil samples consisting of 25 subsamples were taken on a grassland area of 5 x 5 m per transect point with a soil corer (diameter: 13 mm, 10 cm deep, Rasengrün, Ingelheim, Germany) in a depth of 0 – 10 cm after removing vegetation from the soil surface.
41	L318: How was made the selection of the 97 CUP? Only those used in apple fields?	*No the CUPs were selected as substances with high usage in differnt crops and presence in water monitoring. We mention this referring two of our publications but additioally added a sentence in the SI.*	yes	Details on the selection of the target CUPs and analytical HPLC-MS/MS method and instrument parameters can be found in Bakanov et al. (2023) and Brühl et al. (2021) and the SI. IN the SI we added The 97 target CUPs were selected based on use records of the Julius-Kühn-Institute, Germany. CUPS frequently applied in winter wheat, oilseed rape, maize, potato and wine in 2016 and 2017 were selected. Additionally, newer insecticides such as for example chlorantraniliprole were included together with pesticides that were regularly detected in German small streams

42	L323: Did you measure soil characteristics as well? Carbon, clay content etc. Some differences in pesticide content between soils could be explained by soil properties. As said in the supp mat line 43, some soils seem to have a high carbon content!	*Yes we are aware of this and mention it in the text. However no soil characteristics were assessed for the soil samples.*	yes	
43	L330: As said before, why not include soil characteristics in the PCA? And also pesticide characteristics such as Koc that could explain the persistence in soils.	*Preliminary work was conducted to obtain additional geoclimatic data for the PCR approach. However, both climatic/meteorological data (see response above) and geomorphological/soil characteristics were not available or not available in sufficient resolution from the publicly accessible geoportal of South Tyrol. Nonetheless, we feel that some of the land-use / land-cover attributes have a proxy effect on attributes such as the soil organic carbon content (e.g. meadows vs. forests).Still, we added a snetence and highlight the usefulness of incorporating soil data in the future (L.325ff), if readily available. Information on Koc is also incldued (see below)*	yes	
44	L4: A table with physico-chemical properties of soil samples might be included.	*See above, soil characterists at the different sites were not assessed*	not applicable	
45	L34: If I understand correctly, a mixture or isotopically labelled standard with only imidacloprid was added to the reference soil sample known to have no pesticides residues? How about other substances that could be influenced by matrix effects during extraction?	*Thank you for your valuable comment. You are completely right, deuterated internal standards (e.g. imidacloprid-D4) play a critical role in analytical chemistry by enabling accurate and precise quantification of analytes, e.g. by compensating for matrix effects, correcting variability, etc. In the present study, we employed external calibration using matrix-matched standards as a common alternative to deuterated internal standards. We have not used any deuterated standards to compensate matrix effects of the field sample. We used the deuterated standard imidacloprid-D4 to test the extraction recovery and to verify that the extraction of each sample was correct by using imidacloprid-D4 as a representative analytical standard (which was spiked into each field soil and vegetation sample) and to repeat the extraction, if necessary. This procedure requires more time but ensures a high reliability of the method. We have clarified the issue and added new information on the methodology in the manuscript*	yes	We added paragraph in the Mthods description in the SI on matrix-matched calbration.
46	L66: It could be interesting to have the list of the molecules included in the standard mix solution.	*They are all mentioned in the SI Table A2. Te foornote indicates inclusion in the standard mix and other.*	yes	
47	L71: Table A4 you mean?	*Thank you for spotting this. Changed.*	yes	Table A4
48	L122: Physico-chemical properties of soils?	*We refer to the CUPs. We clarified this by adding details in te text.*	yes	changed to: and physical-chemical properties of CUP substances and CUP concentrations.
49	Figure 3A: this figure is of poor resolution making it difficult to read. Please provide a better-quality plot.	*We provided a higher reolution*	yes	
50	L169: I think we should have the results of PCA analysis (cos2 and factor loadings etc.) with both log.conc and the number of substances in a detailed table. Also, the PCR model (coefficient, residuals etc.) and the results should be included, not only the map. To allow the reader to have a critical view of the modelling and the PCA results.	*We agree and added detailed information about the PCA and PCR results to the SI. We added summary tables for the PCA such as cumluative explained variance, factor contribution, factor loadings and cos2. Similarly, we added Tables detailing PCR coefficients and provide additional graphics regarding predicted vs. actual values as well as the residual distributions.*	yes	We added the tables in the SI
51	L161: There is a big issue I think in using PCR to predict pesticides in forest soils covering a large part of the study region when the monitoring was conducted only in meadows. Forests should be excluded of the mapping process to keep only land cover related to the monitoring	*Thank you for metioning this aspect. In response to the Reviewer's suggestion we also provide an additional graphic in the SI that depcits (overlays) all forested area based on the LULC data. thereby excluding forests as requested, which is also noted in the Methods section now.*	yes	Additional figure added in SI and text in methods
52	L86: Do you have information on the amount of use of individual substances? To see if the most concentrated or frequent substances are also the most applied?	*Thank you for this question. The only reference available are spraying records from 2017 (Baumert et al., 2023). The most common fungicides were also in the most applied in 2017. Although not directly comparable we aded a sentence here. To our knowledge no application data are available for our sampling period.*	yes	we added: Although not directly comparable to our sampling period, Fluazinam and penconazole were also among the most commonly used CUPs in 2017, with highest numbers of applications, used by many agricultural opterations resulting in high volume applied (Baumert et al. 2023
53	L99: This is the first time you mention the period of sampling: if I understand well, this period is not considered as the highest pesticide application period? Do you know what type of pesticides are used first in the season? This would definitely impact your results if the orchards treatments begin in march/April with fungicides and continue in May/June with insecticides…	*We clarified the samplimg periods position in the spraying season by providing more details for the only available data for applications in the region - from 2017. Neverthelss it gives an indication about the spraying in the apple orhcards and is useful for the discussion of the findings.*	yes	The only available evaluation of farmers spraying records in the Vinschgau is for 2017 and shows most applications in spring between mid March and End of May, however there is no day between beginning of March and end of September without pesticide applications (Baumert et al. 2017). In 2017 insecticide applications peaked in early April and fungicides at the beginning of May. Application patterns are weather dependend and vary from year to year but our sampling period falls within the time of high pesticide applications. With the continuous applications until the end of September, accumulation of pesticidies, resulting in even higher
54	L102 to 128: I miss a small part in the main text describing the main substances found in the soils and vegetation and the detection frequency by substances. The main cocktail of pesticides found in soil and vegetation would be interesting as well as the contribution of individual substances to the cumulative concentration.	*Thank you. We restructured the results part about the detections and concentrations and added information on main substances and mixtures. WE also refer to the SI tables A2 and A3. The entire raw data will be available in a repository or on request.*	yes	See tables and new text parts.
55	L143: When was conducted the air sampling of the cited study?	*The samples were taken in 2019, added in the text.*	yes	info added: taken in 2019

56	L157: what about the meteorological parameters before the study? To identify the main wind direction for example? The absence of soil parameters makes the interpretation of the results highly questionable especially considering the attempt of predicting the distribution in the valley without any data on forest soils contamination nor any soil characteristics.	*We are aware of this shortcomings of the study and aded some critical sentences.*	yes	… Hence, possible differences among these factors along the altitudinal gradients that contribute to the observed differences in pesticides therefore require further investigations.
57	L169: Where can we find this information in the SI? I do not see any table reporting by transect point the concentration by substance, this would be interesting, also the sum concentration by type (fungicide, insecticide and herbicide).	*WE refer to the tables A2 and A3 in the SI, wher all data mentioned can be seen. We will add the raw data in a repository or make them available on request.*	yes	Tab. A2 gives an overview of detected substances, i.e. mean, sum, max, min etc.
58	L185: The canopy could act as a trap of substances brought by the wind, therefore high concentrations in soils near to forest areas is not surprising.	*Thank you for this suggestion. WE added some text and two references on fog in mountains. However the role fo trees in pestciide trapping is still to be investigated.*	yes	Text included: Potentially trees could lead to higher pesticide concentrations at ground level by trapping fog, that is known to contain high concentrations of pesticides (Glotfelty et al. 1987, Shunthirasingham et al. 2011). We did not compare forested closed canopy conditions with open habitat and the function of trees in CUP residue distribution awaits further investigation.
59	L188: You did not present the substances with the highest concentrations in soils…	*Thanks for this comment. The infomation was added.*	yes	we added: In soil, the insecticide methoxyfenozide was found most frequently in 21 (40 %) of the 53 samples in concentrations ranging from 0.36 - 10.70 µg kg-1, followed by the fungicides fluazinam with 13 (25 %, 0.53 - 3.69 µg kg-1) and trifloxystrobin with 8 (15 %, 0.41 - 3.30 µg kg-1) detections
60	L193: Do you have any information on the amount of those insecticides used? If one is not that volatile but widely used it is quite expected that we will find it everywhere…	*Thanks for this comment. We addded the only available use data for 2017 and compared our most common residues with the most frequently applied CUPs. Although this might change from year to year it nevertheless might give an indication that use is actually more important than phys chem properties. We also added a sentence in the conclusions.*	yes	in results: The only available use data for the region from 2017 (Baumert et al. 2023) reveal that etofenprox was among the most frequently applied CUPs with 13.695 applications, used in 89% of all agricultural operations, 332 kg used). Methoxyfenozid was applied 6.908 times in 48% of operations and 159 kg. The fungicides fluazinam (20.985 applications, used in 76% of agricultural operations, 1.703 kg used) and penconazole (30.060 applications, 85% of operations and 269 kg) were also heavily applied. However, trifloxystrobin was only applied 412 times in 4% of all operations and only 8 kg were used. When comparing our recorded residues with the use data from 2017 it becomes obvious that residues of frequently applied CUPs are also often recorded and widely distributed. The exception is trifloxystrobin, although it might be possible that this fungicide was applied more often in 2022. in conclusion: It also seems likely that frequently applied CUPs that are used in high volumes in many farms simply are recorded more often in teh landscape, independent of any physical-chemcial properties of the molecules.
61	L202: this is very important indeed!! To identify those substances that should not drift in the atmosphere!	*Thank you, the relevant CUPs are already mentioned in the sentence. We also consider this an important point.*	not applicable	
62	L210: What about the Koc to predict their persistence in soil? And in the atmosphere linked to the particles?	*We mention particles here : However, CUPs can be adsorbed to fine particles and then be distributed by air currents and winds over large distances (Degrendele et al. 2016, Unsworth et al., 1999. We added Kfoc values in teh SI and a short sentence regarding particles.*	yes	Text added: However, in our study Kfoc values of the detected CUPs (Si Tab. A2 & A3) showed no relationship with altitude and therefore are not a predictor for affinity to particles and transport to higher regions
63	L219: You never discuss the relationship with expected degradation time. Trifloxystrobin has a DT50 of 1 to 3 days and is reported in the highest transects, this is never pointed out in the discussion…Same thing for fluzinam, 21 to 34 days of DT50.	*Thank you. We added a paragraph on Dt50 values and their importance for the interpretation of CUP presence. We refer to the tables in the SI for values.*	yes	For the detections of CUPs half-lives are also of importance for their interpretation (SI Tab. A2 & A3). For the most frequently detected CUPs longer half-lives are known from soil field studies for the insecticide methoxyfenozide (39-133 days) and the fungicide penconazole (67-115 days). With most quantified concentrations, among them the highest with 1370,85 µg kg^{-1} in vegetation the methoxyfenozide detections are likely to be a result of this year's application, whereas the penconazole detections were mostly below LOQ (23 of 35 in vegetation and 5 of 7 in soil), and therefore residues are assumed to originate from applications in the previous year. The fungicide fluazinam with a DT_{50} range of 21-34 days was always detected in quantifiable concentrations and also the insecticide Etofenprox with a shorter DT_{50} range (7-25 days) was recorded in higher concentrations in the vegetation and recent applications are therefore likely. Transport from recent application is especially obvious in the case of the fungicide trifloxystrobin that has a reported half-live in soil of 1-3 days but was recorded in all but one vegetation and 13 soil samples.

64	L232 to 239: Many limitations question the real interest in doing PCA + PCR: first the absence of soil parameters, and the selection of only soils in meadows when a lot of forests are covering the valley as I mentioned before. Also, the wide differences between the groups of transects which are not disposed regularly. Specifically, between T5 and T7 separated by ~ 15km, the cumulative concentration of pesticides is multiplied by 4.5! With such differences, the final prediction will be influenced by the one very high concentration of almost 3000 µg/kg… This leads to predicted pesticides concentrations of 7000 µg/kg in the valley (Fig4A) when the real measurements of the 6 transect points inside this highly contaminated area do not go beyond 3000 µg/kg for the maximum and then get down to 700 and 800 µg/kg. The overestimation of the concentrations is quite obvious here and probably due to the PCA and PCR based on log concentration.	*We agree with the reviewers constructive criticism and highlighted the potential uncertainties associated with the PCR prediction, which is only meant to approximate or identify the general distribution patterns of CUPs and their concentrations, i.e. the maps should not be misinterpreted in a way that they provide exact, small-scale estimations about concentration profiles. A cautionary note is now added prominently at the end of the PCR results (L324ff) to prevent any overinerpretation of the results. We feel that hihglighting the limitations of this approachis important and also adresses similar cirticism from other reviewers.* *Additionally, we analyzed all predicted concentrations and found that only 0.4% of our predictions actually exceed the highest observed concentrations of appr. 3000 ug/kg, which appears reasonable given that it is highly improbable that we observed the absolute highest concentration in the entirety of the Vinschgau valley. Furthermore, only 3.0% of the predicted concentrations are above the second highest concentration of 776 ug/kg.* *We also removed the value of the highest predicted concentration of ca. 7000 ug/kg, due to the reasons outlined by the Reviewer.*	yes	
65	L248 to 260: You never question your model based on few points, predicting concentration in the middle of the valley where there are only five points! You need to be a bit critical especially when mentioning the concentration of 7000 µg/kg predicted when you found half of this concentration in the real samples!	*See above. We agree with the reviewer and added a cautionary note at the end of the discussions (see previous comment), highlighting the potential uncertainites.* *We also removed the part of the sentence referencing the highest predicted concentration as it may simply be a fragment of multiple spatial drivers interacting (see L.306ff)* *Additionally, we analyzed all predicted concentrations and found that only 0.4% of our predictions actually exceed the highest observed concentrations of appr. 3000 ug/kg.*	yes	
66	L257: Finally, you mention the spay season which may completely impacts your results! This has to be discussed with more hypothesis of why finding some substances might be related to the use in orchards just before your sampling…	*We added more information on the sppraying season based on available data from 2017.*	yes	
67	L278: Yes, but you did not discuss the Koc parameter that could be an indicator of the affinity to particles…	*We analysed if there is any relationship of Kfoc values with altitude, but there was none. We added this result in the text.*	yes	Text added: However, in our study Kfoc values of the detected CUPs (Si Tab. A2 & A3) showed no relationship with altitude and therefore are not a predictor for affinity to particles and transport to higher regions
	Reviewer 3			
68	I think this is an interesting and important paper. The authors present a well-structured manuscript on the assessment of landscape contamination with currently used pesticides (CUP) by analyzing soil and vegetation samples from different altitudes. The aim of this research was to better understand the fate of CUPs, their distribution and the magnitude of residues along altitudinal gradients. I believe this work provides new and important information towards this goal. The study design, the selection of two different sampling matrices, the selection of sampling points and transects, the sampling itself and its duration, the analytical methods as well as the statistical methods applied are appropriate and innovative.	*Thank you*	not applicable	
69	In my opinion, the selection of this well studied area is not a weakness of the study, but rather a strength, as the new data on pesticide residues along elevation transects complements previous research in this field and within this geographical area.	*Thank you, we also appreciate the possibility to discuss our finding with the results of other studies in the same area.*	not applicable	
70	To my knowledge, the submitted work is the first to present a more landscape-oriented approach, whereas in the past studies have mainly focused on specific sites such as sensitive areas. (playgrounds, official places, landscape conservation areas, high altitude places).	*Correct - as mentioned this is the third global study that uses multiple and regular transects for collection of environmental terrestial samples.*	not applicable	
71	Since even glaciers are contaminated with CUPs and old residues, this work again supports the hypothesis and findings of previous research that pesticide use is a landscape-wide problem, including the exposure of remote high alpine habitats and protected areas.	*Yes, this was the aim of the study and is the main conclusion*	not applicable	
72	With these results that the number and concentration of CUPs decreased with altitude and distance from agricultural application sites, the researchers confirmed previous findings that most of the detected CUPs originate from apple production and that distance to source is a major factor in CUP contamination.	*Yes we agree*	not applicable	
73	The results are not entirely new; however, it is very important to confirm previous findings, generate new monitoring data and combine them.	*Yes we agree*	not applicable	
74	This research closes the gap in pesticide residue monitoring between the lower valley and high-altitude measurements such as glaciers.	*This was our main aim, especially regharing the habitats along the slopes*	not applicable	

75	I like the prediction method with PCR, but there are some methodological issues that could lead to too homogeneous overestimation at the valley bottom and underestimation along the elevation transects of the prediction model.. The PCR is appropriate, but can be improved in its application and reporting. However, the current manuscript needs some minor revisions regarding the statistical analysis and the presentation/description of the analysis. It is not clear to me why soil and vegetation samples are combined for prediction, as this work demonstrates that they differ significantly in the number and concentration of CUPs.	*We agree with the constructive criticism of the reviewer and have highlighted the uncertainty associated with such PCR modelling approaches now prominently at the end of the discussion (L. 324ff). This was also added in response to similar remarks by Reviewer 1. Also, additional information regarding validation and statistical summaries (e.g. PCR summary statistics) have also been added to the SI to provide further context and additional details: e.g PCA factor loadings, explained variance, cos2 values and PCR reisudals, coefficients and graphics detailing PCR residuals as well as predicted vs. actual values.We combined soil and vegetataion data as tehy both have an influence on local biodiversity e.g. insects that develop in the soil and feed on vegetataion (see above). We added a separate graph on concentrations of vegetation only and by the simililarity it becomes clear that soil concentrations are lower.*	yes	Text added and additional figure with vegetation sum concentrations only produced for SI.
76	The authors present very interesting and new aspects for the field of CUP monitoring, therefore I recommend the acceptance of the paper after some revisions.	*Thanks*	not applicable	
77	Line 41: Why did the authors not mention measurements of terrestrial matrix vegetation/grass samples from previous studies conducted in the area? Unless there are arguments against it, I suggest mentioning at least one of the studies on vegetation samples:	*Thanks, we are aware of the studies and we mention them at a diffferent position but we now also included them now also here.*	yes	added vegetation (Cech et al. 2022, Linhardt et al. 2019. 2021)
78	Cech, R. et al. Pesticide drift mitigation measures appear to reduce contamination of non-agricultural areas, but hazards to humans and the environment remain. Sci Total Environ 158814 (2022) doi:10.1016/j.scitotenv.2022.158814.	*added*	yes	
79	Linhart, C. et al. Year-round pesticide contamination of public sites near intensively managed agricultural areas in South Tyrol. Environmental Sciences Europe 33, 1 (2021).	*added*	yes	
80	Linhart, C. et al. Pesticide contamination and associated risk factors at public playgrounds near intensively managed apple and wine orchards. Environmental Sciences Europe 31, 28 (2019).	*added*	yes	
81	Line 43: The need for long-term and comprehensive monitoring was mentioned earlier than 2021, already in Linhart et al. (2019 and 2021).	*agreed, also added in the conclusion section.*	yes	
82	Line 49: CUPs were detected in glacial streams near and in South Tyrol. In particular, glaciers north and south of the studied transects were investigated: Forni Glacier in the Ortles-Cevedale group and Giogo Alto Glacier in the Palla Bianca-Similaun group in Italy. I propose to include this reference and to compare the results: Rizzi, C., Finizio, A., Maggi, V. & Villa, S. Spatial-temporal analysis and risk characterization of pesticides in Alpine glacial streams. Environmental Pollution 248, 659–666 (2019).	*Thank you. We added the study. We already mentioned the work of Ferraio. A comparison is difficult as we did not analyse chlorpyrifos. This compound is alos not renewed sicne 2020 so was no applied in 2022.*	yes	While CUPs have already been monitored specifically in meltwater from glaciers (Ferrario et al., 2017, Rizzi et al. 2019),
83	Line 84: ISTAT data for 2019, recent data are not available?	*Unfortunately not, there is usually a time lag and data for 2022 will probabaly be published in 2025.*	not applicable	
84	Line 122: MRL exceedance was found in this area for the valley bottom (Linhart 2021). Linhart, C. et al. Year-round pesticide contamination of public sites near intensively managed agricultural areas in South Tyrol. Environmental Sciences Europe 33, 1 (2021).	*Thank you we added the reference and exceedances.*	yes	Exceedances were were in the Vinschgau in grass samples taken in the valley bottom that could represent e.g. salad grown in gardens (Linhart et al 2021)
85	- Structure of the article The main article presents the most important information about the study design and results. I like the short and straightforward format and the detailed information in the Supplementary Information for interested readers. However, for better readability, I recommend splitting the Results/Discussion section into at least two subheadings: e.g. 1) CUP Residues and Identification of Related Factors, 2) CUP Prediction	*Thanks - done*	yes	headings included
86	- Tables: All Tables are clear and present the important information very well. Good presentation of LOD and LOQ.	*Thanks*	not applicable	
87	- Results and discussion: Line 95-97: The sentence refers to the significant correlation of number and sum concentrations of CUPs with the proportion of apple orchards. However, the mentioned table (Table A1) does not show the results of the correlation. Please also refer to the results of the correlation.	*Spearman correlation parameters added*	yes	added: Spearman correlation test revealed the number of pesticides in soil (rs = 0.32, pS = 0.0099) and in vegetation (rs = 0.58, pS < 0.0001) as well as their sum concentrations (soil: rs = 0.29, pS = 0.0182, vegetation: rs = 0.44, pS = 0.0005) to be significantly positively correlated with percentual area used for apple cultivation.
88	Lines 102-114: I suggest a better order for the sentences. Line 102-106: The maximum number of residues detected per site is discussed. The next sentence jumps to the differences between soil and vegetation samples - which I think is a separate, very important result. Then, at line 112, the sentence jumps back to the multi-residue problem. The paragraph comparing soil and vegetation interrupts the discussion of multi-residue contamination. Please reorder and use a paragraph to separate these two findings (multi-residue results, comparison of soil and vegetation matrix) for better readability. The significant difference in the number and concentration of residues in soil and vegetation samples deserves its own section, in my opinion, as it is a very important result.	*in this paragraph we present and discuss the mixture CUP residues present in soil and vegetation. We added subheadings for the entire text to clarify and rewored and reorded some parts.*	yes	Subheading included
89	Line 130 The term "statistically" is not necessary. Delete.	*as it is a statistically significant result based on a test with the details in te menioned table we keep the statistically in for clarity.*	no	

90	Line 137: Figure 2 "Y-Axis": Continue the axis to 6, for transect #6. I suggest also adding an arrow to indicate the increase in elevation (0 m to 3000 m asl, or the highest sampling point).	*We thank the reviewer for their keen eye. This was a clipping error and has now been corrected. We also indicate the increase in altitute as suggested be the reviewer, although we do not provide exact m a.s.l. values as the height m a.s.l. between transect points is not always the same.* *Additionally, an alternative depiction of this figure is also added to the SI, highlighting the change throughout the Valley and the height transects.*	yes	Figures improverd and additionally figure added in SI
91	It may help to improve the figure to indicate the "x-axis" with a west to east arrow or valley downstream arrow.	*As noted with the previous comment, this change has been made.*	yes	
92	I assume the color scaling of the number of pesticides in soil and vegetation is the same? If so, it would be helpful to show the same numbering and number formatting on the color scale. Either digits for both scales or no digits at all. I understand that the log scales for soil and vegetation should be different.	*The reviewer is correct and we adjusted it accordingly.*	yes	
93	Line 151: The authors state that increasing altitude is not the only factor leading to a decrease in CUPs at higher altitudes. This is quite obvious for a dependent variable under different environmental conditions. It is not always a linear relationship; even linear statistical methods are used. Try to reformulate the first sentence in terms of the other environmental factors. Later, you will present a multivariate statistical method. This sentence weakens the later use of PCA and PCR.	*Thanks. Addressed, we refer in this sentence now to linear relationships.*	yes	The distribution of CUPs along the transects did not always follow a linar decline in numbers and sum concentrations with increasing altitude
94	Line 152: Seasonal factors: A previous study conducted in this valley identified seasonality as a factor in CUP contamination of non-target areas, please cite: Linhart, C. et al. Year-round pesticide contamination of public sites near intensively managed agricultural areas in South Tyrol. Environmental Sciences Europe 33, 1 (2021).	*Thank you for the suggestion. Reference and additional text added.*	yes	Previous research in the valley bottom show complex interactions between meteorological parameters and application patterns, with CUPs decreasing with increasing irradiance and distance from fields, but also influences of rainfall, wind and the proportion of apple orchards in the surrounding landscape (Linhart et al. 2019).
95	Line 154: A previous study identified the most important environmental factors on CUPs for the same valley (bottom only): CUPS decrease with increasing irradiance and distance. At higher altitudes, irradiance is also higher. The work identified rainfall, percentage of apple orchards and wind (from source to sample site) in combination with increasing distance as factors increasing CUPs on non-target areas. I strongly suggest that the results of this work be included in this paragraph. Compare Fig. 2 and Fig. 3 in Linhart, C. et al. Pesticide contamination and associated risk factors at public playgrounds near intensively managed apple and wine orchards. Environmental Sciences Europe 31, 28 (2019).	*See above, additional sentence was added*	yes	Previous research in the valley bottom show complex interactions between meteorological parameters and application patterns, with CUPs decreasing with increasing irradiance and distance from fields, but also influences of rainfall, wind and the proportion of apple orchards in the surrounding landscape (Linhart et al. 2019).
96	Line 157: Meteorological data for different points within the studied valley are available retrospectively from the regional hydrographic service: https://wetter.provinz.bz.it/download-messdaten.asp. I know it is a lot of work to incorporate different meteorological data into a model. Also, the choice of data resolution can be crucial. However, I strongly recommend including these data, as the quality of the work would be improved enormously. The PCR model would be much more realistic and valid. In its current state, the PCR model is, in my opinion, quite rough and overestimated by landscape use variables. The inclusion of temperature data, irradiance, wind and precipitation would improve the validity of the model. These factors have already been shown to be significant factors for CUP contamination. At this stage, the model depends mainly on distance, altitude and different land use variables, which, in my opinion, is not sufficient. The PCR with additional factors previously shown to be significant would result in a similar model, but with less even distribution of CUPs along the valley and elevation transects, reflecting a more realistic situation. CUPs could be even higher or lower than predicted by the actual PCR. So far, the model is most likely overestimated for the bottom valley and underestimated for the slope sites.I agree that the CUPs are likely to be homogeneously distributed in the atmosphere, but they do not precipitate or fall homogeneously, or are homogeneously displaced over non-target areas. If it is not possible to include the meteorological data during the revision of the paper, I suggest to state again that the model is likely to be over/underestimated and dominated by land use factors, since the former have been shown to be significant factors and are not included.	*We agree with the reviewer regarding the use of additional meteorological data and the rough state of the PCR.* *First, we further elaborated on the limitations of the PCR in L 325ff. and highlighted potential improvements for the future.* *Second, we investigated the suggested data, however, their use would require establishing a method of interpolating the suggested meteorological data (from the point measurements) to produce reliable aeral raster data, which we will not achieve in the short time period of the revision.* *However, as also noted by Reviewer 1, we now prominently highlight that future work should investigate the temporal changes of CUPs in the plant and soil matrices, as well as, using meteorological and geological data so that these temporally variable exposure profiles can be described more precisely. Hence, we explicitly stated that the current model iteration primarily reflects land-use and land-cover effects (as noted by the Reviewer), which can, however, be improved in the future via the inclusion of the data mentioned by Reviewer 1 and Reviewer 3.*	yes	we added a critical text on the PCR.
97	Line 166: Compare comment for appendix and "wind" section.	*We included the table SI Table A1 in the text-*	yes	To compare CUP distribution by wind onto north- and south-facing slopes, transects T3-T10 were established directly facing each other on opposite valley slopes (Fig. 1, SI Tab. A1).
98	Line 185: Very interesting that a site with closed canopy cover had a relatively high concentration of CUPs. Are there any additional findings on this? If so, please provide literature.	*This site was a meadow along a trail in a more forested area than all the other sites. We removed the closed as the canopy in hindsight was not really fully closed. We added some more information an text in regard to this finding, also referring to reviewer 2.*	yes	Text added: Potentially trees could lead to higher pesticide concentrations at ground level by trapping fog, that is known to contain high concentrations of pesticides (Glotfelty et al. 1987, Shunthirasingham et al. 2011). We did not compare forested closed canopy conditions with open habitat and the function of trees in CUP residue distribution awaits further investigation.

99	Line 238: Why are CUPs in soil and vegetation not separated for prediction by PCR analysis?	*See above. Soil and vegetation were combined to provide a contamination of the entire habitat as for example insects develop in soil and feed on vegetation*	yes	Text added and additional figure with vegetation sum concentrations only produced for SI.
100	Line 244: Figure 3: Why are transects T9 and T10 not covered by the predicted CUP residues and concentration using PCR? Figure A3: PCA Biplot: log CUP sum concentration of soil or vegetation or the sum of both?	*The reviewer is correct and we updated Figure 3 extending the predicted area so that T9 and T10 are also covered . In addition, we clarified the Figure caption as suggested by the Reviewer.*	yes	Figure corrected
101	The text of the main article and Figure 3 indicate that prediction for soil and vegetation samples are not analyzed separately. Why are these two variables, which are clearly described to differ significantly in number and concentration of CPUs, not distinguished for prediction analysis? Please clarify. I recommend separating the analysis for soil and vegetation.	*See above. For the reason of analyzing soil and plant matrices together, please refer to the comment above.* *However, we now also constructed a third model, predicting the concentration only in plant matrices, which was subsequently applied to the study area. This new figure can be found in the SI with additional tables and graphics detailing e.g. model accuarcy.*	yes	Text added and additional figure with vegetation sum concentrations only produced for SI.
102	Questions and recommendations about PCA and PCR: I recommend restricting the prediction of CPUs by PCR to vegetation samples, as they have a significantly higher contamination rate and therefore less zero inflation of the dependent variable, which would strengthen the PCR. A cluster description of the four clusters of grouped sites would also improve the PCA results and the selection of principal components would be clearer.	*As mentioned in the previous comment, a separate model for the prediction of concentrations in plant matrices is now added separately to the SI.* *A brief cluster description was added to the SI as suggested by the reviewer. The resulting clusters are primarily divided by the associated land-use in die direct vicinity, i.e. clusters (1) close to orchards in the valley floor with high contamination, (2) highly remote clusters on alpine slopes, (3) sites close to urban centers with higher amounts of agriculture and (4) intermediate cluster where aforementioned attributes are moderately expressed (SI L. 161 ff)*	yes	Text added and additional figure with vegetation sum concentrations only produced for SI.
103	The application of PCA regression analysis allowed the prediction of CUP contamination from the valley floor up to nearly 3000 m elevation.	*This is correct. Model prediction was only conducted for areas that shared similar spatial attributes with the acutal field samples, i.e. no predictions were made above 2500 m a.s.l. as the model was not parameterized for these heights and the combination of land-use / land-cover attributes present there.*	not applicable	
104	I like the PCA and PCR approach and don't see anything fundamentally wrong with the methodology. However, there are neglected uncertainties in the form of missing variables and cofactors that probably lead to a homogeneous overestimation of the predicted contamination of the valley bottom. The factors related to contamination of the valley's bottom vegetation are clearly presented and described in Linhart et al. 2019. The factors distance, number of apple orchards, rain, wind and irradiance were shown to contribute significantly to the amount of CUP contamination.	*We agree with the comment. We followed the suggestion of the reviewer and analysed e.g. distance to apple orhacrds in our study. Regarding the use of additional weather data: The available meteorological data was insufficient for matching definite weather attributes with individual transects based on preliminary research. In the study region, only four weather stations exist that were assigned to the 11. However, this rough classification does not allow for a detailed analysis of weather conditions that may contribute to the distribution, deposition and degradation of pesticides in mountains, e.g. precipitation rates at slopes (Daly et al., 2007b, Tremolada et al., 2008), temperature differences between altitudes (Daly et al., 2007a, Feltracco et al., 2022) and incoming solar radiation (Daly and Wania, 2005, Rani and Sud, 2005). Nonetheless, the lack of meteorological data is now highlighted in the added uncertainity discussion in L.325ff.*	yes	We added a critical sentence on the PCR approach.
105	I am missing at least actual weather data; data summarized over three to five days prior to sampling would significantly improve the PCR model and better reflect particle-bound transport.	*See above. We wanted to include high resolution weather data however that is not currently available for high alpine regions in which weather condictions can drastically change between alpine slopes and valley floor conditions*	yes	
106	Also the use of PCA is not new, it is a widely used and quite "old" method in ecology for dimension and factor reduction; nowadays criticized in ecology & environmental science because the selection of variables/predictors and later the selection of principal components does not depend on the response variable. PLS regression would be a better alternative. See: https://doi.org/10.4209/aaqr.2020.01.0001	*We agree with the Reviewer regarding the use of PCA/PCR being outdated to a certain extent. As Reviewer 1 has noted as well, future research should leverage more advanced approaches such as ensemble learning methods (e.g. random-forest-modeling) or PLS (as suggested by Reviewer 3) once a more abundant datasets become available to us. This potential for improvement has now been added to the uncertainty discussion and acknowledges the potential for future improvements.*	yes	We added a critical sentence on the PCR approach.
107	The use of PCR and prediction modeling of possible contamination is a very useful tool. However, there are three issues: Neglect of available cofactors (meteo data), unclear separation or merging of soil and vegetation data, number and concentration of CUPs could be combined by weighting the concentration by the number of CUPs. (cf. Linhart 2019).	*Please refer to the previous responses regarding the grouping of soil and vegetation samples, the new addition of a plant-specific model and the unavailability of detailed meteorological data.*	yes	We added a critical sentence on the PCR approach.
108	Highlight differences between soil and vegetation results. May highlight and discuss the differences between soil and vegetation samples. Why are the results sign. different? For example: Co-factors of soil samples are more complex than vegetation samples. Soil pH, soil microbiome, no irradiance, stable temperature,…	*Thank you for this suggestion. We added infromation on ystemic properties as well as a discussion of Dt50 values to the discussion*	yes	Discussion added.
109	Vegetation samples may be more in line with the CUP concept, as this matrix reflects current use, whereas soil or even sediment samples may also show the presence of previously used pesticides.	*True but we measure CUPs but not legacy pesticidesin soil. There presence is from previous or current application but limted in time by the regultion.*	not applicable	
110	Is PCR better at predicting soil or vegetation CUP contamination? Clarify what was done with the data, combination of CUPs in soil and vegetation?	*See above. Additiona information was added*	yes	
111	The PCR prediction model could be fitted and is a very good starting point for future research in this area. The inclusion of open-source meteorological data is recommended.	*Thank you very much, this fact has now been highlighted prominently in the final chapter of the discussion as an outlook (L. 325 ff)*	yes	

112	The PCR prediction is very interesting at this point, but contamination is not a static process, it is a very dynamic process, strongly influenced by environmental factors.	*We fully agree and included this opinion / outlook in the Results and Discussion and conclusion by (1) mentioning the treatmetn period in apples in Vinschgau and (2) that contamination profiles will most likely change both spatial and temporally, which requires thorough investigation in the future where the suggested meteorological data will most likely be an important new cofactor.*	yes	
113	I recommend using only the results of CUPs detected in vegetation samples for PCR. For two reasons: - Vegetation samples reflect CUPs better - Vegetation samples correlate better with elevation and distance. Or at least distinguish between soil and vegetation, as these matrices reflect different habitats and different levels of contamination.	*We would like to keep the combined figure in the maintext as many insects will be exposed via both matrices directly or indirectly, thus acting as exposure pathways. Still, we feel that the Reviewers comment is justified and merited, hence we now provide a seperate prediction for the concentrations in plants in the SI. We refrained from predicting soil concentrations seperately as the results may be too unreliable, in parts due to the zero-inflation that the Reviewr has already outlined.*	yes	
114	The discrepancies between the results of the CUPs along the altitude transect and the EFSA peer review conclusion on volatility and air transport are a very interesting and important finding. I think this could be highlighted more.	*Thanks we added a stronger sentence.*	yes	The use of various parameters currently used for the description if the CUP distribution potential therefore needs to be questioned and assessed more thoroughly.
115	- Appendix It is confusing that the Appendix is referred to as Appendix A. are there other documents? Appendix B? C? D?	*Changed we now refer directly to the pictures.*	yes	
116	Line 259 The section "Effect of Wind on Pesticide Distribution" is somewhat misleading and confusing. First, there is no mention of wind data being collected or presented. Second, the term "foehn" is mentioned, but the analysis described focuses on differences in pesticide residues as a function of slope orientation. Differences in pesticide residues on north- and south-facing slopes are possible due to this warm upwind, but the statistical test analyzes orientation and does not include wind data. This is somewhat confusing because the reader is expecting wind data. I suggest using the term "effect of slope orientation" instead of "effect of wind". The "foehn" wind could be an obvious influence, but since it is not measured, it should be mentioned afterwards as a probable cause in the interpretation. Data on wind speed or wind hours defined as "foehn" may lead to different results than presented. I suggest changing the terms (orientation instead of wind).	*Thank you, we use now slope orientation and delted effect of wind.*	yes	
117	Line 256 The authors refer to Tab. D13. However, Tab. D13 does not include weather in the main article or in the appendix. Please clarify.	*Thanks for spotting this. Adressed, new table numbering.*	yes	
118	Line 283 Expanded Discussion of Butterfly and CUP Presence. I think this is a very interesting part of the study and provides hypotheses for further research. Is there also species richness data from vegetation samples? If so, this data would complete the link between butterfly biodiversity - vegetation diversity - CUP residues.	*Unfortunately to our knowledge there is no vegetataion data. However , caterpillars could be directly exposed through oral exposure, we added a short prargraph.*	yes	

15th Nov 23

Dear Professor Brühl,

Your manuscript titled "Contaminated landscapes – Current Use Pesticides residues in soil and vegetation along altitudinal gradients in a European alpine valley" has now been seen by our reviewers, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewers. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

Please note that it may still be possible for your paper to be published before the end of 2023, but in order to do this we will need you to address these points as quickly as possible so that we can move forward with your paper.

EDITORIAL REQUESTS:

Please review our specific editorial comments and requests regarding your manuscript in the attached "Editorial Requests Table".

*****Please take care to match our formatting and policy requirements. We will check revised manuscript and return manuscripts that do not comply. Such requests will lead to delays. *****

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If you have any questions or concerns about any of our requests, please do not hesitate to contact me.

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Best regards,

Fiona Tang, PhD
Editorial Board Member
Communications Earth & Environment

Clare Davis, PhD
Senior Editor
Communications Earth & Environment

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REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

Please see the doc file attached

Reviewer #2 (Remarks to the Author):

Dear authors,

The revised manuscript on "Contaminated landscapes – Current Use Pesticides residues in soil and vegetation along altitudinal gradients in a European alpine valley" has been highly improved.
You answered all my specific comments and included it in the manuscript. I therefore suggest the publication.

Kind regards

Reviewer #3 (Remarks to the Author):

The authors have addressed all feedback and suggestions for improvement. Both PCA and PCR and PCR prediction are much clearer and the terminology has been corrected. The risk of "overestimation" has been discussed and made more realistic by including the forest in a version of the regression prediction model.
Overall, the submitted paper has been well reviewed and I recommend its publication.

One comment:
The pdf file has not been updated regarding the numbering of the supplementary figures. For example, line 329: SI Fig.XXXX (revised manuscript marked). Be sure to use the completely revised document for final publication.

Caroline Linhart

I previously reviewed the manuscript now titled “Contaminated landscapes – terrestrial measurement of Current Use Pesticides residues along altitudinal gradients in a European alpine valley”.

I thank the authors for addressing the major suggestions from the two Reviewers.

One aspect that must be fixed is that numbers are sometimes reported using “.” as a decimal separator, and other times it is used “,”. For example L141: “trifloxystrobin: mean vegetation = 20.88 µg kg-1, mean soil =0.88 µg kg-1” and L146 “methoxyfenozide 7,01 µg kg-1 soil 1370,85 µg kg-1 vegetation”.

As reiterated by the other reviewer, weather data are important. Gridded data (1 km resolution) would be available from the Copernicus ECMWF Climate Change Service at https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-single-levels?tab=form

Perhaps things have changed, but last year for example I did the following query:

##########

Product type:

Reanalysis

Variable:

10m u-component of wind, 10m v-component of wind, 2m dewpoint temperature, 2m temperature, Mean evaporation rate, Mean surface downward long-wave radiation flux, Mean surface downward short-wave radiation flux, Surface pressure, Total column rain water, Total precipitation

Year:

2021

Month:

June, July, August, September

Day:

01, 02, 03, 04, 05, 06, 07, 08, 09, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31

Time:

00:00, 01:00, 02:00, 03:00, 04:00, 05:00, 06:00, 07:00, 08:00, 09:00, 10:00, 11:00, 12:00, 13:00, 14:00, 15:00, 16:00, 17:00, 18:00, 19:00, 20:00, 21:00, 22:00, 23:00

Sub-region extraction:

North 44.65°, West 10.91°, South 44.63°, East 10.93°

Format:

GRIB

And used the script to read and plot the data in the R environment

```
################################################################################

### short test

# read data

 a <- brick("adaptor.mars.internal-1664288796.7315109-26445-12-e3483757-c886-4bea-b21d-af89426764bb.grib")

# convert into array

 a_arr<-as.array(a)

 rm(a)


# define temporal extent and resolution of query

 myxinter <- c("01.06.2021 00:00","30.09.2021 23:00")

 myxinter <- as.numeric( as.POSIXct(strptime(as.character(myxinter), format="%d.%m.%Y %H:%M",tz="UTC")) )

 myxtick <- seq(myxinter[1], myxinter[2], by = 3600 )

 myxlab <- as.character(as.POSIXct(myxtick, origin="1970-01-01", tz="UTC"))

 myxlab_pos <- as.POSIXct(myxtick, origin="1970-01-01", tz="UTC")


# define name vaiables selected in the form

 name_var <- c("u_wind_10m", "v_wind_10m", "Td_2m", "T_2m", "Mean_E", "Mean_LW", "Mean_SW", "Surface_pressure", "Total_column_rain", "Total_precipitation")

 n_var <- length(name_var)


# everything as a data.frame for convenience

 mydf <- as.data.frame(matrix(NA, nrow=length(myxlab), ncol=2+ length(name_var)))

 names(mydf) <- c("Date","Date_pos",name_var)

 mydf$Date <- myxlab

 mydf$Date_pos <- myxlab_pos # myxtick
```

```
 for (i in 1:length(name_var)) {
  mydf[, 2+i] <- a_arr[1,1,seq(i,length(a_arr[1,1,]), by=length(name_var))]
 }

 mydf$Td_2m <- mydf$Td_2m - 273.15 # Kelvin to °C
 mydf$T_2m <- mydf$T_2m - 273.15 # Kelvin to °C
 plot(myxlab_pos , mydf$T_2m, type="s")

 mydf$wind_10m <- sqrt(mydf$u_wind_10m^2 + mydf$v_wind_10m^2) # combine wind direction
 plot(myxlab_pos , mydf$wind_10m, type="s")

 mydf$Surface_pressure <- mydf$Surface_pressure / 100 # Pa to hPa; 1 hPa = 1 mb
 plot(myxlab_pos , mydf$Surface_pressure, type="s")

 mydf$Total_precipitation <- mydf$Total_precipitation * 1000 # m to mm
 plot(myxlab_pos , mydf$Total_precipitation, type="s")
##############################################################################
```

Perhaps the authors will make use of the above information in their future studies.

Best regards
Daniele la Cecilia