We would like to thank the reviewers for their thoughtful reading and comments, which helped to improve this manuscript. We have addressed all comments below and have indicated the corresponding modifications in the revised version attached below.

Anonymous referee 1:

Overall:

This manuscript reported multi-annual observations of CUPs and OCPs from a monitoring site in Czech Republic. Active air samples have been collected biweekly over 3 years (CUPs) and 10 years (OCPs) and temporal trends were analysed. From the results, potential sources for the atmospheric presence of pesticides were indicated. Both OCPs and CUPs showed declining trends. The OCPs showed less decrease in the sampling period compared to the CUPs. Recently banned CUPs showed the lowest half-lives reflecting the immediate effects of legislation, but their presence may imply that they are persistent. While there are indications that the levels of OCPs are affected by secondary sources, CUP concentrations seem to correlate with application seasons.

Overall, this manuscript presents novel and important results, which has been sufficiently documented. The manuscript includes a great deal of information, and additional information is recorded in the SI. The approach and data help to advance the field of science. I would therefore suggest it can be accepted after some revision. In addition to the comments below, some parts need rephrasing/elaboration which is commented directly in the attached pdf of the preprint.

Specific comments:

Section 1 Introduction:

1. *The reference (UNEP, 2001) cannot be found in the list and a more precise reference to the Stockholm convention should be given.*

   Thank you for your comment. To clarify the reference, we will update the reference list to refer to the proper reference of the Stockholm Convention official website.

2. *Line 40: Levelling off air concentrations over time may imply that secondary sources are likely to influence to a larger extent, but how can conclusions regarding sources be made from time-trends by itself? I think this statement needs some moderation /elaboration or a reference.*

   Thank you for your comment. The statement should have been more specific in mentioning that we consider seasonal patterns, and specifically the relationship between temperature peaks and pesticide concentration peaks to be an indication of either fresh application (primary sources) or revolatilization (secondary sources). So, to ensure that this idea is also expressed in the introduction, we will modify the sentence, to read as (lines 43-46):

   > "For both banned and authorised chemicals, it is generally possible to distinguish between primary (e.g., application for pesticides) and secondary sources (e.g., re-volatilisation) by examining the seasonal variations of temperature, application and concentrations in air (Hoff et al., 1998; van den Berg et al., 1999)."

Section 2 Methods:

For the OCPs, there were pointed at some methodological differences between the dataset in terms of recovery corrections. This gives rise to two different time trend curves. The half lifes for the two periods were then compared. This seems to be a reasonable way to account for the differences. The analytical methods are well-described with satisfactory measures to assure high quality of the data, but I have a few comments:

3. *The QFF and PUF/XAD samples have been analyzed separately, but the total concentration is used in further assessments. It should therefore be described how the results were combined. Furthermore, the recoveries given in SI (Table S7) should not only be indicated separately for QFF and PUF/XAD, but also for the two combined.*

   Thank you for your comment. To provide more clarity, we expanded the sentence (line 121), to describe how the results were combined. As QFF and PUF/XAD were extracted separately, no recoveries for QFF and PUF/XAD together were determined. An effective recovery could be derived based on the phase mass-weighted mean of the recoveries of the particulate (QFF) and gas (PUF/XAD) phases. The revised sentence will read (lines 130-131, in section 2.6):

   > "As relaxation to phase equilibrium is fast on the time scale addressed (weeks), and both phases were analysed separately, the data analysed was the total concentration i.e., sum of particulate and gaseous concentrations."

4. *For the OCPs, it seems like only 7 out of 30 have internal standards. This may give rise to higher uncertainty in data for the other OCPs and should be specified.*

   Thank you for your comment. It is true that we do not have C13-labelled structural analogues for all OCPs, but in most cases this should lead to limited uncertainty because the internal standard is an isomeric or structurally similar compound, e.g., γ-HCH is used as internal standard for 5 HCH isomers (Table S5). In the cases of the chlorobenzenes, HCHs and DDT compounds, we do not expect the internal standard to introduce uncertainty. However, the reviewer is correct in the case of the second set of pesticides (heptachlors, chlordanes, endosulfans, and drin pesticides); in this case ^{13}C-α-endosulfan was used as an internal standard except for β-endosulfan, for which ^{13}C β-endosulfan was used as internal standard. This may introduce additional uncertainty for a subset of these OCPs. We will add a sentence about uncertainties in the SI when different methods are described (SI, lines 61-64):

   > "As isotopically labelled internal standards were not available for all 17 additional OCPs analysed, ^{13}C-α-endosulfan was used as an internal standard for all compounds (Table S5a and S5b; except for β-endosulfan for which ^{13}C β-endosulfan was used), which may lead to additional uncertainties in expressed concentrations, which are expected to be minor."

5. *Under section 2.5 it is not clear if the recoveries given account for both OCPs and CUPs, and for all samples/time periods, incl. 2013-2017. A reference to Table S7 is also missing. Furthermore, it is not clear if internal standards for all OCPs were introduced in 2018 (line 115). This part could be elaborated.*

   Thank for pointing this out, we will add the reference to table S7 (line 123), as well as extra information to the sentence to fill this gap, to read (lines 119-123):

> "For the 48 CUPs analysed using the HPLC-MS/MS, the method recoveries of individual analytes ranged from (68 ± 14)% (carbaryl) to (153 ± 22)% (iprovalicarb) for QFFs and from (61 ± 3)% (kresoxim-methyl) to (132 ± 10)% (iprovalicarb) for sandwiches (Table S7a), while for OCPs, recoveries ranged from (47 ± 8)% (PeCB) to (100 ± 9)% (*p,p'*-DDD) for QFFs and from (49 ± 6)% (PeCB) to (103 ± 10)% (*p,p'*-DDD) for all the samples from 2013 to 2022 (Table S7b)."

6. *Under 2.6 the term "timing" is not clear and a further explanation is needed.*

   Thank you for comment. In order to clarify the sentence, we will remove the words "timing" and replace them with the more appropriate and precise terms "periodicity" and "application periodicty" (lines 140 and 145).

Section 3 Results:

7. *Line 190-195: Why is 100 pg/m3 used as a limit for CUPs? For both CUPs and OCPs, it should be stated explicitly which compounds that were found in highest concentrations and their concentrations should be specified, e.g. the CUPs Metolachlor, Chlorpyrifos and Pendimethalin, and the OCPs HCB, ppDDE and gHCH. Methoxychlor should also be mentioned. It should explicitly be specified that the concentrations of OCPs are significantly lower than CUPs.*

   Thank you for the comment. We have added the additional information asked for CUPs, as well as explained why 100 pg m^{-3} matter in the context of our study in relation to the previous one done at this site. Will read (line 200-207):

   > "Previous observation at this site during the 2012-2013 period, showcased the observation of chlorpyrifos, metazachlor and s-metolachlor, in addition to acetochlor and isoproturon in concentration surpassing 100 pg m^{-3} (Degrendele et al., 2016). In this study, chlorpyrifos, s-metolachlor, and prosulfocarb were found most abundant (average concentrations of 116, 115, and 79.7 pg m^{-3}, respectively; Table S10a). (…), Similarly, high concentrations of these compounds had previously been reported from rural environments (Debler et al., 2024; Habran et al., 2024; Mayer et al., 2024; Ni et al., 2024)."

   Similarly, the suggested statement comparing OCPs and CUPs concentration has been added. As well as, a clear statement about HCB, *p,p'*-DDE and *γ*-HCH being the OCPs found in the highest atmospheric concentrations (line 211-2215):

   > "Overall, concentration of OCPs were found to be significantly lower than CUP concentrations. The average weekly concentration of Σ_{30}OCPs was 44.3 pg m^{-3}, with HCB, *p,p'*-DDE and *γ*-HCH accounting on average for 38, 29 and 8.1% of Σ_{30}OCPs and found at highest concentrations (Figure 1c,d and Table S10)."

8. *For both OCPs and CUPs the comparison with other studies should be elaborated, i.e. how do the results align with previous findings, e.g passive air studies? Also, a reference to Table S9 and compliance with usage should be included.*

Thank you for your question. We believe the discussion covered other studies for CUPs completely in the original manuscript (lines 192-197), but indeed not so for OCPs (as there were way more). So, we will add references to recent OCPs studies where similar results were observed (lines 2213-214), will read:

> “These three OCPs have also been previously observed as the dominant atmospheric OCPs (Gevao et al., 2018; Wang et al., 2018; Wong et al., 2021; Mamontova et al., 2022; Khuman et al., 2023; Lunder-Halvorsen et al. 2023).”
>
> Additional references:
>
> Gevao, B., Porcelli, M., Rajagopalan, S., Krishnan, D., Martinez-Guijarro, K., Alshemmari, H., Bahloul, M., and Zafar, J.: Spatial and temporal variations in the atmospheric concentrations of “Stockholm Convention” organochlorine pesticides in Kuwait, Sci. Total Environ., 622–623, 1621–1629, 2018.
>
> Khuman, S. N., Park, M. K., Kim, H. J., Hwang, S. M., Lee, C. H., and Choi, S. D.: Nationwide assessment of atmospheric organochlorine pesticides over a decade during 2008–2017 in South Korea, Sci. Total Environ., 877, 162927, 2023.
>
> Lunder Halvorsen, H., Bohlin-Nizzetto, P., Eckhardt, S., Gusev, A., Moeckel, C., Shatalov, V., Skogeng, L. P., and Breivik, K.: Spatial variability and temporal changes of POPs in European background air, Atmos. Environ., 299, 2023.
>
> Mamontova, E. A. and Mamontov, A. A.: Air monitoring of polychlorinated biphenyls and organochlorine pesticides in Eastern Siberia: Levels, temporal trends, and risk assessment, Atmosphere (Basel), 13, 1971, 2022.
>
> Wang, C., Wang, X., Gong, P., and Yao, T.: Long-term trends of atmospheric organochlorine pollutants and polycyclic aromatic hydrocarbons over the southeastern Tibetan Plateau, Sci. Total Environ., 624, 241–249, 2018.
>
> Wong, F., Hung, H., Dryfhout-Clark, H., Aas, W., Bohlin-Nizzetto, P., Breivik, K., Mastromonaco, M. N., Lundén, E. B., Ólafsdóttir, K., Sigurðsson, Á., Vorkamp, K., Bossi, R., Skov, H., Hakola, H., Barresi, E., Sverko, E., Fellin, P., Li, H., Vlasenko, A., Zapevalov, M., Samsonov, D., and Wilson, S.: Time trends of persistent organic pollutants (POPs) and chemicals of emerging Arctic concern (CEAC) in Arctic air from 25 years of monitoring, Sci. Total Environ., 775, 145109, 2021.

Comparison to passive sampler studies are discussed in relation to multi-annual variation in the corresponding. For concentrations however, only comparison to studies done with active samplers were discussed due to the uncertainties associated with recalculation of concentration from passive sampling. A reference to Table S9 will be added to section 3.2 to discuss high atmospheric concentration and national usage, will read: (lines 203-206):

> “As mentioned previously, some of those compounds (i.e., chlorpyrifos, metazachlor, pendimethalin, spiroxamine, tebuconazole and terbuthylazine) were applied on the Czech territory in more than 50 t of active substance per year, which would influence their high atmospheric concentration, notably during application periods (Table S9).”

9. *Line 200-205: The interpretation of DDT ratios is unprecise. Low ppDDT compared to ppDDE/ppDDD is due to degradation of DDT to DDE and indicate possible shift from primary to secondary sources, i.e past usage of technical DDT mixture. Pozo et al (2006) Environ. Sci. Technol. 2006, 40, 4867-4873 should be referenced. Also, the interpretation of HCH ratios should be elaborated. Note that Lindane contains >99% y-HCH (ref. Y.F. Li, R.W. Macdonald / Science of the Total Environment 342 (2005) 87–106). Furthermore, source indications of both DDTs and HCHs should be compared with other studies in Europe assessing sources (e.g Lunder Halvorsen et al 2023, PAS Europe 2016).*

   Thank you for the suggestion, we believe this idea was already expressed in the original manuscript. The indicator ppDDT/(pDDE+ppDDD), to our knowledge, was first used by Rapaport et al., 1985, and we will therefore add a reference to a review, Bidleman 1999, at the end of our original statement. In addition, the information regarding lindane and corresponding reference will be added (lines 226-228) and will read:

   > "Additionally, the ratio β-HCH/(α-HCH+γ-HCH) which ranged 0.01-0.16 can be used to distinguish between technical HCH (β-HCH/(α-HCH + γ-HCH) ≥ 0.5) and lindane (β-HCH/(α-HCH + γ-HCH) < 0.5) (Li and Macdonald, 2005), as sources of environmental contamination."

   In addition, source indication of HCHs were elaborated upon based on the suggested references (lines 231-236), will read:

   > "Similarly, the ratio α-HCH/γ-HCH has been previously used to infer sources of HCHs. A recent study in Europe highlighted a distinction between the northern and southern regions: in the north, high α-HCH/γ-HCH values were observed, indicating the dominance of long-range atmospheric transport and re-suspension of α-HCH, whereas in the south, lower α-HCH/γ-HCH values suggested historical use of γ-HCH (lindane; Lunder-Halvorsen et al., 2023). In our study, the ratio α-HCH/γ-HCH ranged from 0.12 to 1.7, aligning with values reported in southern Europe."

   Additional references:

   Bidleman, T. F.: Atmospheric transport and air-surface exchange of pesticides, in: Fate of Pesticides in the Atmosphere: Implications for Environmental Risk Assessment (van Dijk, H. F. G., van Pul, W. A. J., and de Voogt, P., eds.), Springer, Dordrecht, Netherlands, pp. 115–166, 1999.

   Li, Y. F. and Macdonald, R. W.: Sources and pathways of selected organochlorine pesticides to the Arctic and the effect of pathway divergence on HCH trends in biota: a review, Sci. Total Environ. 342, 87-106, 2005.

10. *Figure 1: The X-axis with months is not very consistent and it should be easier to see where the application/spring seasons are. Metolachlor and Chlorpyrifos should also be shown or mentioned in section 3.3 given their high concentrations.*

    Thank you for your suggestion. We will update the X-axis for Figure 1 to keep a consistent base for the month for CUPs and for OCPs. High concentrations of chlorpyrifos and s-metolachlor have been mentioned and discussed in section 3.2 total concentration, while seasonal variation and application period are discussed in section 3.3, where s-metolachlor, although not mentioned by name specifically, is part of the 16 compounds with peak application in spring (Table S12).

11. *Temperature dependence (line 230): An overview of temperature during the sampling period is missing and should be included e.g. in Figure S6. How is the temperature dependence and can a peak be expected from volatilisation by itself?*

    Thank you for your suggestion, we will add a sentence at the beginning of section "3.4 Influence of temperature on pesticide revolatilisation" reading (lines 274-275):

    > "For the 2013-2022 OCPs collection period, the temperature ranged from -9.5 to 23.7 °C while for the 2019-2021 CUPs collection period, it ranged from -5.8 to 22.1°C."

    The conclusion regarding the temperature dependence and whether a peak can be expected from volatilisation alone is discussed and answered in section "3.4 Influence of temperature on pesticide revolatilisation", as mentioned in lines 296-299 of the version published as preprint.

    > "For OCPs, temperature dependent volatilisation is the main influence on OCP atmospheric concentration. For authorised CUPs, atmospheric concentrations were mainly influenced by application, while temperature-dependent resuspension and LRAT determined CUPs atmospheric levels for banned compounds."

    This statement follows the discussion of the results from examining the Clausius-Clapeyron equation for OCPs and CUPs. It, indeed, concludes that atmospheric concentration for OCPs and some CUPs are temperature-dependent, and a seasonal peak can be due to elevated temperature, especially for OCPs.

12. *For the OCPs, the Clausius-Clapeyron equation is used to look into re-volatilisation. However, it is not clear to me how good correlation between partial pressure and the inverse ambient temperature implies that air concentrations and temperature are correlated, and this should therefore be elaborated. Correlation between air concentrations and temperature may be more intuitive and align better with Figure S6. In that case, R2 and p-values should be added to the figure.*

    Thank you for the suggestion, we will add the R^2 and p-value from Spearman correlation to selected chemicals presented in Figure S6, and update the figure caption, to show the relationship between atmospheric concentration and temperature.

13. *Line 270: How do the results of this study imply that the atmospheric concentrations of banned CUPs are dominated by resuspension and LRAT? This claim needs further explanation.*

    Thank you for your comment and question. Since for the Czech Republic, information about the amount of active substances applied yearly is available, it is possible to specify whether the level encountered in the atmosphere is due to primary emissions only. For banned pesticides, obviously this is not the case: Therefore, OCP levels encountered are due to resuspension from other media (e.g., soil) or long-range atmospheric transport and individual compound persistence. Whereas for authorized CUPs, levels are generally more influenced by seasonal applications. This is explicitly described in the version of the manuscript published as preprint (lines 268-270).

14. *Section 3.5: I question the assessment of time trends for OCPs/CUPs down to 20% detection frequency, as it is a possibility for bias due to MDLs. I would therefore suggest to use 80% as cut-off instead.*

    Thank you for the comment and suggestion. We agree that 20% is a relatively low detection frequency threshold, but we have specifically selected this as relevant for pesticides, given their strong seasonality. Many pesticides are detected only in summer and/or around time of application and applying a threshold such as 80% would lead to the exclusion of substantial amounts of valuable data.

15. *The discussion of time trends for CUPs should highlight Chlorpyrifos given the decline in usage (Table S9) and high concentrations. The comparison with Fenpropimorph (line 290) is interesting, given that the use of Chlorpyrifos has been more drastically reduced, while the half-life for Fenpropimorph is lower. Time trends for Metolachlor and Pendimethalin should also be considered given their dominance in the atmosphere.*

    Thank you for your suggestion, we will replace terbuthylazine by s-metolachlor in Figure 2 showcasing the significantly decreasing trends for selected CUPs. As pendimethalin trend was not significant, it has not been included in figure 2. However, results for all CUPs trends are presented in Tables S14 and S15.

16. *Figure 2: Not easy to see purple line for the OCPs and I therefore suggest to change to a darker color.*

    Thank you for your suggestion, we will update the figure and caption accordingly. We decided to go with a brighter color (red) which adds to trend visibility.

17. *Line 300: The steepest slope is found for a-Endosulfan in accordance with regulation from 2013. Data for b-Endosulfan could have given valuable information regarding past/present usage. However, the number of compounds in the study is already extensive and it is understandable that not all isomers can be included in the instrumental analysis.*

    Thank you for your comment. Indeed, it would have brought forth valuable information. *β*-endosulfan was included in the instrumental analysis, but only found in 6.3 % of the samples (Table S8), which we judge inconclusive with regard to time trends.

18. *Line 315: Time trends are compared with data extracted from EBAS, but it should be specified in SI how these trends have been developed. Also, HCB trends are specifically mentioned in EMEP status report 2/2022, which should be referenced.*

    Thank you for the suggestion, we will add a reference to the EMEP status report 2/2022. The reference to the EBAS database will be dropped, as the data available on that website is the same as the one used in references already made (i.e., Ilyin et al., 2022; Lunder-Halvorsen et al., 2023; UNEP 2023).

19. *Line 300: Time trends of 12 OCPs indicate that the concentrations are levelling off, but to a less extent compared to the CUPs (i.e. higher half-life). This should be highlighted.*

    Thank you for the suggestion, an additional sentence will be added (line 337-338):

    > "Compared to CUPs with insignificant trends, these 12 OCPs levelled-off on a shorter time scale (i.e., longer half-life than for CUPs), highlighting the persistence of these compounds."

Section 4 Conclusion:

20. *While Kosetice is a regional background site, it is pointed out in section 2.2 that local sources exist for pesticides. Generalization and the impact of this study should therefore be moderated, given that these data are based on one rural site only and variations may exist within central Europe.*

    Thank you for your comment, indeed, NAOK is considered a regional background site in the context of air quality studies and research, which includes also some organic pollutants such as PAHs, but, as mentioned in section 2.2, NAOK is not (and never was) a background site in the context of pesticides studies and research. This article aims to bring forth data and interpretation of a time series of atmospheric CUPs from a site located in a source area. To make sure that we are showcasing the results from one central European agricultural site only, we will add a corresponding statement (line 385-386), which will read:

    > "As this study focuses on a single agricultural site, the findings cannot capture the variability across the entire CUP source area i.e., rural central Europe. However, the observations were consistent…"

Anonymous referee 2:

This study presents seasonal and multi-annual trends of organic pesticides at a rural agricultural site in Central Europe and investigates their primary and secondary sources. Over ten years of weekly air monitoring for a wide range of pesticides (48 CUPs and 30 OCPs) has produced a large dataset that enables a comprehensive assessment of atmospheric pesticide contamination in Central Europe and allows for the determination of seasonal and temporal trends with high confidence. The statistical analyses appear sound, and the resulting trends generally align with known usage patterns, regulatory restrictions, and dominant release pathways. However, some statements should be clarified, and the language could be improved for better readability. Detailed comments are provided below.

Detailed comments

(1) *The application of two separate equations for OCPs and CUPs is scientifically reasonable, but the manuscript would benefit from a clearer justification of why different models were chosen, especially for readers less familiar with OCP or CUP-specific seasonality patterns.*

Thank you for your comment. We have added a sentence in the methodology to clarify the use of different time functions to model the observations, now reading (Lines 144-145):

> "Since at rural sites, CUPs in air will be dominated by primary emissions (i.e., application on agricultural land), long-term trends were analysed using Eq. (2), which captures up to two annual amplitudes and their application periodicity."

(2) *The authors employed HPLC, GC, and tandem mass spectrometry (MS/MS) to analyze the collected samples and identify OCPs and CUPs. However, both OCPs and CUPs can naturally degrade or transform under specific environmental conditions, potentially forming structurally similar degradation products. Therefore, the authors should discuss how they distinguish between parent compounds and their degradation/metabolic products. Is it possible that the parent compounds could be potentially interfered by their degradation/metabolic products during the analysis?*

Thank you for your comment. For CUPs, the qualification and quantification of peaks was done using isotope dilution methods, which use deuterated version of the compounds of interest to ascertain their presence and properly discern their levels. For OCPs, the method employed are accredited (SOP-LSA-055; EPA- MethodTO-13A) based on inter-laboratory comparison with the standardisation agency and therefore we are sure that no interference occurred from degradation/metabolic products during the analysis.

(3) *Title: I am not entirely sure if the modifier "unidentified" is necessary. I think the secondary sources primarily refer to volatilization from land and water surfaces; The "current-use and organochlorine pesticides" can be refined as "legacy and current-use pesticides". Consider a refined title of something like "Multi-annual trends of legacy and current-use pesticides in air in Central Europe: primary and secondary sources."*

Thanks for this discussion. Identification of sources vs. of source types was indeed not clear. Even so many studies have pointed to re-volatilisation as the main secondary source of banned OCPs, we are still far from having these sources identified: Neither the contributions of the various types of surfaces (such as urban, agricultural and natural soils and canopies, surface waters) nor the geographic location of the source areas has commonly been characterised on the appropriate spatial scale i.e., regional to global. Moreover, current understanding of release processes is deficient as often limited to few case studies (Bogdal et al., 2009; Nizzetto et al., 2010, Christensen, 2024), such that current knowledge cannot assess the significance of episodic mobilisation from surface compartments by climate events (floods, large-scale biomass burning, melting cryosphere) for the long-term trends in air. As to the summer maximum, note, that co-variation of local air temperature with pesticide concentration in air may indicate local sources (Hoff et al., 1998), but could also be explained by sources located upwind under co-varying temperature, such as the larger region or even the entire climate zone. This would be indistinguishable, the lower the time resolution of the OCP (ours is 2 weeks only), the more. Potential source areas for HCB, γ-HCH and DDT at our site were mapped within ca. 1000 km based on trajectory analysis (Dvorská et al., 2009). Using a global chemistry-transport model with air-soil exchange parameterisations for land and water surfaces, seasonal HCH and DDT secondary source areas were mapped and identified as regional and even global scale phenomena (Semeena & Lammel, 2005; Semeena et al., 2006). We will extend the discussion of results accordingly, include the conclusion, and prefer to keep the ‘unidentified’ in the title.

Additionally, we agree that the word “legacy” in the title is more concise than “organochlorine”. The (i) new title and the (ii) extended discussion text and (iii) conclusion section will read (Title header; lines 364-379; lines 394-398):

> (i) “Current-use and legacy pesticides’ multi-annual trends in air in central Europe: primary and unidentified secondary sources”
>
> (ii) “In general, the atmospheric levels of banned OCPs could be sustained by reversal of the direction of air-surface exchanges driven by historic contamination and chemical equilibria (Bidleman et al., 1995; Mackay and Parnis, 2020) or by mobilisation from surface compartments driven by climate events, such as melting of glaciers, permafrost soils or polar ice, flooding or heating of soils by wildfires (Holoubek et al., 2007; Bogdal et al., 2009; Nadal et al., 2015; Jiang et al., 2023). However, for only few OCPs (HCH, DDT) historic primary emissions, needed for modelling the large-scale chemodynamics are available (Hansen et al., 2004; Semeena and Lammel, 2005; Stemmler and Lammel, 2009; Wöhrnschimmel et al., 2012) and mobilisation processes have hardly been described beyond case studies (Bogdal et al., 2009; Nizzetto et al., 2010; Christensen, 2024). In recent years, the influence of such mobilisation processes on OCP cycling in the study region has not become evident, but cannot be excluded, considering on-going climate change with global impacts. Note, that the pesticide concentration in air summer maximum may indicate local sources (Hoff et al., 1998; Wania et al., 1998), but could also be explained by sources located upwind under co-varying temperature, such as the larger region (e.g., Zhan et al., 2024). Using a global chemistry-transport model with air-soil exchange parameterisations for land and water surfaces, seasonal HCH and DDT secondary source areas were mapped and identified as regional and global

scale phenomena (Semeena and Lammel, 2005; Semeena et al., 2006). In conclusion, current knowledge cannot assess the significance of these two types of secondary sources for long-term trends in the region, hence, cannot identify the sources, even not for HCH and DDT."

(iii) "…levelling off of these OCPs results from enhanced secondary sources, i.e., recent reversal of the direction of air-surface exchange or recent mobilisation of their reservoirs in soils, water bodies or the cryosphere. Current knowledge cannot assess the significance of these two types of secondary sources or locate their distributions."

Related additional references are:

Christensen, K.: Thawing permafrost releases industrial contaminants into Arctic communities. Environ. Health Persp., 132, 032001, 2024.

Dvorská A., Lammel G., Holoubek I.: Recent trends of persistent organic pollutants in air in Europe - Air monitoring in combination with air mass trajectory statistics as a tool to study the effectivity of regional chemical policy. Atmos. Environ., 43, 1280-1287, 2009.

Hansen, K. M., Christensen, J. H., Brandt, J., Frohn, L. M., Geels, C.: Modelling atmospheric transport of persistent organic pollutants in the northern hemisphere with a three-dimensional dynamic model: DEHM-POP, Atmos. Chem. Phys., 4, 1125–1137, 2004.

Jiang, H.Y., Li, J., Zhang, R.J., Pansak, W., Zhong, G.C., Li, K.C., Zhao, S.Z., Bualert, S., Phewnil, O., Zhang, G.: Mapping the contribution of biomass burning to persistent organic pollutants in the air of the Indo-China Peninsula based on a passive air monitoring network. Environmental Science & Technology, 57, 2274-2285, 2023.

Nizzetto, L.,MacLeod, M., Borgå, K., Cabrerizo, A., Dachs, J., di Guardo, A., Ghirardello, D., Hansen, K.M., Jarvis, A., Lindroth, A., Ludwig, B., Monteith, D., Perlinger, J.A., Scheringer, M., Schwendenmann, L., Semple, K.T., Wick, L.Y., Zhang, G., Jones, K.C.: Past, present, and future controls on levels of persistent organic pollutants in the global environment - Understanding the legacy of persistent organic pollutants requires studying the transition from primary to secondary source control. Environ. Sci. Technol., 44, 6526–6531, 2010.

Semeena V.S., Lammel G.: The significance of the grasshopper effect on the atmospheric distribution of persistent organic substances, Geophys. Res. Lett., 32, L07804, 2005.

Zhan, F.Q., Li, Y.N., Shunthirasingham, C., Oh, J., Lei, Y.D., Lu, Z., ben Chaaben, A., Lee, K., Gobas, F.A.P.C., Hung, H., Breivik, K., Wania, F.: Archetypes of spatial concentration variability of organic contaminants in the atmosphere: Implications for identifying sources and mapping the gaseous outdoor inhalation exposome. Environ. Sci. Technol., 58, 18273−18283, 2024.

(4) Line 12, "with revolatilisation from soil apparent in summer." For OCPs, they can not only be volatilized from soil but also from water bodies, in some cases, e.g. HCH, volatilization from water is more pronounced compared to direct volatilization from soil. (https://doi.org/10.1021/acs.est.4c05204; https://doi.org/10.1021/es034998k)

Thank you for spotting it. We corrected it.

(5) Line 16: "reversal of the direction of air-surface exchange or recent mobilisation from soils, water bodies, or the cryosphere" this is sentence is not clear. Reversal from what to what? Convert the direction from surface volatilization to atmospheric deposition? This statement repeatedly occurs in the manuscript that need to be well clarified. In addition, mobilisation from soil, water bodies, or the cryosphere share the same mechanism as air-surface exchange when regarding the emission to the atmosphere. Therefore, this sentence can be refined.

Thank you for your comment. The concept of reversal of the direction of air-surface exchange is explained in the introduction section (including references to the literature). We will change the sentence in the abstract to make it clearer , will read (lines 15-17).

> "…related to reversal of the direction of air-soil exchange in response to historic atmospheric depositions or recent mobilisation from ground compartments, such as water bodies, the cryosphere, or soils heated by wildfires."

(6) *Line 22, rather than semivolatility, long-range transport ability is more precise accounting for the global cycling.*

For semivolatiles (SVOCs) which are only slowly or not degradable in ground compartments, the long-range transport potential (LRTP) is not determined by atmospheric degradation kinetics and size of the carrying aerosol mode as is the case for non-volatiles (NVOCs), or by vapour pressure, water solubility (because of scavenging) and atmospheric degradation kinetics, as is the case for volatiles (VOCs), but from the combination of vapour pressure, atmospheric, aqueous and soil degradation kinetics, and gas-particle partitioning, and may include several subsequent cycles of deposition and volatilisation (Goldberg, 1975; Wania and Mackay, 1993; Scheringer and Wania, 2003; Semeena and Lammel, 2005). The gas-particle partitioning, in turn, is determined by water and organic solubilities (the latter commonly parameterised as function of K_{ow}) and the composition of the aerosol particulate phase (Goss and Schwarzenbach, 2001). Most of the total environmental burden of semivolatiles which are only slowly or not degradable in ground compartments is stored in soils, water bodies or sediment. As persistence of multicompartmental substances results from degradability in all environmental compartments (Mackay, 2001), the statement under discussion here ('Because of persistence and semivolatility, these substances cycle globally.') is correct and concise. For the sake of clarity for a multidisciplinary readership we will extend the statement, including compartmental degradability and references to the literature. The new sentence will read (Lines 24-25):

> "Because of long residence time in ground compartments and air, these substances cycle globally, enhanced by semivolatility, which allows for several cycles of volatilisation and deposition (Wania and Mackay, 1993; Semeena and Lammel, 2005)."

Related additional references are:

Goldberg, E.D.: Synthetic organohalides in the sea. Proc. R. Soc. Lond. B, 189, 277-289, 1975.

Goss K.U., and Schwarzenbach R.P.: Linear free energy relationships used to evaluate equilibrium partitioning of organic compounds. Environ. Sci. Technol. 35, 1–9, 2001.

Mackay, D.: Multimedia environmental models – the fugacity approach, 2nd ed.; CRC Press, Boca Raton, USA, 2001, 272 pp.

Scheringer, M, and Wania, F.: Multimedia models of global transport and fate of persistent organic pollutants, Handbook of Environmental Chemistry, O3 (Fiedler, H., ed.), Springer, Berlin, Germany, 2003, pp. 237-269.

Semeena V.S., Lammel G.: The significance of the grasshopper effect on the atmospheric distribution of persistent organic substances, Geophys. Res. Lett., 32, L07804, 2005.

Wania, F., and Mackay, D.: Global fractionation and cold condensation of low volatility organochlorine compounds in polar regions, Ambio, 22, 10-18, 1993.

(7) *Section 2.4: To prevent potential breakthrough, PUF/XAD2/PUF sandwich was used for sampling gas-phase CUPs, while only two PUF plugs were used for sampling gaseous OCPs.*

Why did the author not apply PUF/XAD2/PUF sandwich to OCP sampling? Can the author assure that there is no breakthrough issue for hi-vol sampling of OCPs without XAD? If not, a limitation on this point should be included in the context.

Thank you for your question. XAD is a resin that has been shown to efficiently sorb gaseous polar chemicals in ambient air (Dobson et al., 2006; López et al., 2018), whereas it is not needed for apolar organics, like OCPs. PUF has a long-term record of reliable use for sampling OCPs. XAD is quite expensive compared to PUF plugs, and more delicate to handle. Regarding breakthrough, a previous study (Degrendele et al., 2016), which was the precursor to this study, had a similar sampling design and ensured breakthrough did not occur for OCPs. The mention of this was added to the manuscript (lines 80-82), now reading:

> "This configuration has been shown to be the most efficient for the collection of gaseous CUPs (Dobson et al., 2006; López et al., 2018) and has been applied and optimized previously for CUPs and OCPs (Degrendele et al., 2016)."

Dobson, R., Scheyer, A., Rizet, A.L., Mirabel, P., Millet, M.: Comparison of the efficiencies of different types of adsorbents at trapping currently used pesticides in the gaseous phase using the technique of high-volume sampling. Anal. Bioanal. Chem., 2006, 386, 1781-1789. https://doi.org/10.1007/s00216-006-0737-2

(8) *Line 91-92, "100 µL of MilliQ water were then added to a 100µL aliquot of the respective extracts which were finally used for analysis." This sentence is not clear. 100µL water was added to both OCP extracts and CUP extracts? Since n-nonane was previously added in the extracts, water and nonane are immiscible and extracts for OCPs analysis using GC-MS/MS should not contain any water. Please explain this.*

Thank you for spotting that. Indeed, the water is only added for the CUPs in LC analysis. For OCPs and GC, its major constituents are a DCM:hexane mix (1:1) and nonane. We have corrected this part of the manuscript, now reading (lines 93-99):

> "CUPs extract clean-up was done by filtration through a 0.22 µm pore size cellulose acetate membrane (Corning Costar Spin-X, United States) and concentrated under a gentle stream of nitrogen to a final volume of 500 µL. 100 µL of MilliQ water were then added to a 100 µL aliquot of the extract which was finally used for analysis. After extraction, OCP extracts were transferred to a glass column (30 mm i.d.) filled with 0.5 g of activated silica, 30 g of H_2SO_4 modified activated silica and 1 g of non-activated silica and were eluted with 40 mL of DCM:hexane (1:1). 50 µL of n-nonane was added, and the extract was then concentrated under a gentle stream of nitrogen to a final volume of 100 µL."

(9) *Line 95, four different methods were used for CUP analysis. Why did not author combine these methods into one or two since they all are based on LC-MS/MS analysis?*

The four methods for CUPs have been described in the cited paper Mayer et al., 2024 (see SI). In order to not overload the column and ensure good time efficiency / cost of analysis. The targeted CUPs were allocated to method based on their chemical family, (e.g., one method for pyrethroids, …)

(10) *Line 108-114, for the recoveries, were they calculated based on spiked native standards or isotopic standards? If based on the former, a series of recovery tests are required. Then, how many replicates were conducted? Otherwise, recoveries can be calculated for each real sample based on their ratio of labelled spike standards (surrogates) to the injection standards in samples relative to those in the calibration standards. Please clarify.*

This information was presented in the manuscript, methods section and SI. The recoveries were calculated based on spiking clean collection material (i.e., filter or PUF/XAD2/PUF sorbent combination) with both native and isotopic standards, (lines 108-114), while the number of replicates is presented in Table S7.

(11) *Line 115, "so was the internal standards for OCPs only." does it mean that internal standards were changed as well" what dose the "only" refer to?*

Thank you for your comment. Internal standards and instrument were changed for analysis related to OCPs only, not for CUPs. Hence what the "only" refers to. Based on a previous comment from a reviewer, the sentence was slightly adjusted for clarity, now reading (lines 124)

> "In 2018, the analytical instrument and internal standards were changed for OCP analysis only."

(12) *Line 147, what are the Non-parametric Mann-Whitney tests used for? Test for difference with previous measurements?*

Thank you for your question. Yes, it was used to compare CUP concentrations observed 2012-2013 (Degrendele et al., 2016) with those from 2019-2021. Results from this analysis are briefly discussed (lines 244-246) and presented in Table S11.

(13) *Line 160, I believe the Clausius-Clapeyron equation works for gaseous phase only. Please include a reference to support your statement of using total concentrations is more appropriate.*

Thank you for this question. While the Clausius-Clapeyron predicts the vapour pressure (in air above contaminated surfaces), among the observables the total concentration, $c_g + c_p$, is the appropriate, due to our sampling design (weekly samples, long-lived substances): As explained in the text, upon entry into the atmosphere, pesticides rapidly equilibrate between the phases according to environmental conditions. Hence, c_g is operationally not conservative. Furthermore, most of OCPs were in the gas phase, so using total concentration yielded similar conclusions and results. Will be rephrased for better clarity to (lines 168-169):

> "c_{tot} is more appropriate than c_g, because inherent to our sampling design (weekly samples, long-lived substances), the total concentration, $c_g + c_p$, is operationally conservative, unlike c_g."

(14) *Line 201-202: Can you explain why the low and decreasing (o,p'-/(o,p'-+p,p'-) ratios ratio indicates that dicofol was not apparently a viable source for presence of DDT in the atmosphere. To make it understandable, additional information on dicofol and its relationship with DDT and DDT should be included in the context.*

Thank you for your comment. Unlike for technical DDT, dicofol contains a higher fraction of *o,p'*-DDT (Qiu et al., 2005). In general a ratio of (*o,p'*-DDX/(*o,p'*-DDX + *p,p'*-DDX) > 0.5 suggest a dicofol source (Qiu et al., 2005; Ricking and Schwarzbauer, 2012). Which is not the case in our observations. These additional information about dicofol were added, the sentence was slightly revised, now reading (lines 220-223)

> "Technical DDT contains a higher fraction of *p,p'*-DDT, while dicofol contains a higher fraction of *o,p'*-DDT (Qiu et al., 2005). For both DDT and DDD, this ratio decreased over time and remained low (0.37 and 0.31 for DDT and DDD, respectively), indicating that dicofol was seemingly not a viable source for presence of DDT in the atmosphere, not during years of declining concentration nor later (Ricking and Schwarzbauer, 2012)."

Additional reference: Qiu, X., Zhu, T., Yao, B., Hu, J., Hu, S.: Contribution of dicofol to the current DDT pollution in China, Environ Sci. Technol., 39, 12, 4385-4390, 2005.

(15) *Line 207-208, please indicate how to use the ratio β-/(α-+γ-) HCH to distinguish between HCH isomers, and include a reference for it.*

Thank you for your comment, based on a previous reviewer's comment and yours, the sentence will be modified (lines 226228):

> "Additionally, the ratio *β*-HCH/(*α*-HCH+*γ*-HCH) which ranged 0.01-0.16 can be used to distinguish between technical HCH (*β*-HCH/(*α*-HCH + *γ*-HCH) ≥ 0.5) and lindane (*β*-HCH/(*α*-HCH + *γ*-HCH) < 0.5) (Li and Macdonald, 2005), as sources of environmental contamination."

(16) *Line 228, how was application of pendimethalin in winter observed? In what cases people apply the pesticide in winter? It's rare to apply pesticides in winter. In fact, it contradicts with the statements in lines 232-233.*

Thank you for your question. This herbicide is applied to prevent unwanted weed to germ, germination season (typically late autumn and early spring). This is visible looking at atmospheric concentration (Figure 1 a and b). We define winter as December to March (Table S12). I think this is where the confusion comes from. Application of this compounds can still happen in early December (Figure 1a; year 2020). To clarify, this sentence and the phrasing in line 232 will be updated, reading (line 253-254 and 258-260):

> "For pendimethalin, as a pre-emergence herbicide, an early winter/late spring application is also seen in 2019 and 2020."
>
> "In general, during winter months, pesticide application are not expected due to low soil temperature (frozen or near), CUPs occurrence in ambient air indicates low degradability"

(17) *Line 240-241: Were the maxima referred to the pre-ban period? If so, the maxima should be higher during the same period in the following years if the legislation works out.*

Thank you for your question. No, the maxima refer to the application period (here the spring and autumn), which were high in 2019 (pre-ban period) and up to six times lower in 2020 and 2021. This shows that legislation was immediately effective.

(18) *Line 245 255: how did author determine whether the slope is steep or not? One approach is to use the slope of Clausius-Clapeyron plot can derive ΔHexp, and then the ΔHexp can be compared with theoretical ΔH, if derive ΔH_{exp} is close to theoretical ΔH, we assume that local volatilization is dominant. If ΔH_{exp} is much small than theoretical ΔH (shallow slope), the long-range transport is dominant (DOI:10.1016/j.ese.2022.100209; DOI: 10.1126/sciadv.adi808).*

Thank you for your question. The approach used was based on previous work from Hoff et al. (1998), Wania et al. (1998), Degrendele et al., (2016) and interpret our data based on their findings. Based on their work, the threshold for slope to be considered steep is -3520. In addition, another criterion was the scatter quantified by R^2. Therefore, conclusive steepness is diagnosed based on both slope < -3520 and $R^2 > 0.6$.

(19) *Line 255-258, how can the "narrow" or "wide" spread be linked to the sources, e.g., soil temperature or secondary sources" Please elaborate.*

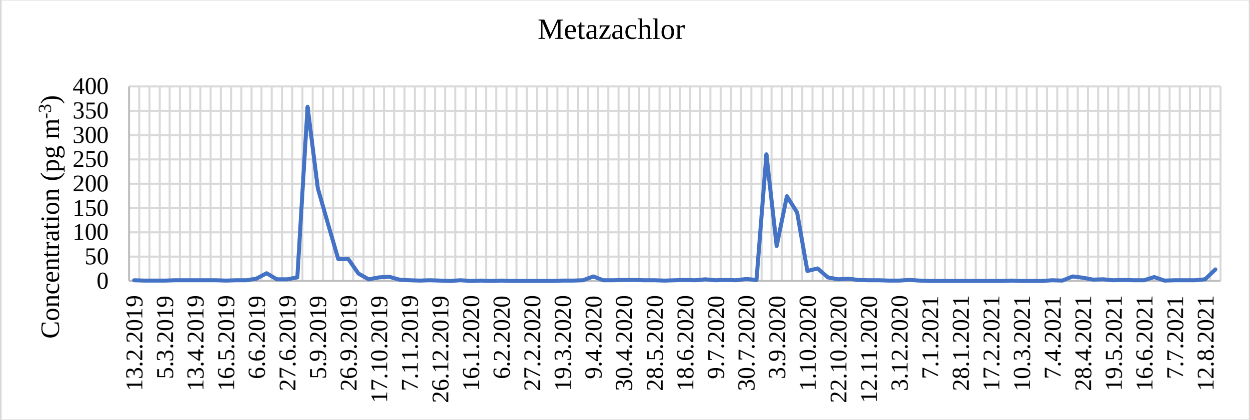
Thank you for your comment. Following Hoff et al. (1998), "…the scatter in the CC plot is due to the varied exposure air parcels encounter before arriving at our sampling site. These air parcels arrive from different directions on days with widely varying temperatures and with varied potential sources." For better clarity will be rephrased, new text will read: (lines 283-286)

> "The results from the Clausius-Clapeyron analysis suggest that soil temperatures play a significant role in DDD levels at this site, as indicated by only little scatter, also in the DDD sinusoidal time series ($R^2 = 0.34$, Table S13b). In contrast, the bigger scatter for DDE ($R^2 = 0.46$, Table S13b), suggests that DDE is more influenced by secondary sources located far from the sampling area (Hoff et al., 1998)."

(20) *Line 266-267, a CUP metazachlor, peaked at summer, how can it be distinguished between application in summer and re-volatilization during the warmest period (summer)?*

Thank you for this discussion. Upon further investigation, it seems that metazachlor application is most likely in August (Wijewardene et al., 2021; https://doi.org/10.1016/j.ecoenv.2021.113036), adding to the revolatilisation-induced summer maximum. This is consistent with our data showing high concentration in air in August, unlike in July when air temperature was high as well (see graph below). Text will be extended accordingly, reading (lines 262-265):

> "…metazachlor peak in summer was particularly pronounced (Table S12, Figure S4), as observed previously (Mai et al., 2013; Degrendele et al., 2016). It was highest in August. This can be explained by coincidence of application, as metazachlor is most used for seed oil plants, usually applied in August for weed control of winter cereals (Wijewardene et al., 2021)."



Added reference:

Wijewardene, L., Wu, N.C., Hörmann, G., Messyasz, B., Riis, T., Hölzel, C., Ulrich, U. and Fohrer, N.: Effects of the herbicides metazachlor and flufenacet on phytoplankton communities – A microcosm assay. Ecotox. Environ. Safety, 228, 113036, 2021.

(21) *Line 300-301: is there a reason why pp-DDD increased during 2013-2017 while decreased during 2018-2022? Has a similar shift of the trend been reported?*

Thank you for your question, From the best of our knowledge, this trend has not been reported before. The reason is unknown. Will be stated reading (line 329)

> "…for unknown reason."

(22) *Line 309-310: why this shift of ratio does not indicate any influence of fresh inputs of the pesticide? Because the ratios are both below 1?*

Thank you for your question, It is actually not a shift, so we should not have used that term. In addition, we have added a reference for the interpretation of these values We have updated the sentence, will read (lines 338-340):

> "For DDT compounds, the ratio of the pesticide over its metabolites, DDT/(DDE+DDD), ranged from 0.27 to 0.34 during the 2013-22 period, which does not indicate any influence of fresh inputs of the pesticide, as generally a ratio <1 implies aged DDT (Yang et al., 2008)."

(23) *Line 314-316, Industrial emissions could be more pronounced compared to the role of being a pesticide for HCB and PeCB. This point can be involved in the discussion in the context rather than being included at the end.*

We had pointed to industrial emissions further down in the text. Thank you for pointing to confusing order of presentation. We will move the sentence on HCB sources up and will slightly extend the discussion of the HCB trend by previous observations at the site and by adding a reference listing more recent unintended sources; will read (lines 345-351)

> "For HCB, a long-term increase was reported in European background air for the years 2016 to 2019 compared to the previous decade (Fiedler et al., 2023; Lunder Halvorsen et al., 2023). Even one decade earlier, higher HCB had been observed at the site in 2004-06 as compared to 1997-99 (Dvorská et al., 2009).
>
> (…)
>
> "HCB are long phased-out from agricultural usage, but are unintentionally released by industries and combustion processes, such as waste incineration (Thomsen et al., 2009; Gong et al., 2017; UNEP 2024)."

Additional references:

Dvorská, A., Lammel, G. and Holoubek, I.: Recent trends of persistent organic pollutants in air in Europe - Air monitoring in combination with air mass trajectory statistics as a tool to study the effectivity of regional chemical policy. Atmos. Environ., 43, 1280-1287, 2009.

Gong, W.W., Fiedler, H., Liu, X.T., Wang, B., Yu, G.: Emission factors of unintentional HCB and PeCBz and their correlation with PCDD/PCDF. Environ. Pollut., 230, 516-522, 2017.

(24) *Line 331-332, could consider additionally include the evidence of net HCH volatilization from water to the atmosphere (https://doi.org/10.1021/acs.est.4c05204; https://doi.org/10.1021/es034998k).*

Thanks for spotting. We will correct it .

(25) *Line 339-342: Despite that gas-particle partition calculation is not available, separate temporal trend analysis for gas- and particle phases is possible. Is there any difference between the two trends as well as compared with the trend for the total concentrations? It would be worthwhile to bring up this point or a short discussion.*

Thank you for your question. Separate time trend analysis of c_g and c_p would not be conclusive, because of the mismatch between sampling periods (weekly samples) and the characteristic time for relaxation to gas-particle partitioning (< 1 h; Meng and Seinfeld, 1996; Wilson et al., 2021).

Meng, Z.Y. and Seinfeld, J.H.: Time scales to achieve atmospheric gas-aerosol equilibrium for volatile species, Atmos. Environ. 30, 2889-2900, 1996.

Wilson, J., Ulrich Pöschl,U., Shiraiwa, M., and Berkemeier, T.: Non-equilibrium interplay between gas–particle partitioning and multiphase chemical reactions of semi-volatile compounds: mechanistic insights and practical implications for atmospheric modelling of polycyclic aromatic hydrocarbons. Atmos. Chem. Phys., 21, 6175-6198, 2021.

(26) Line 349, if the case for δ- and ε-HCH is the same, why was it separately mentioned rather than being included with other OCPs.

Thank you for your question, unlike for the other isomers, there is no previous finding regarding *δ*- and *ε*-HCH temporal trends. However, these compounds also showed negative trends. This is what the sentence means. To clarify the sentence, we will update, then reading (line 391-392):

> "For the OCPs *α*-HCH, *cis*-heptachlor epoxide, *γ*-chlordane, oxychlordane, and *α*-endosulfan significant negative trends are found until 2023, which were consistent with previous findings in the region. Additionally, similar trends were found for *δ*- and *ε*-HCH."

Small things:

Line 15, a comma should be inserted between "mirex" and "levelling off".

Thanks, will be followed.

Line 19 For clarity, consider changing "one-year post-ban" to " one year after the bans"

Thanks, will be followed.

Line 22-24, consider splitting into two sentences for clarity, rephrase like "Due to their severe health and environmental impacts, OCPs have been restricted in most countries (UNEP, 2001). For compounds like DDT and HCH, this has been consistently reflected in declining air concentrations."

Thanks, will be followed.

Line 32, and line 33: inserting a comma between "(CUPs)" and "have", between "neonicotinoids" and "are", could make the sentence clearer.

Thanks, will be followed.

Line 33-34: consider rephrasing the sentence as "are chemically varied and subject to different environmental fates accordingly"

Thanks, will be followed.

Line 36-39, consider splitting sentence into two for clarity. Something like "CUPs can enter the atmosphere during application, where up to 90% of the mass applied can be released directly into the atmosphere (van den Berg et al., 1999). CUPs can be volatilised from surfaces such as soil, plants and surface water over longer periods of time following application (Bedos et al., 2002), and mobilised through wind erosion of soil particles containing CUPs

Thanks, will be followed.

Line 44: " from a few European countries"

Thanks, will be followed.

Line 46, "have become "

Thanks, will be followed.

Line 50-53: consider rephrasing the sentence as: "In this study, biweekly samples of OCPs and CUPs were collected in both the gas and particulate phases at a rural site in an agricultural region of Central Europe, spanning 2013–2022 for OCPs and 2019–2021 for CUPs, allowing for the assessment of seasonal variations and time trends"

Thanks, will be followed.

Line 82: change "on location until transported to the" to "on site until transport to the"

Thanks, will be followed.

Line 110: "processed as per samples" sounds wired. It should be "which were then processed as samples" or in a clearer way like "which were processed in the same way as the samples". And "with a few exceptions, "

Thanks, will be followed.

Line 114: "for QFFs and from 49 % ± 6 (PeCB) to 103 % ± 10 (p,p'-DDD) for PUFs"

Thanks, will be followed.

Line 116, suggest use concise and sample expressions for "from 2018 onward" , e.g., "after 2018" , the word "for" in the "for both OCPs and CUPs" can be removed. The sentence can be rephrased as "the concentration of both OCPs and CUPs have been adjusted by the recoveries."

Thanks, will be followed.

Line 123, for clarity, here is a rephrased version, "For OCPs, with expected one annual amplitude, Equation (1) was used, which .."

Thanks, will be followed.

Line 168, The sentence can be rephrased in a clearer manner. "Eleven CUPs had detection frequencies (DF) greater than 80%, with two CUPs (pendimethalin and tebuconazole) being detected in all samples."

Thanks, will be followed.

Line 174, "these CUPs were all quantified in >65% of air samples"

Thanks, will be followed.

Line 198, "38, 29 and 8 .1% of Σ30OCPs, respectively (Figure 1c,d Table S10)."

Thanks, will be followed.

Line 214, suggest that a revised figure title of " Time series of Σ CUP (a) and Σ OCP (c) concentrations and their relative abundances(b,d) in the atmosphere.

Thanks, will be detailed for better clarity.

Line 222, change "pointing to" to "aligned with"

Thanks, will be followed.

Line 239-240, The sentence can be refined as "High concentrations were due to clearly evident application."

Thanks, will be followed.

Line 242-243, can be improved as "... with spring being quite broad, ranging from ..."

Thanks, will be followed.

Line 255: "….suggest that soil temperatures play a significant role in influencing DDD levels at this site…"

Thanks, will be followed.

Line 274, the sentence can be clarified as "Long-term annual variations in atmospheric concentrations were assessed for 22 CUPs with sufficient concentration data (DF > 20%) using Eq. (2) and Eq. (1) for CUPs and OCPs, respectively."

Thanks, will be followed.

Line 285: "one of them" can be removed.

Thanks, will be followed.

Line 287: The sentence can refined as "Boscalid was the only in-use CUP in the Czech Republic that was decreasing."

Thanks, will be followed.

Line 289-291 , The sentences can be rephrased as " ..was reported to approach zero or in a low amount in 2021. The observed decline was accelerated from 2020 to 2021 compared to the period during 2019-2020, reflecting the combination of these applications and the degradation in the total environment after ban"

Thanks, will be followed.

Line 304-307, the sentence is not clear. Please clarify. Doubled period in the end of the sentence.

Thanks, will be followed.

Line 325, equilibrium rather than equilibria?

No, plural is correct here

Line 328, "which is global" seems not complete. Something like " which impacts globally" or "which is a global concern"?

Thanks, will be followed.

Line 329-330, the sentence is not clear. Please clarify.

This sentence will be removed.

Line 336, " have been reported" rather than "have been reporting"

No, grammar is correct here.

Line 340, the full name of SVOCs should be specified, Semivolatile Organic compounds (SVOCs)

Thanks, will be made consistent throughout.

Line 344, the sentence can be refined as " Overall, this study provided long-term time series data for atmospheric OCPs and CUPs at a Central European site. Consistent with the perception of low degradation rate of many SVOCs in soils, Clausius-Clapeyron analysis results showed that…."

Changed based on comment of previous reviewer.

*Line 356, "investigating" rather than "investigate" ; " for the global OCP cycling"?***;**

No, grammar is correct here

Line 367, The sentence can be rephrased as " In addition, we observed that CUPs' temporal trends are… They were generally negative or insignificant, during which CUPs national use …"

Followed, to be rephrased will read (line 401-402)

> “…we observed that CUPs’ temporal trends were apparently dominated by applications. They were generally negative or insignificant, while during our study period, CUP use in the Czech Republic”

Line 359, “the decreasing trends were directly related to the revocation of their use authorization. “

Thanks, but we feel that rephrasing would not improve clarity.

Line 360, the sentence can be refined as “…in the atmosphere at detectable levels, suggesting a potential atmospheric persistence. “

Thanks for pointing to this, will be corrected, actually suggesting environmental or atmospheric persistence (as long as the compartmental residence times are incompletely known)

Line 362-364, “…highlights… provide sufficient insights into .. and to further develop accurate models to predict key environmental processes such as.. “

Thanks, will be followed.