Reviewer 6:

This paper investigates emitted microplastics aerosols from five sources in northwest China. The authors present a comprehensive characterization of the sources and plasticizer profiles. An eco-health risk assessment was conducted, providing key details like daily exposure risks. These findings are especially valuable for informing risk-mitigation policies and protecting individuals who are often close to sources like those in rural households which burn plastics, agricultural workers or incineration plant workers. This paper merits publication in ACP after some minor revisions.

Dear reviewer,

Thanks very much for taking your time to review this manuscript. We have carefully considered each of your suggestions and have made the adequate revisions to improve the quality of our manuscript.

General comments:

1. The current manuscript misses some clarification and details on the different source categories. For example, it only became apparent in Table 1 that the plastic burning occurred in a household setting not in an industrial one. This distinction is important, as emissions from household and industrial plastic burning may differ significantly, and readers should not conflate the two. Thus, this point should be clarified and emphasized in the Introduction. While it appears that fruit bags were not included in the household plastic burning, it would be helpful to define 'fruit bags' more thoroughly. What polymer types are they made out of - line 105 only states 'also containing some plastics components'. How do they differ from common household plastic waste? Clarifying this will help the readers to better understand why you chose the two categories 'fruit bag' vs 'plastic burning' and their differences.

Response:

Thank you for the reviewer's insightful comments. We have now explicitly stated that our study focused on life pollution sources and the plastic burning occurred in a household setting in the Introduction and Abstract sections. The related statements are as follows.

"Current research on atmospheric MPs sources focuses on industrial emissions and natural processes, but neglects air pollution sources closely related to daily life sources. Given that living sources significantly affect human health, this study pays particular attention to such sources."

"Due to the relatively limited facilities and means of waste disposal, residents in rural areas often resort to open burning when disposing of plastic waste (Pathak et al., 2023). In addition, given the flammability of plastics, residents also tend to use plastics as igniters or even burn them directly when using stoves for cooking or heating, which is an important household source of MPs."

"The aims of this research are to characterize the distributions of MPs and plasticizers in dual-size PMs ($PM_{2.5}$, $PM_{2.5\text{-}10}$) from typical MP sources (anthropogenic sources from daily life) in the Guanzhong Plain."

The reason we distinguish between plastic burning (PB) and fruit bag burning (FB) rather than classifying them as a single combustion source is that the wax layer in fruit bags cannot be separated from the plastic. This is a featured source in the Guanzhong region (This is also quite common in fruit producing areas in northern China), and local residents typically burn fruit bags directly without separating the wax. We have revised this point in the manuscript as follows.

"The Guanzhong Plain is an important fruit production base in China, with the highest

consumption of fruit bags. Local residents often use the above-mentioned plastic products to ignite solid fuels for indoor heating or cooking. The reason we distinguish between Plastic Burning and Fruit-bag Burning rather than classifying them as a single combustion source is that the wax layer in fruit bags cannot be separated from the plastic. This is a featured source in the Guanzhong region (This is also quite common in fruit producing areas in northern China), and local residents typically burn fruit bags directly without separating the wax. Table 1 provides a summary of the essential details for each source."

The more details of fruit bags and their differences with household plastic waste have been added in the manuscript as follows.

"Fruit bags are typically lightweight, thin-film, which are designed for single-use and are often discarded after a short period, differing from common household plastic waste. These bags are usually made from low-density polyethylene and Nylon, which is known for its flexibility and transparency (Ali et al., 2021; Yang et al., 2022)."

Reference:

Ali, M. M., Anwar, R., Yousef, A. F., Li, B. Q., Luvisi, A., De Bellis, L., Aprile, A., and Chen, F. X.: Influence of Bagging on the Development and Quality of Fruits, Plants-Basel, 10, 16, 10.3390/plants10020358, 2021.

Yang, H. G., Gu, F. W., Wu, F., Wang, B. K., Shi, L. L., and Hu, Z. C.: Production, Use and Recycling of Fruit Cultivating Bags in China, Sustainability, 14, 18, 10.3390/su142114144, 2022.

Pathak, G., Nichter, M., Hardon, A., Moyer, E., Latkar, A., Simbaya, J., Pakasi, D., Taqueban, E., and Love, J.: Plastic pollution and the open burning of plastic wastes, Glob. Environ. Change-Human Policy Dimens., 80, 9, 10.1016/j.gloenvcha.2023.102648, 2023.

2. Details on the collection of field blanks are limited and would benefit from further clarification. Line 118: How were the field blanks collected? By synchronously, do the authors mean that the blank was taken at the same location, same duration, and same time as the samples?

Response:

We have provided the information on how the field blanks collected in Line 163-166.

"The field blank of each type of source was synchronously collected with active sampling. Unused filters (the same batch as sampling filters) were loaded into identical sampling devices, which were placed adjacent to operational samplers for the entire duration of one sampling event."

3. Line 128: Why were phthalates, benzothiazole and its derivatives, and bisphenol A chosen to be the 3 classes for in-depth analysis? What criteria guided this choice?

Response:

The selection of BTs, PAEs, and BPA as the focus of our study is driven by their widespread use, ubiquitous in the environment and potential health risks.

Phthalate esters (PAEs) are the most widely used plasticizers globally, dominating the plastic additive market. He et al. (2020) demonstrated that during 2007-2017, the annual global production of PAEs increased from 2.7 million tons to 6 million tons. Moreover, China is recognized as the largest importer of PAEs worldwide. Benzothiazoles (BTs) are extensively used in automotive tires and agrochemicals. High concentrations of BTs were discovered in the street runoff, suggesting that these tire material-related compounds can persevere in the environment (Zhang et al., 2018). Exposure to BTs may result in central nervous system depression, liver and kidney damage, dermatitis, and pulmonary irritation (Ginsberg et al., 2011). Bisphenol A (BPA) as a common industrial chemical component in many products, has steadily grown over the last 50 years (Corrales et al., 2015). Growth of global production has consistently ranged between 0% and 5% annually (Corrales et al., 2015). PAEs and BPA considered as endocrine disruptors, are demonstrated to impair reproductive function and development in laboratory animals (Wang et al., 2018).

Despite the well-documented health risks associated with these plasticizers in laboratory settings, there is a significant gap in understanding their real-world emissions and health impacts. In this study, we aim to fill this gap by investigating the emission characteristics of these plasticizers from various sources and evaluating their potential health impacts based on real-world concentration levels.

The related sentences have been revised as follows.

"Plasticizers are widely used in the production of plastics in order to achieve the desired material properties (Demir and Ulutan, 2013). Since plasticizers are not chemically bound to the plastic products, they can easily diffuse into the surrounding environment during the life-time (Yadav et al., 2017; Demir and Ulutan, 2013). PAEs, BTs, and BPA are the most common plastic additives that are ubiquitous in the environment and pose potential health risks. Phthalate esters (PAEs) are the most widely used plasticizers globally, dominating the plastic additive market. He et al. (2020) demonstrated that during 2007-2017, the annual global production of PAEs increased from 2.7 million tons to 6 million tons. China is recognized as the largest importer of PAEs worldwide. Benzothiazoles (BTs) are extensively used in automotive tires and agrochemicals. High concentrations of BTs were discovered in the street runoff, suggesting that these tire material-related compounds can persevere in the environment (Zhang et al., 2018). Exposure to BTs may result in central nervous system depression, liver and kidney damage, dermatitis, and pulmonary irritation (Ginsberg et al., 2011). Bisphenol A (BPA) as a common industrial chemical component in many products, has steadily grown over the last 50 years (Corrales et al., 2015). Growth of global production has consistently ranged between 0% and 5% annually (Corrales et al., 2015). PAEs and BPA considered as endocrine disruptors, are demonstrated to impair reproductive function and development in laboratory animals (Wang et al., 2019).

Previous studies have investigated the emission characteristics of plasticizers from various sources. Simoneit et al. (2005) illustrated that the major plasticizers detected in particulate matters (PMs) from open-burning of plastics were dibutyl phthalate (DBP), diethylhexyl adipate (DEHA), and diethylhexyl phthalate (DEHP). Zeng et al. (2020) reported phthalate concentrations in greenhouses air were higher than that in ambient air. Liu et al. (2023) found that phthalates were the most dominant plasticizer compositions in tunnel PM2.5, accounting for 64.8% of the detected plasticizers. Zhang et al. (2018) demonstrated that tire material-related compounds, benzothiazole (BT) and 2-hydroxybenzothiazole (2-OH-BT) were the major compounds in both tire and road dust samples. The majority of existing studies on atmospheric MPs and plasticizers have focused on analyzing the emission characteristics of individual source and lacked a comprehensive and comparative analysis of the MPs emission profiles of various sources."

Reference:

Corrales, J., Kristofco, L. A., Steele, W. B., Yates, B. S., Breed, C. S., Williams, E. S., and Brooks, B. W.: Global Assessment of Bisphenol A in the Environment: Review and Analysis of Its Occurrence and Bioaccumulation, Dose-Response, 13, 29, 10.1177/1559325815598308, 2015.

Demir, A. P. T. and Ulutan, S.: Migration of phthalate and non-phthalate plasticizers out of plasticized PVC films into air, Journal of Applied Polymer Science, 128, 1948-1961, 10.1002/app.38291, 2013.

Ginsberg, G., Toal, B., and Kurland, T.: Benzothiazole Toxicity Assessment In Support Of Synthetic Turf Field Human Health Risk Assessment, J. Toxicol. Env. Health Part A, 74, 1175-1183, 10.1080/15287394.2011.586943, 2011.

He, M. J., Lu, J. F., Wang, J., Wei, S. Q., and Hageman, K. J.: Phthalate esters in biota, air and water in an agricultural area of western China, with emphasis on bioaccumulation and human exposure, Sci. Total Environ., 698, 9, 10.1016/j.scitotenv.2019.134264, 2020.

Liu, M. X., Xu, H. M., Feng, R., Gu, Y. X., Bai, Y. L., Zhang, N. N., Wang, Q. Y., Ho, S. S. H., Qu, L. L., Shen, Z. X., and Cao, J. J.: Chemical composition and potential health risks of tire and road wear microplastics from light-duty vehicles in an urban tunnel in China, Environmental Pollution, 330, 9, 10.1016/j.envpol.2023.121835, 2023.

Simoneit, B. R. T., Medeiros, P. M., and Didyk, B. M.: Combustion products of plastics as indicators for refuse burning in the atmosphere, Environ. Sci. Technol., 39, 6961-6970, 10.1021/es050767x, 2005.

Wang, Y., Zhu, H. K., and Kannan, K.: A Review of Biomonitoring of Phthalate Exposures, Toxics, 7, 28, 10.3390/toxics7020021, 2019.

Yadav, I. C., Devi, N. L., Zhong, G., Li, J., Zhang, G., and Covaci, A.: Occurrence and fate of organophosphate ester flame retardants and plasticizers in indoor air and dust of Nepal: Implication for human exposure, Environmental Pollution, 229, 668-678, 10.1016/j.envpol.2017.06.089, 2017.

Zeng, L.-J., Huang, Y.-H., Chen, X.-T., Chen, X.-H., Mo, C.-H., Feng, Y.-X., Lu, H., Xiang, L., Li, Y.-W., Li, H., Cai, Q.-Y., and Wong, M.-H.: Prevalent phthalates in air-soil-vegetable systems of plastic greenhouses in a subtropical city and health risk assessments, Sci. Total Environ., 743, 10.1016/j.scitotenv.2020.140755, 2020.

Zhang, J., Zhang, X., Wu, L., Wang, T., Zhao, J., Zhang, Y., Men, Z., and Mao, H.: Occurrence of benzothiazole and its derivates in tire wear, road dust, and roadside soil, Chemosphere, 201, 310-317, 10.1016/j.chemosphere.2018.03.007, 2018.

Zhang, H., Yang, R. F., Shi, W. Y., Zhou, X., and Sun, S. J.: The association between bisphenol A exposure and oxidative damage in rats/mice: A systematic review and meta-analysis, Environmental Pollution, 292, 9, 10.1016/j.envpol.2021.118444, 2022.

4. Near the source you may have metals and other aerosols depositing onto the filter, were these non-plastic sources accounted for in your analysis? How was the mass contribution from microplastics determined?

Response:

In this study, we employed Mass Spectrometry methods (Py-GCMS) for the identification and quantification of microplastics and plasticizers. These methods detect marker ions unique to microplastics (MPs) and plasticizers, while metals and other aerosols do not produce these fragments (Hermabessiere et al., 2018).

The mass contribution from microplastics was determined by calculating the mass proportion of each type of MP to the total nine detected MPs in the samples.

Reference：

Hermabessiere, L., Himber, C., Boricaud, B., Kazour, M., Amara, R., Cassone, A. L., Laurentie, M., Paul-Pont, I., Soudant, P., Dehaut, A., and Duflos, G.: Optimization, performance, and application of a pyrolysis-GC/MS method for the identification of microplastics, Anal. Bioanal. Chem., 410, 6663-6676, 10.1007/s00216-018-1279-0, 2018.

5. The manuscript uses too many abbreviations, some of which are uncommon or used only once, making the text hard to follow. I recommend writing out PB, FB, RT, AF, and LB, and remove single-use abbreviations like FL (line 166). Replace OP with "oxidation potential" throughout. Limiting abbreviations to widely recognized terms like the different microplastics and plasticizers will improve clarity and readability.

Response:

Thank you for your suggestion. We have revised the manuscript to remove abbreviations of PB, FB, RT, AF, LB, and OP in the main text to enhance readability. However, in order to maintain conciseness and adhere to the word limit, we have retained the abbreviations in the Abstract and Figures/Tables.

Specific comments:

- Line 89: Can you elaborate why is the UV so strong in this area and why does it matter for this study? MPs emitted in this area may undergo increased photooxidation, is that something that can be discussed with your results? Or is that something that needs to be explored further in future studies?

Response:

The Guanzhong Plain, our study area is located in northwestern China, and according to the study "Ultraviolet radiation over China: Spatial distribution and trends (Sust. Energ. Rev., 76, 1371-1383)", this region experiences relatively strong UV radiation (14486 kJ m^{-2} day^{-1}, the average solar radiation of Xi’an, Tongchuan, and Xianyang). The high UV radiation in this area can be attributed to the elevated altitude (497 m, the average elevated altitude of Xi’an, Tongchuan, and Xianyang) (reducing atmospheric path length) and less cloud cover (especially low cloud cover). Ultraviolet radiation drives photooxidation of plastics, breaking polymer chains and accelerating fragmentation into secondary MPs. The high UV intensity in the Guanzhong Plain exacerbates this process.

However, our current research primarily focused on the characteristics and health risks of MPs and plasticizers emitted from various sources in the region. We did not specifically examine how UV-induced aging of MPs might influence their environmental behavior or toxicity. This aspect is important as ultraviolet radiation can alter the physical and chemical properties of MPs, potentially increasing their toxicity and capacity to adsorb other pollutants. We accept the reviewer's suggestion and plan to explore the effects of UV on MPs and their associated health risks in future studies. This will provide a more comprehensive understanding of the eco-health impacts of MPs in the Guanzhong Plain.

Reference:

Liu, H., Hu, B., Zhang, L., Zhao, X. J., Shang, K. Z., Wang, Y. S., and Wang, J.: Ultraviolet radiation over China: Spatial distribution and trends, Renew. Sust. Energ. Rev., 76, 1371-1383, 10.1016/j.rser.2017.03.102, 2017.

■ Line 113: State what type of sampler MiniVol samplers are (e.g. impinger, impactor, etc.)

Response:

Thank you for this comment. The MiniVol air samplers used in this study are impactors. This point has been revised as follows.

"MiniVOL samplers (Airmetrics, Springfield, OR, USA), which are inertial impactors, were employed for collection, operating at a steady flow rate of 5 L min^{-1}."

■ Figure 1: errors bars used represent standard deviation of PM10 however the figure is plotting PM5 and PMcourse. Is that error representation suitable for this plot? Consider just focusing on PM2.5 and PMcourse throughout the paper and removing PM10 to be more consistent with how results are discussed.Is that error representation suitable for this plot? Consider just focusing on PM2.5 and PMcourse throughout the paper and removing PM10 to be more consistent with how results are discussed.

Response:

We have revised Figure 1 to illustrate the characteristics of microplastics and plasticizers in $PM_{2.5}$ and $PM_{2.5\text{-}10}$ (PM_{coarse}) and remove PM_{10} from this figure. Additionally, we have included error bars representing the standard deviation for both $PM_{2.5}$ and $PM_{2.5\text{-}10}$.

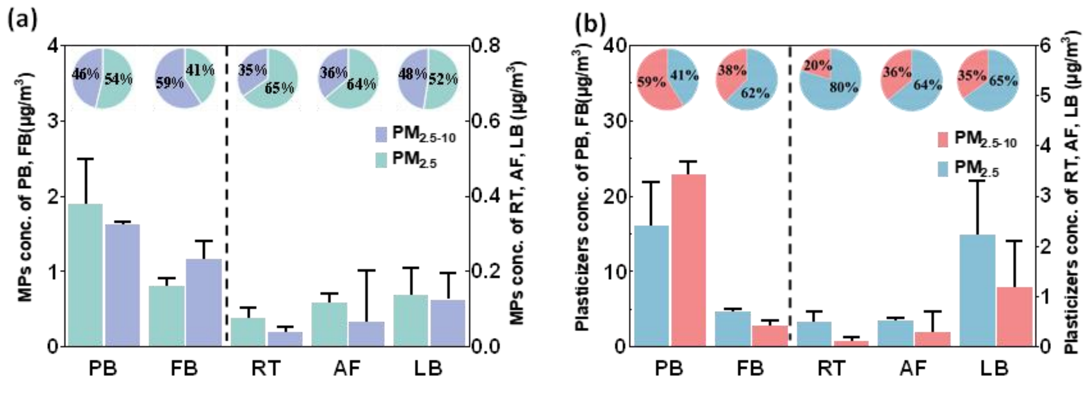

Figure 1 Average concentrations of MPs (a) and plasticizers (b) in $PM_{2.5}$ and $PM_{2.5\text{-}10}$ from five sources (PB: Plastic Burning, FB: Fruit-bag Burning, RT: Road Traffic, AF: Agricultural Film, LB: Livestock Breeding).

■ Figure 1: I would suggest increasing the spacing between (a) and (b) to improve readability. The secondary y-axis for (a) is very close the first y-axis of (b).

Response:

Suggestion taken. The figure was shown as above.

- Line 282: In addition to the most abundant plasticizer, I would suggest the authors list the next two most abundant plasticizer types detected in PB.

We apologize for causing confusion to the reviewer. It has been revised as follows.

"The highest concentrations of PAEs, BTs, and BPA still appear in PB among five sources."

- Line 288-289 and Figure 5: Use consistent spelling of tire or tyre throughout the paper.

Response:

Sorry for the error. We have revised line 288-289 and Figure 5 to consistently use the spelling "tire" throughout the revised manuscript.

- Figure 3: Redefine abbreviations when they appear in captions e.g., PAE and BT

Response:

Suggestion taken.

- Figure 4: Consider adding visual cues (e.g. labels by the arrow, highlighted section or color cues) to help guide the reader's eye from the black arrow to the corresponding source marker on the x-axis. In the main text, please expand the discussion for these identified markers. How confident are these marker classifications? Have previous studies reported the same markers?

Response:

We have added the abbreviation of the fingerprint species next to the arrow in Figure 4 to highlight the substance. The revised Figure is as follows.

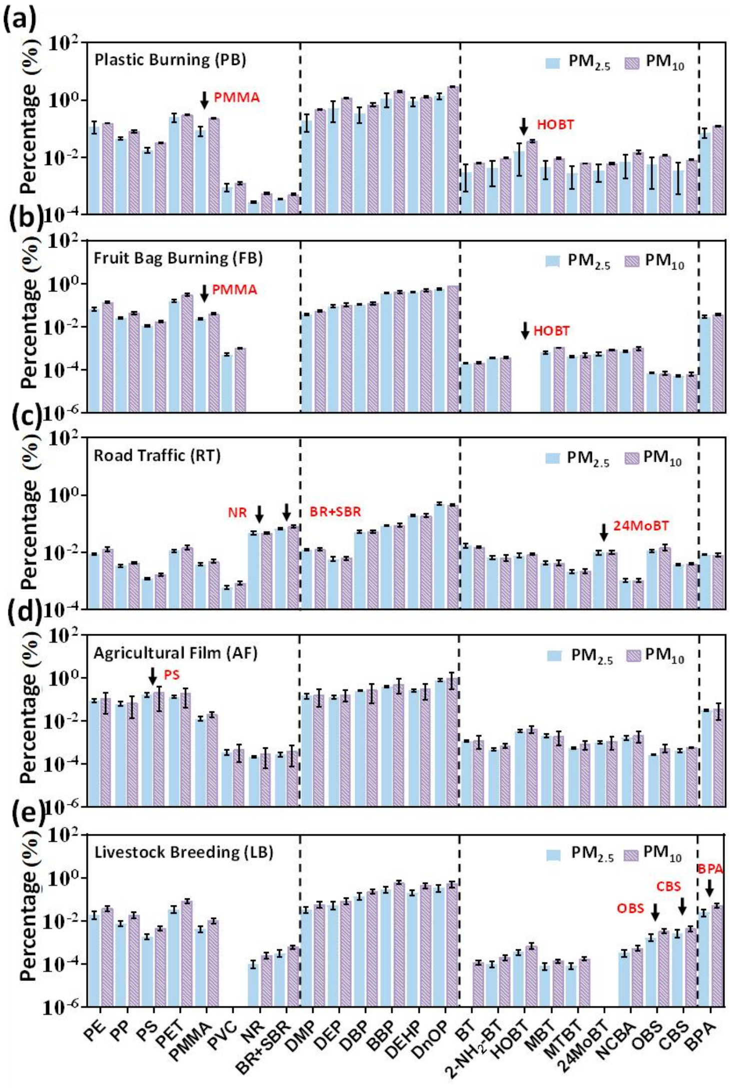

Figure 4 Source profiles of microplastics and plasticizers in $PM_{2.5}$ and PM_{10} (The black arrows indicate the source markers).

The marker classifications in this study were based on the distinct emission characteristics of different sources. We compared and selected these markers to represent each source accurately based on our real-world experiment results. While previous studies on the emission characteristics of microplastics from these sources are relatively limited, there are some findings that corroborate our results. We have expanded the discussion for the identified markers in the manuscript as follows.

"Liu et al. (2019) documented that polystyrene (PS) were the predominant polymer in agricultural sources."

"Liu et al. (2023) revealed that natural rubber (NR) and other rubber particles are emitted at high levels in tunnel traffic, emerging as the dominant microplastic in traffic-dominated environments."

Reference:

Liu, K., Wang, X., Fang, T., Xu, P., Zhu, L., and Li, D.: Source and potential risk assessment of suspended atmospheric microplastics in Shanghai, Sci. Total Environ., 675, 462-471, 10.1016/j.scitotenv.2019.04.110, 2019.

Liu, M. X., Xu, H. M., Feng, R., Gu, Y. X., Bai, Y. L., Zhang, N. N., Wang, Q. Y., Ho, S. S. H., Qu, L. L., Shen, Z. X., and Cao, J. J.: Chemical composition and potential health risks of tire and road wear microplastics from light-duty vehicles in an urban tunnel in China, Environmental Pollution, 330, 9, 10.1016/j.envpol.2023.121835, 2023.

■ Figure 6: Please add a legend to define the colour groupings.

Response:

Suggestion taken.

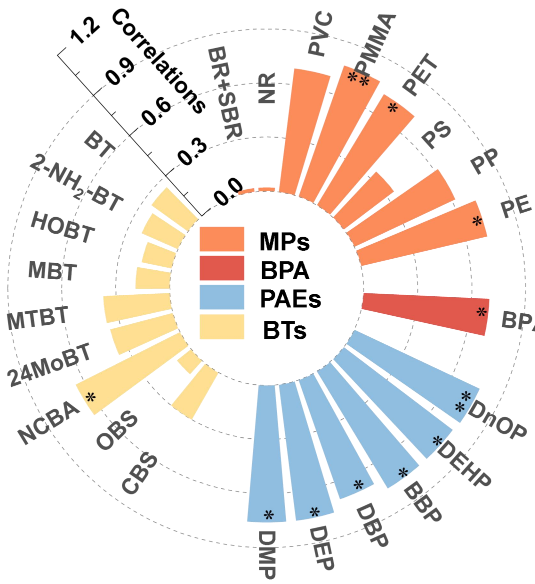

Figure 6 Correlations between DTT, MPs, and plasticizers (*P < 0.05; **P < 0.01).

Technical corrections:

■ Line 19: (phthalates, benzothiazole and its derivatives, and bisphenol A)

Response:

Suggestion taken. It has been revised as follows.

"the characteristics and source profiles of eight types of common MPs and three classes of plasticizers (i.e., phthalates, benzothiazole and its derivatives, and bisphenol A)."

■ Line 31: missing closing bracket: poly(methyl methacrylate)

Response:

Corrected.

■ Line 122: missing space: …frozen at -20°C

Response:

Corrected.

■ Line 180: and absorbance was measured at 412 nm using a microplate reader

Response:

Corrected.

- Line 246: This is because plastic products...

Response:

Corrected.