Response to Reviewers

"Characteristics of Legacy and Emerging Per- and Polyfluoroalkyl Substances in Atmospheric Total Suspended Particulate from The Coastal Areas in China"

Dear reviewers:

We would like to thank you for careful and thorough reading of this manuscript. Thanks for your professional and valuable comments, which are significantly helpful to improve the manuscript. According to these comments, we try our best to revise the manuscript carefully and thoroughly. All revisions were highlighted with red font in Revised Manuscript with marked changes. The following pages contain the detailed responses to these comments.

Sincerely,

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Reviewer 1#

General comment:

The authors collected particulate matter from Laoshan (along the coast of the East China Sea) and from the Xisha Islands (in the South China Sea) and tested for 30 PFAS. They quantified 19 PFAS at Laoshan and 14 at Xisha. As in other studies, long-chain PFCAs were most prevalent, so this result is not particularly surprising. Among the emerging PFAS tested, HFPO-DA, 6:2 Cl-PFESA, and PFOSA were detected at Laoshan only; DONA at Xisha Islands only; and 6:2 FTSA at both sites. It is intriguing to see the spike of DONA on March 15-17, though not explored much in the manuscript.

The scientific approach is generally appropriate. The authors describe QA/QC measures, but they should include more details about blanks to demonstrate full scientific rigor, as described in more detail below. I also caution against over-interpreting the sectors of PFAS sources identified from PCA-MLR. With the current selection of figures in the manuscript, it is not easy for the reader to make direct comparison between measured PFAS concentrations at the two sampling sites. See below for suggested changes.

Response: Thank you for the constructive suggestions. We have provided more details about the blanks in the SI (as Comment 4) and toned down the interpretation of the PCA-MLR results throughout the manuscript, using more conditional language (e.g., "may", "suggest") as suggested in Comments 9 and 10. In response to Comment 5, we have integrated the original Figures S1 and Figure S2 into the main manuscript as Fig 3 (the previous Fig 3 has been renumbered as Figure S1 in the updated Supplementary Materials) to facilitate a clearer visual comparison between the two sampling sites.

Comment 1: Lines 16-18 of Abstract: What do the authors mean by "the similarity of PFAS distribution characteristics"? The distribution characteristics of Figure S1 vs S2 do not look similar to me, nor do the pie charts in Fig 5.

Response: Thank you for your comment. The description of "the similarity of PFAS distribution characteristics" was indeed ambiguous. It has been revised as "It suggested that the predominance of long-chained PFCAs (e.g., PFOA) at both Laoshan and Xisha Islands may due to a same long-distance atmospheric transport route." In addition, Line 359-361 "Combined with the similarity of PFAS distribution characteristics between the two regions, it revealed that long-distance atmospheric PFAS transport builds Bridges between these geographically different coastal systems." was revised as "It revealed that long-distance atmospheric PFAS transport could explain the predominance of PFOA at the two sites."

Comment 2: Lines 41-42: I question the authors' statement that few studies have focused on atmospheric PFAS in China. They cite at least 8 works in the manuscript published between 2015-2019. There are also multiple papers published more recently. As most relevant, the authors should recognize the coastal and marine measurements from southeastern China by Yamazaki et al. (DOI 10.1016/j.chemosphere.2021.129869).

Response: Thank you for your constructive comment. Indeed, it is an inaccurate description in the original statement, which may have caused confusion for readers. Therefore, "To date, few studies have focused on atmospheric PFASs in China." has been revised as " To date, the legacy and emerging PFASs have been detected in atmospheric aerosol particles in worldwide (Faust et al., 2021; Yamazaki et al., 2021). However, few studies focused on the marine atmosphere aerosol particles of PFASs，especially on potential long-range transport between different coastal regions." We also thank you for recommending the highly relevant study by Yamazaki et al. (2021). It strongly supported the work for coastal PFAS measurements in China and has been added in the introduction (Line 50).

Comment 3: Line 102: Some of the target analytes lacked a corresponding isotopically labelled standard. Were the concentrations of 6:2 Cl-PFESA, HFPO-DA, and DONA corrected by the percent recoveries in Table S4? If yes, please specify. If no, I recommend to acknowledge that detections and absolute quantitations of 6:2 Cl-PFESA, HFPO-DA, and DONA are likely to be underestimates because of analyte loss during sample prep, e.g., from sorption to the nylon filter.

Response: Thank you for your constructive comment. 6:2 Cl-PFESA, HFPO-DA, and DONA were lack of isotopically labelled standards, and concentrations of them were not corrected by the percent recoveries. "It should be noted that 6:2 Cl-PFESA, 8:2 Cl-PFESA, HFPO-DA, and DONA were lack of isotopically labelled standards, and concentrations of them were not corrected by the percent recoveries. Thus, they would be underestimates in the present study due to the loss during sample pretreatment, e.g., from sorption to the nylon filter." has been added in the manuscript of Quality assurance and quality control (QA/QC).

Comment 4: Lines 106-108: It seems misleading to say that PFAS levels in all blanks were either not detected or below MDLs because the MDLs were defined based on blank concentrations. Could the authors please clarify? I recommend to add blanks to the data tables in the SI for full transparency. For example, it seems counterintuitive for the MDL for 6:2 FTS to be so low when its percent recovery is >> 100, suggesting background contamination.

Response: Thank you for your valuable comments. "The PFAAs were not detected in all the blank samples or were below their corresponding MDLs in the procedural blanks, filed blanks, and methanol." was misleading, and it has been revised as your suggestion: "PFNA, PFOS, PFHxS, and 6:2 FTS et. al were detected in blanks and the values were list in Table S4." It has been revised as follows:

Table S4 The method detection limits (MDLs) and recoveries of target compounds

Target compounds	Internal Standard	MDLs (pg·m^{-3})	Recovery (%, Avg±SD)	Blank(pg·m^{-3})
PFBA	MPFBA	0.009	93.1±0.9	n.d.
PFPeA	MPFPeA	0.023	94.2±0.9	n.d.
PFHxA	MPFHxA	0.060	93.0±0.4	n.d.
PFHpA	MPFHpA	0.044	94.7±0.3	n.d.
PFOA	MPFOA	0.046	92.5±0.6	n.d.
PFNA	MPFNA	0.144	90.5±0.2	0.103
PFDA	MPFDA	0.062	90.9±0.4	n.d.
PFUnDA	MPFUnDA	0.069	92.9±0.5	n.d.
PFDoDA	MPFDoDA	0.071	93.2±0.2	n.d.
PFTrDA	MPFDoDA	0.061	118.4±0.7	0.034
PFTeDA	MPFTeDA	0.313	94.0±0.4	0.229
PFBS	MPFBS	0.093	94.9±0.4	n.d.
PFPeS	MPFBS	0.115	91.7±0.6	0.085
PFHxS	MPFHxS	0.197	93.1±0.5	0.152
PFHpS	MPFHxS	0.092	96.6±0.4	n.d
PFOS	MPFOS	0.381	93.6±0.7	0.293
PFNS	MPFOS	0.222	101±5.6	0.171
PFDS	MPFOS	0.350	90.2±0.2	0.269
N-MeFOSAA	d3-N-MeFOSAA	0.841	95.1±0.5	0.647
N-EtFOSAA	d3-N-EtFOSAA	0.449	94.2±1.7	0.345
4:2 FTS	M4:2 FTS	0.515	128.4±0.3	0.396
6:2 FTS	M6:2 FTS	0.012	124.3±0.5	0.009
8:2 FTS	M8:2 FTS	0.108	126.8±0.4	0.073
FBSA	MPFBS	0.089	93.6±0.3	n.d
FHxSA	MPFOS	0.056	98.3±1.5	n.d
PFOSA	MPFOSA	0.069	90.8±0.1	n.d
6:2 Cl-PFESA	/	0.018	72.1±1.5	n.d
8:2 Cl-PFESA	/	0.102	69.3±1.5	0.078
HFPO-DA	/	0.128	73.0±1.7	0.098
ADONA	/	0.052	88.4±3.0	n.d

Comment 5: Figure 2 and 3: If the goal is to compare PFAS profiles at Laoshan and Xisha Islands, then I suggest that the authors make Figure 2 & 3 two-panel figures with one panel for each site. Otherwise, it's hard to make a visual comparison of

concentrations when the Laoshan data in Fig 2 are displayed in a different format from the Xisha Islands data in Fig 3.

Response: Thank you for your excellent suggestion. To facilitate a direct visual comparison of the spatial distribution patterns of PFASs between Laoshan and Xisha Islands, the profiles of PFASs at Laoshan and Xisha Islands were displayed by Fig. 3, and it has been revised as follows:

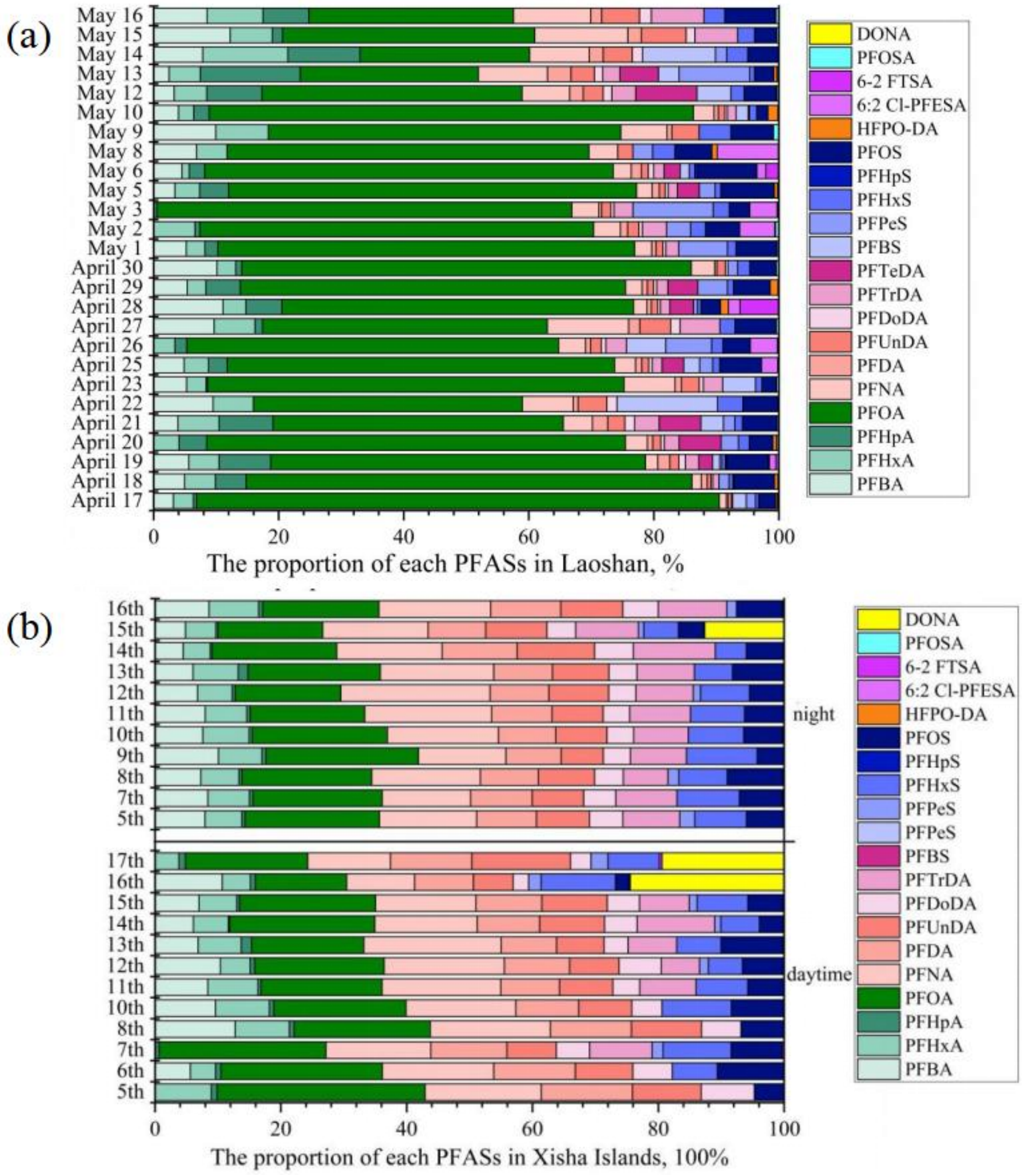


Fig. 3. Proportion of PFASs in TSP at Laoshan and Xisha Islands, China

Comment 6: Figure 3: Is the daily average concentration of PFAS integrated over a full 24-hour period?

Response: Thank you for raising this important question regarding concentration calculations. The concentrations reported in this study are volumetric concentrations, calculated as the mass of PFAS collected on the filter divided by the total volume of air sampled during the corresponding period, with the unit of pg/m^3. Therefore, the daily average concentrations shown in Figure 3 (in original manuscript) represent the average concentrations over their respective sampling periods, rather than 24-hour integrated averages. We acknowledge that this approach would induce some uncertainties, such as temperature, wind speed, light duration, et al.

Comment 7: Sample Collection for Xisha Islands: In lines 197-205, the authors hypothesize that day vs night differences occur because the ship was sailing in the day and stationary at night. I find the discussion somewhat confusing.

(a) What can the authors learn from the exceptions, i.e., night samples when the ship was in motion (XS-02 and XS-22)?

(b) I have a related clarifying question... Based on my interpretation of Table S2, the day and night samples associated with 20210316 are XS-21 and XS-22, and the ship was sailing for both and stationary for neither.

(c) It could be helpful if the authors number the sampling sites in Figure 1 to connect them to the samples in Table S2.

Response: We thank the reviewer for pointing out the lack of clarity in this section.

(a) We originally intended to use Original Figure 3 (Current Figure S1) to illustrate the potential impacts of environmental factors on PFAS concentrations during sampling, particularly the concentration variations that may be caused by environmental conditions at different sampling locations or time periods. However, we recognize the limitations of discussion on navigation status of PFAS concentrations due to the limited sampling period. Therefore, we have revised content is as follows:" As shown in Figure 3, long-chained PFCAs were the main PFASs, with the proportion of 72.0%. Similar compositional characteristics have also been observed in the East China Sea (Sun et al., 2025), Taiwan Strait (Yamazaki et al., 2021). Long-chained PFCAs, such as PFOA and PFUnDA, were also identified as major pollutants in rivers and adjacent coastal water of Hainan Province, China. (Tang et al., 2025; Hu et al., 2025). Generally speaking, the similar profiles of PFASs in the south coastal area may due to the sources of wastewater treatment plants and industrial emission. The concentrations of PFASs in TSP of Xisha Islands were lower than Bohai Sea and Yellow Sea (Yu et al., 2018b), the East China Sea (Sun et al., 2025), and the Pearl River Delta (Liu et al., 2023). Notably, ADONA showed relatively low detection frequency (Fig. S1, Table S6), which may be due to its oxidative degradation to PFCAs in the environment (Zhang et al., 2019).

(b) Our initial statement regarding March 16 was misleading. XS-21 was collected at 7:00-7:52 and 16:31-18:30, respectively, when the ship was stationary. XS-22 was collected at 19:00-6:30 (next day) when the ship was moving. Table S2 has been revised to make the sampling information clearer.

(c) Sample numbers corresponding to those listed in Table S2 have been added to Figure 1 to make sampling site and time visible.

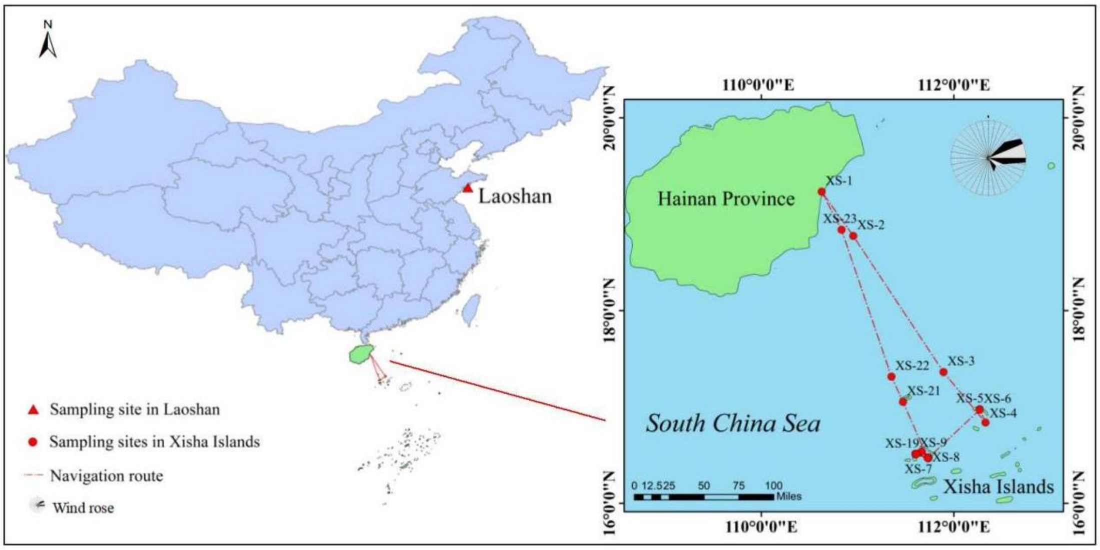


Fig. 1. The sampling sites at Laoshan and Xisha Islands, China

Comment 8: Lines 225-239: I caution that the authors are unlikely to find significant and strong correlations for the emerging PFAS given that the data set is heavily censored (lots of n.d.'s and <MDL's).

Response: Thank you for this suggestion. The strong correlations were mainly found among legacy PFASs (e.g., PFCAs and PFSAs), which were detected in most samples. The MDL was substituted by dividing the $\sqrt{2}$ if the detected value is lower than MDL.

In addition, PFASs with a detection frequency lower than 20% were excluded from the correlation analysis according to Jian Zhou et al. (DOI: 10.1016/j.envint.2021.107007). We have incorporated the following statement in the manuscript (lines 247–249): "It should be noted that PFASs with a detection rate lower than 20% were excluded from the correlation data analysis according to Jian Zhou et al. (2021)."

Tables S8 have been revised as follows:

Table S8 Pearson rank correlations between the PFASs components in Laoshan TSP samples

Laoshan(n=26)	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFPeS	PFHxS	PFOS	HFPO-DA	6:2 Cl-PFESA	6:2 FTSA	PFOSA
PFBA	.715**	.658**	.703**	.435*	.596**	.529**	.657**	.496**	.556**	.126	-.010	.374	.657**	.588**	.174	.646**	.152
PFHxA		.800**	.540**	.471*	.716**	.647**	.770**	.419*	.387	.359	.006	.417*	.591**	.329	.016	.166	-.048
PFHpA			.665**	.272	.866**	.390*	.730**	.547**	.709**	.023	.276	.084	.686**	.475*	.139	.283	.137
PFOA				.134	.648**	.160	.565**	.513**	.600**	-.015	.266	.200	.832**	.668**	.265	.331	.293
PFNA					.467*	.916**	.639**	.554**	.171	.397*	.033	.644**	.282	.117	-.142	.022	.205
PFDA						.560**	.901**	.670**	.574**	.127	.078	.172	.806**	.201	.181	.205	.185
PFUnDA							.786**	.517**	.157	.537**	-.094	.698**	.348	.034	-.093	.042	-.001
PFDoDA								.646**	.467*	.427*	-.010	.508**	.757**	.126	.160	.191	.062
PFTrDA									.490*	-.227	.207	.142	.580**	.292	.201	.177	.671**
PFTeDA										-.131	.415*	.066	.640**	.582**	.124	.371	.342
PFBS											-.144	.704**	.065	-.165	-.021	-.086	-.579*
PFPeS												.108	.187	.391*	.167	-.127	.269
PFHxS													.297	.165	.044	-.042	-.245
PFOS														.328	.277	.248	.226
HFPO-DA															-.024	.354	.281
6:2 Cl-PFESA																.424*	-.158
6:2 FTSA																	.028

*: Correlation is significant at the 0.05 level (2-tailed).

**: Correlation is significant at the 0.01 level (2-tailed).

The revisions to the main text are as follows:

The Pearson correlation coefficients were further investigated between the PFASs in APM (Table S7-S8), a significant correlation generally indicated similar sources, transport processes and transformation processes for the two components (Lai et al., 2016). It should be noted that PFASs with a detection rate lower than 20% were excluded from the correlation data analysis according to Jian Zhou et al. (2021). Moderate to strong correlations were shown between PFCAs, suggesting that PFCAs in the atmosphere from Laoshan and Xisha Islands may originate from common sources, such as atmospheric transport. In Laoshan, PFOS showed moderate to strong correlations with PFCAs, especially PFOA (r = 0.832, p = 0.000) and PFDA (r = 0.806, p = 0.000). HFPO-DA was found to be moderately correlated with PFBA (r = 0.588, p = 0.002), PFOA (r = 0.668, p = 0.000) and PFTeDA (r = 0.582, p = 0.002), while PFOSA only showed moderate correlation with PFTrDA (r = 0.671, p = 0.000). Both 6:2 Cl-PFESA and 6:2 FTSA showed weaker and less significant correlations with others, except for 6:2 FTSA and PFBA (r = 0.646, p = 0.000). In Xisha Islands, PFOA as the predominant PFASs showed significantly positive correlations with PFHxA (r = 0.868, p = 0.000), PFNA (r = 0.855, p = 0.000), PFDA (r = 0.906, p = 0.000) and PFDoDA (r = 0.907, p = 0.000). As an alternative to PFOS, 6:2 FTSA was found to be moderately correlated with PFPeS (r = 0.669, p = 0.000).

Comment 9: Section 3.3, Source Apportionment: How distinct are the different groupings? An individual PFAS has many uses, and in addition to direct emissions, PFCAs can also form from atmospheric degradation of FTOHs.

Response: Thank you for this critical insight. In principal component analysis (PCA), eigenvalues represent the variance of data after dimensionality reduction and also indicate the amount of original information carried by each component. The groups with eigenvalues greater than 1 were interpreted as source components. Differences between groups are determined by the distinct characteristic substances selected for each group. PFASs in each group with the load greater than 0.8 were selected as the characteristics to display the main pollutant source. Each characteristic PFAS in a group may have multiple sources, only the common sources of these characteristic PFASs were identified as the source of the corresponding group. Tables S10 have been revised as follows:

Table S10 Source profiles of PFASs in Laoshan obtained from PCA-MLR models (*n*=26)

Species	KMO measure	Rotated Component Coefficients	
		F1	F2
PFBA	.941	0.691	0.472
PFHxA	.733	0.511	0.657
PFHpA	.678	0.776	0.44
PFOA	.700	**0.899**	0.167
PFNA	.643	-0.027	**0.884**
PFDA	.733	0.605	0.67
PFUnDA	.651	0.005	**0.963**
PFDoDA	.794	0.446	**0.843**
PFTrDA	.946	0.452	0.592
PFTeDA	.706	**0.811**	0.137
PFOS	.829	0.758	0.418
HFPO-DA	.519	0.76	-0.104
Eigenvalue		7.055	1.973
% of Variance		58.8	16.4
Cumulative % of Variance		58.8	75.2
MLR results			
Possible sources		fluoropolymer manufacturing	material intermediates preparation /fluoropolymer processing aids
Profile contributions		0.902	0.344
Source contributions (%)		72.4%	27.6%

The total KMO test :.739;

Bartlett’s test :.000;

The values with bold font represent the components with positive loading greater than

The revisions to the main text are as follows:

“In Laoshan, three principal components explain the sources of 82.6% of PFASs in the atmosphere at this sampling site. FL1 accounted for 56.7% of the total variances, among which PFUnDA and PFNA are in high loading of 0.976 and 0.930, respectively. PFUnDA was used for the preparation of material intermediates (Xiao et al., 2012); PFNA has been used for many decades as an essential “processing aid” in the manufacture of pfluoropolymers (Buck et al., 2011), thus FL1 was interpreted as the source of material intermediates preparation and fluoropolymer processing aids. FL2 explained 15.2% of the total variances and was characterized by HFPO-DA with high loading of 0.938, which was used as PFOA alternative in the fluoropolymer manufacturing industry (Wang et al., 2013). FL3 explained 10.7% of the total variances, among which PFHpS and PFOS are the marker of pollutants with loading of 0.948 and 0.801, respectively. PFOS has been widely used in the metal electroplating industry in Qingdao city, China (Wang et al., 2020), and the fluorine industry usually produces PFOS and other PFSAs by electrofluorination derivatization(Liu et al., 2015), therefore, FL3 was defined as the source of metal electroplating and electrochemical industry." It has been revised as "In Laoshan, two principal components explain the sources of 75.2% of PFASs. FL1 accounted for 58.8% of the total variances, among which PFOA and PFTeDA have high loadings of 0.899 and 0.811, respectively. PFOA is commonly used in the fluoropolymer manufacturing industry (Meng et al., 2017); PFTeDA is found in industrial and commercial products including photographic films, firefighting foams, detergents, and insecticides (Patel et al., 2022). Thus, FL1 was interpreted as the source of fluoropolymer manufacturing. FL2 explained 16.4% of the total variances and was characterized by PFUnDA, PFNA, and PFDoDA with high loadings of 0.963, 0.884, and 0.843, respectively. PFUnDA was used for the preparation of material intermediates (Xiao et al., 2012); PFNA has been used for many decades as an essential "processing aid" in the manufacture of fluoropolymers (Buck et al., 2011). Therefore, FL2 was interpreted as the source of material intermediates preparation and fluoropolymer processing aids."

"The results showed that in Laoshan, the fluoropolymer manufacturing sources FL2 contributed 46.9% to the $\sum_{13}$PFASs, followed by the metal plating and electrochemical sources (36.3%, FL3), the metal electroplating and electrochemical sources (16.8%, FL1) the material intermediates preparation and fluoropolymer processing aids. The 100% (25.6 pg/m^3) of the observed $\sum_{13}$PFASs was explained by PCA-MLR model. These three sources represented the average concentration contributions of 4.3, 12.0 and 9.6 pg/m^3 to the $\sum_{13}$PFASs, respectively (Table S9)."

has been revised as "The results showed that in Laoshan, the fluoropolymer manufacturing sources FL1 contributed 72.4% to the $\sum_{12}$PFASs, followed by the material intermediates preparation and fluoropolymer processing aids (27.6%, FL2), which could represented the average concentration contributions of 18.5 and 7.1 pg/m^3 to the $\sum_{12}$PFASs, respectively (Table S10)."

"The main sources of PFASs in Laoshan area are fluoropolymer manufacturing and metal electroplating and electrochemistry. The Xisha Islands are mainly based on textile treatment and precious metals, but a small part is still derived from metal plating and electrochemistry. This is due to the industrial structure in different regions." has been revised as "Generally speaking, the main sources of PFASs in the Laoshan area may be fluoropolymer manufacturing and material intermediates preparation, while the main sources of PFASs in Xisha Islands may be textile treatment and precious metals, indicating the different industrial structure between Laoshan and Xisha Islands."

Beyond direct contributions, there are indeed indirect contributions—for example, certain substances can transform into other PFASs in the atmosphere (e.g., FTOHs converting to PFCAs). However, for atmospheric PFASs, the proportion of PFASs derived from such indirect sources is relatively small. Thus, this study primarily focuses on PFAS sources from direct emissions. We will add a note on limitations in the discussion of this section (Lines 276–278): " It should be noted that the present study focused on analyzing the direct emission sources of atmospheric PFASs and the impacts of indirect sources (such as the transformation of different PFASs in the atmosphere) was ignored."

Comment 10: Lines 278-290: PCA-MLR provides evidence but not proof. I suggest the authors use conditional language for their conclusions. For example, "The main sources of PFASs in Laoshan area may be..." or something similar.

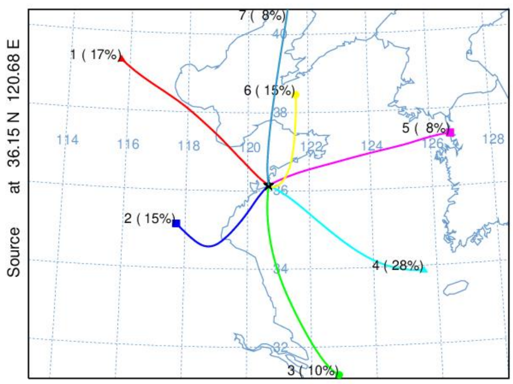
Response: We agree completely and thank you for this suggestion. We have revised the language throughout Section 3.3 to be more conditional. For example (Lines 307-309):

Original: "The main sources of PFASs in Laoshan area are fluoropolymer manufacturing and metal electroplating and electrochemistry."

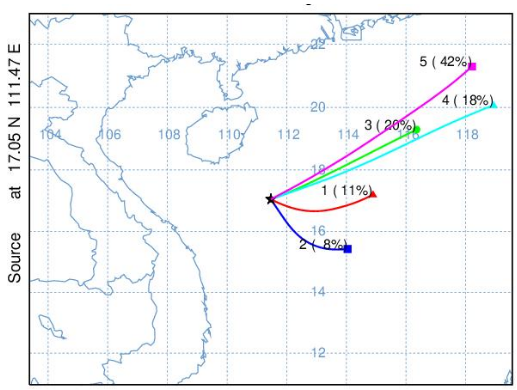
Revised as: "Generally speaking, the main sources of PFASs in the Laoshan area may be fluoropolymer manufacturing and material intermediates preparation, while the main sources of PFASs in Xisha Islands may be textile treatment and precious metals, indicating the different industrial structure between Laoshan and Xisha Islands." This change has been applied to all conclusive statements in this section.

Comment 11: Figure 6: I do not understand the display of dual-source backward trajectory clusters. The caption says that (c) and (d) show different sampling time periods. When are the periods?

Response: We apologize for the lack of clarity in the original figure caption. Panels (c) and (d) in Figure 6 could illustrate two distinct time periods selected from the HYSPLIT analysis Backward trajectory (120-hour) simulations, which were carried out from 15th March 2021 to 16th May 2021 at both sites. And Panels (c) and (d) were selected as representatives that could clearly demonstrate the existence of common air mass transport pathways between the two sites. Panels (c) and Panels (d) were the 120-hours backward trajectory from 15th March 2021, 22nd April 2021, respectively. The information has been added in Figure 6.



a



b

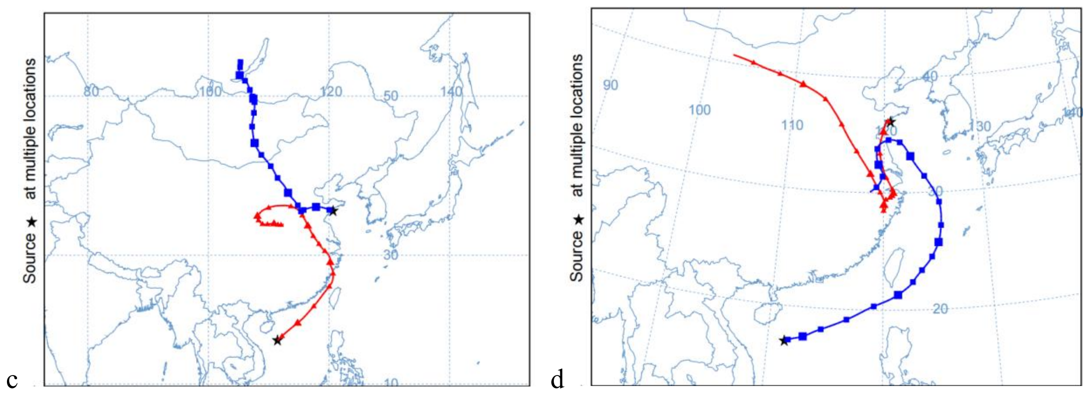


Fig. 6. Backward clustering trajectories at the sampling sites of Laoshan (a) and Xisha Islands (b). Dual-source backward clustering trajectories at the sampling sites of Laoshan and Xisha Islands in different sampling time periods including (c, 5th March 2021) and (d, 22nd April 2021)

Comment 12: Tables S5 and S6: It would be helpful to add row(s) with some summary statistics like min-max range, average and standard deviation.

Response: Thank you for your suggestion. We have added summary rows to both Tables S5 and S6 showing the min-max range, Mean, and Standard Deviation for the concentration of each PFAS across all samples from each location. It has been revised as follows:

Table S5 Concentrations of 30 legacy and emerging PFASs (19 PFAS were detected) in Laoshan atmosphere (pg/m^3)

	PFBA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFPeS	PFHxS	PFHpS	PFOS	HFPO-DA	6:2 Cl-PFESA	6-2 FTSA	PFOSA	Mean±SD	Min-Max	ΣPFASs
20210417	0.92	0.93	0.15	24.5	0.33	0.08	0.17	<0.071	n.d.	n.d.	0.64	0.43	<0.197	n.d.	0.81	<0.128	n.d.	<0.012	<0.069	2.90±7.60	n.d.-24.5	29.3
20210418	2.02	2.02	2.01	29.5	0.60	0.39	0.24	0.13	0.41	n.d.	n.d.	0.66	0.24	0.092	2.63	0.24	n.d.	<0.012	<0.069	2.94±7.69	n.d.-29.5	40.9
20210419	4.52	3.85	6.62	48.0	1.57	1.53	1.14	0.87	1.71	1.77	0.92	0.30	0.41	0.166	5.34	0.15	0.84	0.33	<0.069	4.45±11.04	<0.069-48	80.1
20210420	n.d.	0.63	0.69	10.4	0.54	0.14	0.20	0.09	0.37	1.05	n.d.	0.43	0.27	n.d.	0.59	<0.128	n.d.	<0.012	<0.069	1.28±2.88	n.d.-10.4	15.6
20210421	0.65	1.09	1.44	7.72	0.78	0.41	0.45	0.26	0.65	1.11	0.60	0.30	0.21	n.d.	0.91	n.d.	n.d.	<0.012	<0.069	1.18±1.91	n.d.-7.72	16.6
20210422	2.71	1.83	n.d.	12.3	2.32	0.22	1.31	0.46	n.d.	n.d.	4.58	n.d.	1.15	n.d.	1.69	n.d.	n.d.	0.04	n.d.	2.60±3.47	n.d.-12.3	28.5
20210423	0.72	0.43	<0.044	9.10	1.12	0.14	0.39	0.09	0.43	n.d.	0.71	n.d.	<0.197	n.d.	0.31	n.d.	n.d.	<0.012	<0.069	1.34±2.74	n.d.-9.1	13.7
20210425	1.60	1.29	0.98	20.4	1.10	0.33	0.36	0.19	0.51	1.14	0.84	0.69	0.37	n.d.	2.19	n.d.	0.86	<0.012	<0.069	2.19±5.07	n.d.-20.4	32.9
20210426	n.d.	0.87	0.49	15.3	1.11	0.20	0.45	0.19	0.85	n.d.	1.61	1.89	0.46	n.d.	1.12	n.d.	1.12	<0.012	<0.069	1.97±4.04	n.d.-15.3	25.6
20210427	2.47	1.67	0.31	11.6	3.33	0.45	1.26	0.38	1.63	n.d.	n.d.	n.d.	0.61	n.d.	1.67	n.d.	n.d.	0.02	0.10	1.96±3.06	n.d.-11.6	25.5
20210428	5.56	1.82	2.88	28.2	1.08	0.35	0.50	0.23	0.75	1.90	n.d.	0.31	0.29	n.d.	1.56	0.62	0.94	3.07	<0.069	3.13±6.83	n.d.-28.2	50
20210429	4.08	2.24	4.15	46.6	1.99	0.63	0.77	0.38	1.35	3.60	n.d.	3.60	0.71	n.d.	4.45	0.95	n.d.	0.02	0.09	4.73±11.28	n.d.-46.6	75.6
20210430	1.24	0.37	0.11	8.82	0.46	<0.062	0.16	<0.071	n.d.	n.d.	n.d.	0.19	0.24	n.d.	0.51	n.d.	n.d.	<0.012	<0.069	1.34±2.82	n.d.-8.82	12.3
20210501	0.86	0.49	0.34	11.0	0.45	0.11	0.18	0.08	0.34	n.d.	n.d.	1.29	0.23	n.d.	1.08	n.d.	n.d.	n.d.	<0.069	1.37±3.06	n.d.-11	16.5
20210502	n.d.	0.59	0.07	5.60	0.38	0.10	0.16	<0.071	0.34	n.d.	n.d.	0.34	0.21	n.d.	0.49	n.d.	0.50	<0.012	<0.069	0.80±1.60	n.d.-5.6	8.89
20210503	n.d.	n.d.	0.11	13.6	0.89	0.09	0.30	0.10	0.62	n.d.	n.d.	2.64	0.53	n.d.	0.68	n.d.	0.89	<0.012	<0.069	1.86±3.96	n.d.-13.6	20.5
20210505	1.06	1.24	1.45	20.5	0.79	0.35	0.29	0.16	0.46	1.10	n.d.	0.79	0.29	n.d.	2.69	0.17	n.d.	<0.012	<0.069	2.24±5.30	n.d.-20.5	31.3
20210506	2.58	0.66	1.41	37.2	1.65	0.92	0.62	0.51	0.93	1.47	0.83	n.d.	0.51	0.284	5.37	n.d.	0.81	1.13	<0.069	3.56±9.05	n.d.-37.2	57
20210508	0.72	0.52	n.d.	6.12	0.49	n.d.	0.26	n.d.	n.d.	n.d.	n.d.	0.34	0.38	n.d.	0.63	<0.128	1.03	<0.012	n.d.	1.17±1.87	n.d.-6.12	10.6
20210509	0.56	0.47	n.d.	3.18	0.42	<0.062	0.25	n.d.	n.d.	n.d.	n.d.	n.d.	0.29	n.d.	0.39	n.d.	n.d.	n.d.	<0.069	0.79±1.06	n.d.-3.18	5.65
20210510	1.76	1.08	1.10	34.4	1.47	0.31	0.47	0.17	0.61	n.d.	0.86	<0.115	0.52	n.d.	0.79	0.72	n.d.	<0.012	<0.069	3.40±9.32	n.d.-34.4	44.4
20210512	0.51	0.80	1.36	6.43	1.18	0.34	0.49	0.21	0.59	1.52	0.84	n.d.	0.33	n.d.	0.80	n.d.	n.d.	<0.012	<0.069	1.18±1.63	n.d.-6.43	15.4
20210513	0.45	0.91	2.91	5.19	2.01	0.69	0.68	0.25	0.50	1.12	0.60	2.06	<0.197	n.d.	0.56	0.09	n.d.	<0.012	<0.069	1.29±1.38	n.d.-5.19	18.2
20210514	1.73	3.00	2.54	5.98	2.11	0.49	1.02	0.37	n.d.	n.d.	2.56	0.39	0.74	n.d.	1.10	n.d.	n.d.	n.d.	n.d.	1.84±1.60	n.d.-5.98	22.1

20210515	1.71	0.94	0.23	5.63	2.08	0.30	1.00	0.19	0.96	n.d.	n.d.	n.d.	0.38	n.d.	0.50	n.d.	n.d.	n.d.	<0.069	1.27±1.57	n.d.-5.63	14.0
20210516	0.87	0.91	0.75	3.34	1.26	0.18	0.62	0.19	0.86	n.d.	n.d.	n.d.	0.34	n.d.	0.83	n.d.	n.d.	<0.012	<0.069	0.92±0.87	n.d.-3.34	10.2

Table S6 Concentrations of 30 legacy and emerging PFASs (14 PFAS were detected) in Xisha Islands TSP samples (pg/m^3)

	PFBA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFPeS	PFHxS	PFOS	ADONA	6-2 FTSA	Mean±SD	Min-Max	ΣPFASs
20210305d[a]	n.d.	0.82	0.08	3.03	1.69	1.33	1.00	0.77	n.d.	n.d.	n.d.	0.43	n.d.	n.d.	0.65±0.89	n.d.-3.03	9.15
20210305n[b]	0.50	0.37	<0.044	1.36	0.98	0.61	0.54	0.34	0.57	0.15	0.52	<0.381	n.d.	n.d.	0.50±0.39	n.d.-1.36	6.36
20210306d	0.20	0.14	<0.044	0.92	0.64	0.47	0.33	0.22	n.d.	n.d.	0.26	<0.381	n.d.	n.d.	0.27±0.29	n.d.-0.92	3.59
20210307d	n.d.	0.00	<0.044	1.24	0.78	0.57	0.37	0.25	0.47	<0.115	0.50	<0.381	n.d.	0.01	0.38±0.39	n.d.-1.24	4.68
20210307n	0.46	0.37	<0.044	1.14	0.78	0.54	0.45	0.28	0.54	n.d.	0.55	<0.381	n.d.	0.01	0.43±0.34	n.d.-1.14	5.53
20210308d	1.00	0.67	0.06	1.69	1.49	1.01	0.87	0.49	n.d.	n.d.	n.d.	0.54	n.d.	n.d.	0.56±0.59	n.d.-1.69	7.82
20210308n	0.49	0.41	<0.044	1.39	1.16	0.62	0.60	0.31	0.48	0.12	0.52	0.60	n.d.	0.01	0.52±0.4	n.d.-1.39	6.74
20210309n	1.84	1.26	0.12	4.42	2.54	1.60	1.22	0.79	1.62	n.d.	2.05	0.78	n.d.	n.d.	1.30±1.22	n.d.-4.42	18.2
20210310d	1.09	0.98	0.08	2.40	1.99	1.14	0.96	0.55	0.00	n.d.	1.26	0.96	n.d.	n.d.	0.82±0.77	n.d.-2.4	11.4
20210310n	0.50	0.48	<0.044	1.41	1.16	0.60	0.53	0.28	0.57	n.d.	0.58	0.41	n.d.	0.01	0.50±0.42	n.d.-1.41	6.57
20210311d	0.55	0.53	<0.044	1.26	1.24	0.61	0.56	0.28	0.59	n.d.	0.54	<0.381	n.d.	n.d.	0.51±0.42	n.d.-1.26	6.58
20210311n	0.48	0.40	<0.044	1.10	1.22	0.58	0.49	0.26	0.59	n.d.	0.52	<0.381	n.d.	n.d.	0.47±0.39	n.d.-1.22	6.06
20210312d	0.60	0.27	0.04	1.18	1.10	0.59	0.45	0.39	0.35	<0.115	0.31	<0.381	n.d.	n.d.	0.44±0.39	n.d.-1.18	5.74
20210312n	0.47	0.38	<0.044	1.17	1.64	0.65	0.66	0.30	0.63	<0.115	0.54	<0.381	n.d.	n.d.	0.59±0.48	n.d.-1.64	6.93
20210313d	0.60	0.61	0.14	1.58	1.93	0.78	0.67	0.34	0.68	n.d.	0.62	0.87	n.d.	0.02	0.63±0.57	n.d.-1.93	8.84
20210313n	0.57	0.68	0.15	1.99	1.70	0.88	0.85	0.42	0.86	n.d.	0.57	0.76	n.d.	0.01	0.67±0.60	n.d.-1.99	9.45
20210314d	0.96	0.88	<0.044	3.64	2.56	1.56	1.63	0.82	1.95	0.15	0.97	0.60	n.d.	0.02	1.21±1.05	n.d.-3.64	15.8
20210314n	0.47	0.45	<0.044	2.07	1.75	1.25	1.28	0.65	1.36	n.d.	0.52	0.61	n.d.	0.01	0.80±0.68	n.d.-2.07	10.5
20210315d	0.47	0.40	<0.044	1.46	1.08	0.71	0.70	0.35	0.54	<0.115	0.54	<0.381	n.d.	<0.012	0.63±0.40	n.d.-1.46	6.75
20210315n	0.46	0.46	<0.044	1.57	1.58	0.87	0.92	0.43	0.94	<0.115	0.52	<0.381	1.19	0.01	0.81±0.50	n.d.-1.58	9.44
20210316d	1.86	0.78	0.13	2.53	1.88	1.62	1.10	0.43	n.d.	0.34	2.06	<0.381	4.26	0.03	1.31±1.23	n.d.-4.26	17.4
20210316n	0.45	0.41	<0.044	0.96	0.92	0.58	0.51	0.29	0.57	<0.115	n.d.	<0.381	n.d.	0.01	0.43±0.34	n.d.-0.96	5.21
20210317d	n.d.	0.31	0.08	1.57	1.06	1.04	1.27	0.26	n.d.	0.22	0.66	n.d.	1.57	0.04	0.58±0.60	n.d.-1.57	8.07

[a] d is present sampling in daytime, [b] n is present sampling in nigh

Technical Corrections Response: We thank the reviewer for identifying these errors. They have all been corrected in the revised manuscript.

(1) Line 210: Line 210: ADONA is misspelled.

Response: "ADNOA" has been corrected to "ADONA".

(2) Figure 3: The x axis is missing a title (date in March 2021). The figure caption should indicate that the red line goes with the right axis.

Response: Figure 3 has been revised as Fig S1: The x-axis title "Date in March 2021" has been added. The caption now specifies "The red line (∑PFASs) corresponds to the right axis."

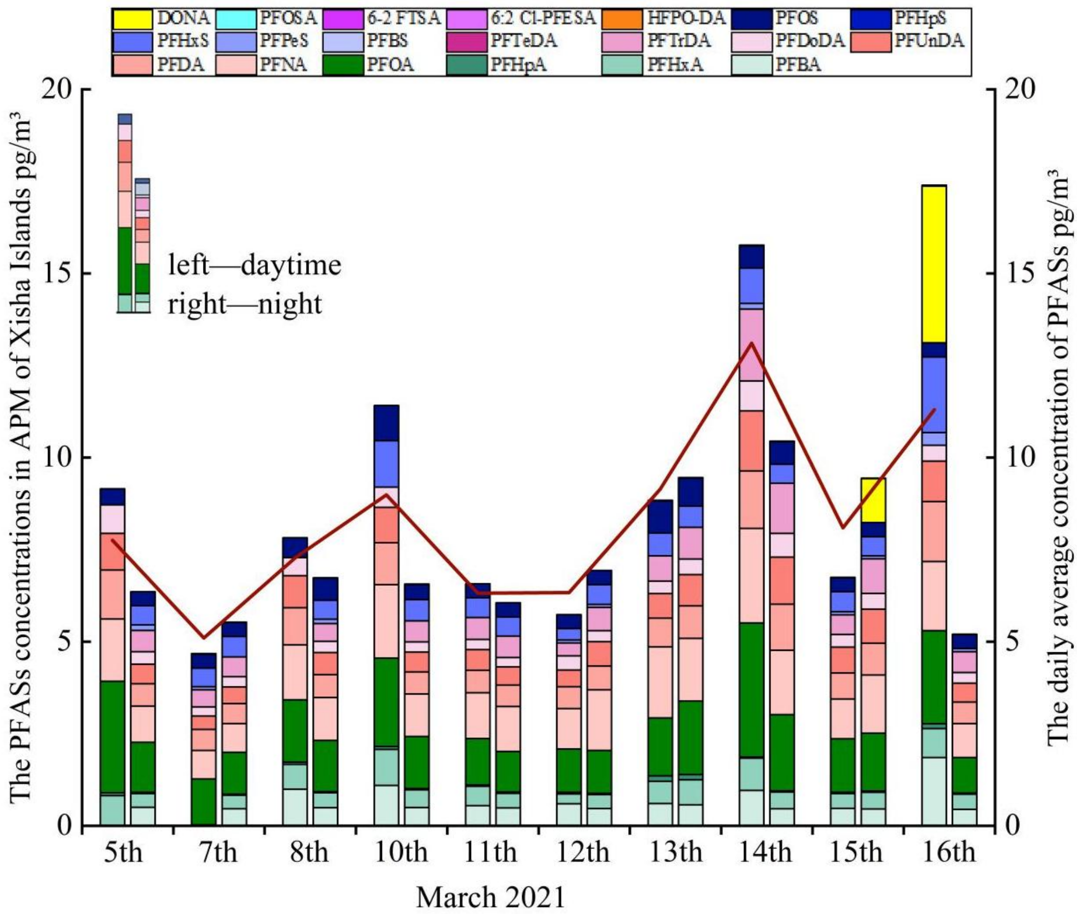


Fig S1. Concentrations (pg/m^3) and proportion (%) characteristics of PFASs in TSP of Xisha Islands, China. Note: Values corresponding to the red line are referenced to the right axis.

(3) I advise the authors to use the acronym LC-PFCAs for long chain PFCAs because L-PFCAs could be misinterpreted as linear PFCAs.

Response: Thank you for this suggestion. We have replaced "L-PFCAs" with

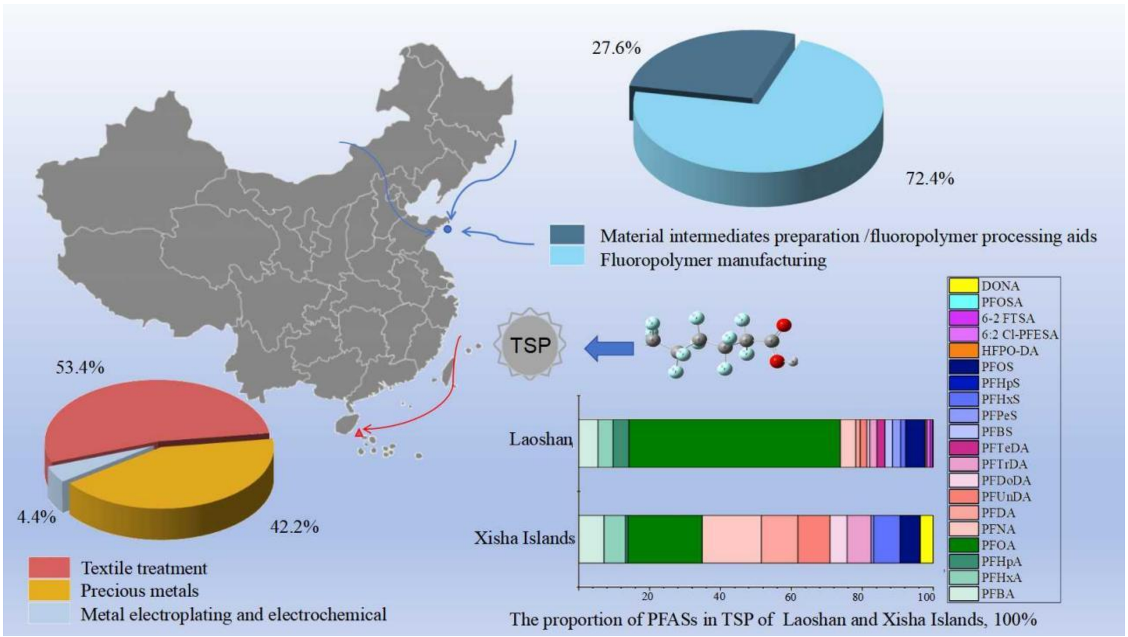
"LC-PFCAs" throughout the manuscript (e.g., Line 197, Fig. 4c) to avoid confusion with "linear PFCAs".

(4) What type of correlation analysis did the authors conduct? Line 118 says Spearman, but line 225 says Pearson.

Response: Thank you for this suggestion. We used Pearson correlation analysis for this study. We have corrected Line 130 to "Pearson correlation coefficients" to be consistent.

(5) TOC art: There is a lot of information in this figure. It will likely be difficult to interpret at scale.

Response: Thank you for this suggestion. We have simplified the TOC/Abstract art figure to improve clarity and legibility when scaled down. It has been revised as follws:



(6) Text S1, third line of first paragraph: Internal standard mix should be MPFAC-MXA.

Response: Text S1: "MPFAC-MAX" has been corrected to "MPFAC-MXA".

Reviewer 2#

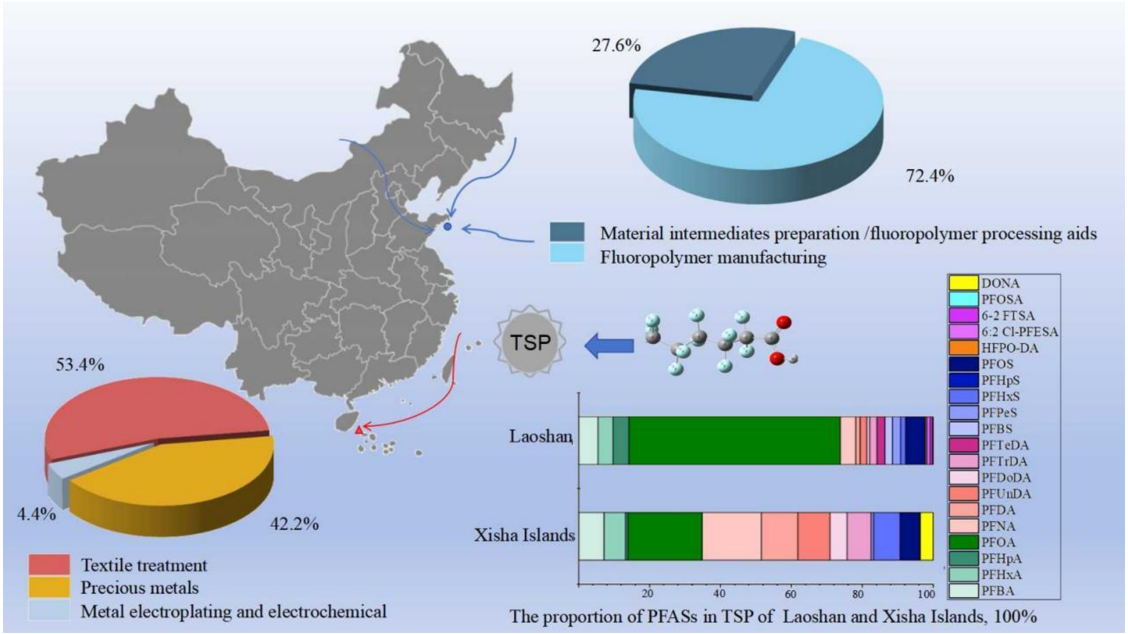
General comment:

While potentially interesting, the poor grammar and odd phrasing make it difficult to follow the manuscript. Typos and missing spaces/words contribute further to this problem. The novelty aspect of the manuscript is also questionable, mainly because the Introduction does not provide sufficient context for understanding the goal of the study. The choice of study locations is not well described, nor does it become clear until the Results section that sampling occurred on a ship. That some of the differences in observations are attributed to the movement of the ship during sampling seems like an overall flaw in the study design that needs to be addressed (Line 197-205). Several of the conclusions (e.g., line 152-153, line 188, line 302-304) are not sufficiently supported by the data or references provided. Thus, I strongly suggest that the authors revise their manuscript to improve the overall presentation before it can be considered for publication. This includes the title of the manuscript and the abstract.

Response: Thank you for the constructive suggestions. We have revised the manuscript thoroughly and added more detailed information about sampling campaign. Moreover, further data analysis and related references have been added to support the conclusions.

Comment 1: I also recommend that the authors simplify the TOC Art, which includes too much information with very small fonts, making it difficult to interpret.

Response: Thank you for this suggestion. We have simplified the TOC/Abstract art figure to improve clarity and legibility when scaled down. It has been revised as follows:



Comment 2: Additional details about how, when, and where samples were collected should be included in the main text.

Response: We appreciate your suggestions. We had revised detailed descriptions of how, when, and where samples were collected in the "Sample Collection" section of the Materials and methods. The original text: "In March 2021, atmospheric suspended particulate matter was sampled in the Xisha Islands with 12 h at day and 12 h at night, with a total of 23 samples." has been revised as: "From March 5 in 2021 to March 17 in 2021, TSP samples were collected among the Xisha Islands of Hainan Province by ship. Nearly 12 h samples were collected during day and night on the voyage, respectively. Finally, a total of 23 samples were obtained." Specific sampling information of the Laoshan and Xisha Islands was presented in Table S1 and Table S2 of the Supplementary Materials.

Table S1 The date, time, volume and meteorological parameters during the sampling campaign in Laoshan.

Date	Number	Time	Volume[a]	Weather	AQI
		36.15°N，120.68°E			
20210416	LS-01	7:53~7:58	field blank	cloudy	194
20210417	LS-02	8:03~7:34 (next day)	440.97	sunny	72
20210418	LS-03	8:00~7:30 (next day)	434.46	sunny	37
20210419	LS-04	8:00~7:30 (next day)	434.46	sunny	52
20210420	LS-05	8:05~7:40 (next day)	448.73	overcast	58
20210421	LS-06	8:00~7:35 (next day)	461.78	rainy	41
20210422	LS-07	8:00~11:15	60.04	rainy	36
20210423	LS-08	8:10~7:38 (next day)	420.55	sunny	37
20210425	LS-09	8:17~7:40 (next day)	431.99	sunny	59
20210426	LS-10	8:05~17:42	196.14	rainy	69
20210427	LS-11	8:25~13:50	110.99	cloudy	68
20210428	LS-12	8:12~21:12	254.55	cloudy	108
20210429	LS-13	8:16~7:30 (next day)	134.15	rainy	69
20210430	LS-14	8:06~7:55 (next day)	453.18	rainy	42
20210501	LS-15	8:09~8:15 (next day)	458.57	sunny	45
20210502	LS-16	8:30~7:40 (next day)	440.82	sunny	34
20210503	LS-17	8:15~21:00	235.55	rainy	41
20210505	LS-18	8:11~8:00 (next day)	446.77	sunny	74
20210506	LS-19	8:22~8:00 (next day)	449.68	cloudy	107
20210508	LS-20	20:16~8:55 (next day)	233.70	cloudy	59
20210509	LS-21	9:06~22:12	242.02	rainy	44
20210510	LS-22	13:36~7:39 (next day)	333.46	overcast	45
20210512	LS-23	15:37~8:10 (next day)	318.09	sunny	33
20210513	LS-24	8:22~7:14 (next day)	447.76	sunny	36
20210514	LS-25	7:25~13:18	108.69	rainy	34
20210515	LS-26	9:30~19:13	195.65	rainy	26
20210516	LS-27	20:18~7:58 (next day)	215.54	overcast	26

a: the total volume of each sample at normal atmospheric pressure, m^3

Table S2 The date, time, volume, position and type of Xisha Islands samples.

Date	Number	Time	Total time	Volume[a]	Position (start-end)		Type
20210305	field blank2	13:45~13:50			Tanmen Port		Blank
	XS-01	14:00~18:30	4h30min	89.06	Tanmen Port	**N** 18°46.461‘ **E** 110°57.244’	Day
	XS-02	18:50~6:30 (next day)	11h40min	230.90	**N** 18°46.461‘ **E** 110°57.244’	**N** 17°21.686‘ **E** 111°53.490’	Night
20210306	XS-03	6:50~11:50 12:00~18:30	11h30min	227.60	**N** 17°21.686‘ **E** 111°53.490’	**N** 16°50.306‘ **E** 112°19.643’	Day
20210307	XS-04	6:52~18:30	11h38min	230.24	**N** 16°50.306‘ **E** 112°19.643’	**N** 16°58.596‘ **E** 112°16.065’	Day
	XS-05	18:40~6:33 (next day)	11h53min	238.18	**N** 16°58.596‘ **E** 112°16.065’		Night
20210308	XS-06	6:47~12:54	6h07min	122.60	**N** 16°58.294‘ **E** 112°16.051’	**N** 16°28.483‘ **E** 111°44.193’	Day
	XS-07	18:40~6:40 (next day)	12h	237.49	**N** 16°28.483‘ **E** 111°44.193’		Night
20210309	XS-08	19:00~22:15	3h15min	64.32	**N** 16°28.133‘ **E** 111°43.974‘		Night
20210310	XS-09	7:00~9:40 16:30~18:30	4h40min	92.36	**N** 16°28.133‘ **E** 111°43.974’	**N** 16°28.531‘ **E** 111°43.581’	Day
	XS-10	19:00~6:30 (next day)	11h30min	227.60	**N** 16°28.531‘ **E** 111°43.581’		Night
20210311	field blank3	18:40~18:45			**N** 16°28.456‘ **E** 111°44.181’		Blank
	XS-11	7:00~18:30	11h30min	227.60	**N** 16°28.531 **E** 111°43.581’	**N** 16°28.456‘ **E** 111°44.181’	Day
	XS-12	19:00~6:30 (next day)	11h30min	227.60	**N** 16°28.456‘ **E** 111°44.181’		Night
20210312	XS-13	7:00~11:15 11:35~16:36	9h16min	183.40	**N** 16°28.456‘ **E** 111°44.181’	**N** 16°30.358‘ **E** 111°36.150’	Day
	XS-14	19:00~6:30 (next day)	11h30min	227.60	**N** 16°30.358‘ **E** 111°36.150’		Night
20210313	XS-15	7:00~16:57	9h57min	196.92	**N** 16°30.358‘ **E** 111°36.150’	**N** 16°28.029‘ **E** 111°43.894’	Day
	XS-16	19:00~6:30 (next day)	11h30min	230.50	**N** 16°28.029‘ **E** 111°43.894’		Night
202103014	XS-17	7:00~13:28	6h28min	127.98	**N** 16°28.029‘ **E** 111°43.894’	**N** 16°30.496‘ **E** 111°36.274’	Day
	XS-18	19:00~6:30 (next day)	11h30min	230.50	**N** 16°30.496‘ **E** 111°36.274’		Night
20210315	XS-19	7:00~16:12; 16:25~18:30	11h17min	223.31	**N** 16°30.496‘ **E** 111°36.274’	**N** 16°28.126‘ **E** 111°43.623’	Day

	XS-20	19:00~6:30(next day)	11h30min	227.60	**N** 16°28.126' **E** 111°43.623'		night
	XS-21	7:00~7:52	52min	56.40	**N** 16°28.126' **E** 111°43.623'		Day
20210316		16:31~18:30	1h59min		**N** 17°15.138' **E** 111°22.538'		
	XS-22	19:00~6:30 (next day)	11h30min	230.50	**N** 17°18.861' **E** 111°21.033'	**N** 18°46.438' **E** 110°51.676'	Night
20210317	XS-23	7:00~13:00	4h	88.29	**N** 18°50.321' **E** 110°49.943'	**N** 19°14.248' **E** 110°37.151'	Day

a: the total volume of each sample at normal atmospheric pressure, m^3**E** 111°43.623'

Comment 3: The authors mention "preprocessing" or "pretreatment" of samples, but no details are given. Please add what kind of preprocessing was done.

Response: Thank you for your question. Given the limited words in the manuscript, the specific details regarding the pretreatment had been detailed in Text S1 of the Supplementary Materials, and the specific content is as follows: Cut the quartz membrane with particles attached into thin strips about 0.5 cm wide, put into a 50 mL polypropylene centrifuge tube (PP tube), add 2 ng of mixed internal standards (PFAC-MAX), vortex for 30 s, and let stand overnight. The samples were extracted with 25 mL 0.1% NH_4OH/methanol in a sonication water bath for 30 min, centrifuged at 4000 r/min for 10 min and collecting the supernatant into new PP tubes. 10 mL 0.1% NH_4OH/methanol was added to the remaining part and the extraction procedure repeated. The two extracts were combined and evaporated to 5 mL under a gentle stream of dry nitrogen gas. The concentrated extracts were purified by Cleanert PestiCarb SPE cartridges (made of graphitized carbon, 500 mg/6 mL, Bonna-Angla Technologies, China). The PestiCarb cartridges were activated with 5 mL methanol, 5 mL of ultrapure water, and 5 mL of methanol at a rate of 1-2 drop (s) per second, respectively. The sample extracts were cleaned up with activated PestiCarb cartridges, the effluent was collected and eluted with 5 mL 0.1% NH_4OH/methanol. The combined eluates (~10 mL) were evaporated to 0.5 mL under a gentle stream of dry nitrogen gas, filtered through a 0.22 μm nylonfilter and transferred into an injection vial, and finally stored at 4 °C for analysis.

Comment 4: The interpretation of the results from the PCA requires more discussion. The assignment of sources to the factors seems somewhat arbitrary.

Response: Thank you for this critical insight. In principal component analysis (PCA), eigenvalues represent the variance of data after dimensionality reduction and also indicate the amount of original information carried by each component. The group with eigenvalues greater than 1 were interpreted as source components. Differences between groups are determined by the distinct characteristic substances selected for each group. PFASs in each group with the load greater than 0.8 were selected as the characteristics to display the main pollutant source. Each characteristic PFAS in a group may have multiple sources, only the common sources of these characteristic PFASs were identified as the source of the corresponding group. Tables S10 have been revised as follows:

Table S10 Source profiles of PFASs in Laoshan obtained from PCA-MLR models (*n*=26)

Species	KMO measure	Rotated Component Coefficients	
		F1	F2
PFBA	.941	0.691	0.472
PFHxA	.733	0.511	0.657
PFHpA	.678	0.776	0.44
PFOA	.700	**0.899**	0.167
PFNA	.643	-0.027	**0.884**
PFDA	.733	0.605	0.67
PFUnDA	.651	0.005	**0.963**
PFDoDA	.794	0.446	**0.843**
PFTrDA	.946	0.452	0.592
PFTeDA	.706	**0.811**	0.137
PFOS	.829	0.758	0.418
HFPO-DA	.519	0.76	-0.104
Eigenvalue		7.055	1.973
% of Variance		58.8	16.4
Cumulative % of Variance		58.8	75.2
MLR results			
Possible sources		fluoropolymer manufacturing	material intermediates preparation /fluoropolymer processing aids
Profile contributions		0.902	0.344
Source contributions (%)		72.4%	27.6%

The total KMO test :.739;

Bartlett's test :.000;

The values with bold font represent the components with positive loading greater than

The revisions to the main text are as follows:

"In Laoshan, three principal components explain the sources of 82.6% of PFASs in the atmosphere at this sampling site. FL1 accounted for 56.7% of the total variances, among which PFUnDA and PFNA are in high loading of 0.976 and 0.930, respectively. PFUnDA was used for the preparation of material intermediates (Xiao et al., 2012); PFNA has been used for many decades as an essential “processing aid” in the manufacture of pfluoropolymers (Buck et al., 2011), thus FL1 was interpreted as the source of material intermediates preparation and fluoropolymer processing aids. FL2 explained 15.2% of the total variances and was characterized by HFPO-DA with high loading of 0.938, which was used as PFOA alternative in the fluoropolymer manufacturing industry (Wang et al., 2013). FL3 explained 10.7% of the total variances, among which PFHpS and PFOS are the marker of pollutants with loading of 0.948 and 0.801, respectively. PFOS has been widely used in the metal electroplating industry in Qingdao city, China (Wang et al., 2020), and the fluorine industry usually produces PFOS and other PFSAs by electrofluorination derivatization(Liu et al., 2015), therefore, FL3 was defined as the source of metal electroplating and electrochemical industry.” It has been revised as “In Laoshan, two principal components explain the sources of 75.2% of PFASs. FL1 accounted for 58.8% of the total variances, among which PFOA and PFTeDA have high loadings of 0.899 and 0.811, respectively. PFOA is commonly used in the fluoropolymer manufacturing industry (Meng et al., 2017); PFTeDA is found in industrial and commercial products including photographic films, firefighting foams, detergents, and insecticides (Patel et al., 2022). Thus, FL1 was interpreted as the source of fluoropolymer manufacturing. FL2 explained 16.4% of the total variances and was characterized by PFUnDA, PFNA, and PFDoDA with high loadings of 0.963, 0.884, and 0.843, respectively. PFUnDA was used for the preparation of material intermediates (Xiao et al., 2012); PFNA has been used for many decades as an essential “processing aid” in the manufacture of fluoropolymers (Buck et al., 2011). Therefore, FL2 was interpreted as the source of material intermediates preparation and fluoropolymer processing aids.”

“The results showed that in Laoshan, the fluoropolymer manufacturing sources FL2 contributed 46.9% to the $\sum_{13}$PFASs, followed by the metal plating and electrochemical sources (36.3%, FL3), the metal electroplating and electrochemical sources (16.8%, FL1) the material intermediates preparation and fluoropolymer processing aids. The 100% (25.6 pg/m^3) of the observed $\sum_{13}$PFASs was explained by PCA-MLR model. These three sources represented the average concentration contributions of 4.3, 12.0 and 9.6 pg/m^3 to the $\sum_{13}$PFASs, respectively (Table S9)." has been revised as "The results showed that in Laoshan, the fluoropolymer manufacturing sources FL1 contributed 72.4% to the $\sum_{12}$PFASs, followed by the material intermediates preparation and fluoropolymer processing aids (27.6%, FL2), which could represented the average concentration contributions of 18.5 and 7.1 pg/m^3 to the $\sum_{12}$PFASs, respectively (Table S10)."

"The main sources of PFASs in Laoshan area are fluoropolymer manufacturing and metal electroplating and electrochemistry. The Xisha Islands are mainly based on textile treatment and precious metals, but a small part is still derived from metal plating and electrochemistry. This is due to the industrial structure in different regions." has been revised as "Generally speaking, the main sources of PFASs in the Laoshan area may be fluoropolymer manufacturing and material intermediates preparation, while the main sources of PFASs in Xisha Islands may be textile treatment and precious metals, indicating the different industrial structure between Laoshan and Xisha Islands."

Beyond direct contributions, there are indeed indirect contributions—for example, certain substances can transform into other PFASs in the atmosphere (e.g., FTOHs converting to PFCAs). However, for atmospheric PFASs, the proportion of PFASs derived from such indirect sources is relatively small. Thus, this study primarily focuses on PFAS sources from direct emissions. We will add a note on limitations in the discussion of this section (Lines 288–290): " It should be noted that the present study focused on analyzing the direct emission sources of atmospheric PFASs and the impacts of indirect sources (such as the transformation of different PFASs in the atmosphere) was ignored."

Comment 5: Please define "ADM".

Response: Thank you for your comment. However, we have not mentioned "ADM" in the text. We speculate that you may be referring to "APM", which is defined as the abbreviation for atmospheric particulate matter. We apologize for the inconsistent use of TSP (total suspended particles) and APM in the manuscript. All instances of "APM" in the text has now been revised to "TSP."

Comment 6: The comparison of the data to literature values (Line 154-166) could be simplified or presented as a table. At the same time, no actual discussion of the observed differences is provided, but this should be added for further context.

Response: Thank you for your suggestions. We have streamlined the comparison between our data and literature values, and the literature values have been presented in Table S6. Specific revisions (Lines 173–187) are as follows: As shown in Table S6, the PFOA levels in TSP from the Laoshan area were slightly higher than those in inland regions, including Guiyang in China (Yu et al., 2018a), Tsukuba in Japan (Ge et al., 2017), Geesthacht in Germany (Dreyer et al., 2015), as well as Jinju in South Korea and Delhi in India (Lin et al., 2020). Unlike Laoshan, these cities exhibit no direct industrial sources of PFOA emissions (Yu et al., 2018b; Lin et al., 2020). The levels of PFOA in Laoshan are also higher than those in coastal regions such as the Pearl River Delta (Liu et al., 2022), Xiamen in China (Wang et al., 2022), and Gujarat in India (Lin et al., 2020). The levels of PFOA in Laoshan were lower than Tianjin, Yantai, Jinan, and Changshu (Yu et al., 2018a; Yu et al., 2018b), as well as Weifang (Yao et al., 2017) in China. These cities have direct or indirect PFOA emission sources such as the fluorochemical industry and fluoropolymer manufacturing industry, and are simultaneously affected by atmospheric transport from surrounding industrial sources (Liu et al., 2017; Meng et al., 2017). The winter heating in northern cities leads to increased PM10 concentrations, which further exacerbates the adsorption and enrichment of PFOA (Yu et al., 2018b).

Table S6 PFOA in total suspended particles (TSP)

Country	City	Range($pg \cdot m^{-3}$)	Mean($pg \cdot m^{-3}$)	Ref.
China	Jinzhou	0.1-90.0	10.3	Yu et al., 2018b
	Tianjin	3.4-329.3	47.2	
	Yantai	0.8-362.9	30.7	
	Yancheng	0.7-24.0	8.3	
	Lianyungang	0.6-65.6	18.3	
	Beijing	/-18.8	12.5	Yu et al., 2018a
	Jinan	/-544	325	
	Nanjing	/-24.8	11.6	
	Changshu	/-3515	556	
	Guiyang	/-2.51	2.07	
	Xiamen	/	4.89	Wang et al., 2022
	Weifang	16.0-3850	/	Yao et al., 2017
	Guangzhou	/	7.9	Liu et al., 2023
	Zhuhai	/	8.0	
	Foshan	/	6.58	
	Shenzhen	/	5.62	
	Zhongshan	/	4.85	
	Maoming	/	3.91	
Japan	Tsukuba	1.2-5.4	2.6	Yamazaki et al., 2017
Germany	Geesthacht	0.1-4.8	0.7	Dreyer et al., 2015
South Korea	Jinju	0.21-7.84	1.47	Lin et al., 2020
India	Delhi	0.323-1.07	0.571	
India	Gujarat	0.12-2.06	0.558	

Comment 7: Line 240-244: Suggest moving this paragraph to the Methods section.

Response: Thank you for your suggestions. The original text of line 240-244 content has been moved to Lines 139–142 of the Methods section, with the specific content as follows: Principal component analysis and multiple linear regression (PCA-MLR) were implemented to analyzing pollution sources of PFASs. The species with poor linear correlation ($p>0.05$), Kaiser-Meyer-Olkin (KMO) value less than 0.5 were excluded to participating in principal component analysis.

Comment 8: A review article about PFAS in atmospheric aerosol particles (J.A. Faust, 2022, https://doi.org/10.1039/D2EM00002D) should be used to provide more context and motivation for the work.

Response: Thank you for your constructive comments. The study on highly relevant by J.A. Faust et al. and its related content that you recommended has provided strong support for our analytical work, both in its introduction and subsequent discussion. In Lines 41-43, we have added a sentence stating: "To date, the legacy and emerging PFASs have been detected in atmospheric aerosol particles in worldwide (Faust et al., 2021; Yamazaki et al., 2021)."

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