

Dear respected professor:

We appreciate the reviewers' comments, which surely improve our manuscript. According to reviewer's comments, we revised this manuscript carefully. All responses and answers are listed below. All revisions were marked as the highlighted text in the manuscript.

Q1: The authors might want to list the limitations of their study in a paragraph in the Results and Discussion. For example, if other researches asked you for advice on doing a study like this, and they had unlimited resources, what would you tell them to do differently?

A: The limitation of the current study includes, but not limited to, 3 parts: 1) the total numbers of sampling sites (16) were not enough since the Tibetan Plateau is about 250 km²; if resources are unlimited, 40 sampling sites are minimum; 2) sampling rates of SIP were estimated using absorption equation, while they should be calibrated using active air sampler that electricity is needed and air volume is accurate; 3) given south Asia is an important source region and the Yarlung Tsangpo Grand Canyon is a channel for receiving pollutants from southern Asia, deploying a sets of sampler along this Canyon can provide direct evidence regarding the monsoon transport of emerging pollutants, such as neutral per-fluoroalkyl substances, and cyclic volatile methylsiloxanes.

Q2: P3,L61 and 62: need a citation for there striction of VBS by European Chemical Agency.

A: A reference was included in line 62 and line 552-555.

Please see the following reference:

European Chemical Agency (ECHA). Identification of PBT and vPvB substance. Results of evaluation of PBT/vPvB properties for decamethylcyclopentasiloxane; 2012 [Available online at http://echa.europa.eu/documents/10162/13628/decamethyl_pbtSheet_en.pdf].

Q3: A few statistical analysis were performed. It would be better to let readers know how did you design and perform your statistical analysis?

A: There are two statistical analysis performed in this study, ANOVA and multiple linear regression, respectively. Analysis of Variance (ANOVA) consists of calculations that provide information about levels of variability within a regression model and form a basis for tests of significance. Multiple linear regression attempts to model the relationship between two or more explanatory variables and a response variable by fitting a linear equation to observed data. All these two statistical methods are common used methods. In the revision, some brief introductions about the methods are included. "One-way analysis of variance (ANOVA) was performed to determine the statistical differences in the values of individual chemicals among different sampling sites. If the *p*-value is lower than 0.05, we conclude the significant differences occur." Please see line 308-311.

For the multiple linear regressions, the R^2 values can be used to explain the variation of

the dependents. Please see line 432-433.

Q4: P14, L375: how "poor-relationship" was that between short- and long- chain PFASs?

A: Correlation coefficients ($r=0.283$) between concentrations of NMeFBSE and those of NMeFOSE, obtained by Li et al., (2011) is provided in line 384.

Reference:

Li, J., Vento, S. D., Schuster, J., Zhang, G., Chakraborty, P., Kobara, Y., and Jones, K. C.: Perfluorinated Compounds in the Asian Atmosphere, *Environ. Sci. Technol.*, 45, 7241-7246, 2011.

Q5: The author should use alternative scientific explanations for this.

A: Sorry about this comment. Since you did not mention the line number, I don't know which place need explanation.

Q6: P7, sample analysis: it would be better if more detail instrumental analysis information was provided in the main manuscript or supplementary information.

A: Detailed analysis regarding GC program and MS detection ions are provided in Text S2.

Thermo DSQ GC-MS was used for analysis of neutral PFASs. The GC temperature program is as follow follows: 50 °C (held 1 min), ramped at 3 °C/ min to 70 °C, ramped at 10 °C /min to 130 °C, then ramped at 20 °C/ min to 225 °C (held 11.4 min), and finally cooled at 80°C /min to 50°C (total run time 32min). A constant injection temperature of 200 °C was used, with a 2 mL splitless injection. Helium was employed as a carrier gas at a constant flow of 1 mL/min. The GC-MS transfer line temperature was set at 250 °C. Mass spectral analysis was performed in PCI-selected ion monitoring (PCI-SIM) mode, using methane as the reagent gas for quantification of target analytes (a seven point calibration curve (20, 50, 100, 200, 500, 1000 and 2000 pg injected) was used.

Quantification of cVMS was performed on a Trace GC Ultra (Thermo Electron Corp.) coupled to a MD800 MS detector (Fisons Instruments SpA) using electron ionization (EI). The GC temperature program incorporated an initial temperature of 40 °C with a hold time of 3 min, increased by 25 °C min⁻¹ to 190 °C, followed by a second temperature ramp of 40 °C min⁻¹ to 240 °C and held for 4 min. The MS was operated in selected ion monitoring (SIM) mode. The following ions were monitored in SIM mode: recording ions m/z 281/282 for D4, 355/267 for D5, 341/429 for D6, 284/285 for 13C4-D4, 360/270 for 13C5-D5, and 345/435 for 13C6-D6, respectively.

Please also see the following references:

- 1) Jonathan L. Barber, et al., Analysis of per- and polyfluorinated alkyl substances in air samples from Northwest Europe. *J. Environ. Monit.*, 2007, 9, 530–541
- 2) Nicholas .A. Warner, et al., Positive vs. false detection: A comparison of analytical methods and performance for analysis of cyclic volatile methylsiloxanes (cVMS) in environmental samples from remote regions. *Chemosphere* 2013, 93, 749-756
- 3) De-goWang, et al., Determination of cyclic volatile methylsiloxanes in water, sediment, soil, biota, and biosolid using large-volume injection–gas chromatography–mass spectrometry. *Chemosphere* 2013, 93, 741-748
- 4) Amelie Kierkegaard et al., Determination of linear and cyclic volatile methylsiloxanes in air at a regional background site in Sweden. *Atmospheric Environment*, 2013, 80, 322-329