Supporting information for

Polychlorinated naphthalene congener profiles in common vegetation on the Tibetan Plateau as bio-monitors of their sources and transportation

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- **Prediction of the atmospheric PCN concentrations on the Tibetan Plateau from the PCN concentrations in lichens**

In the Figure 2 (a), there is the significant relationship between log (K_{LA}/f_{OC}) and log K_{OA} (Equation 6). The K_{LA} could be predicted from the log K_{OA} of the PCN congeners and lipid content of the lichens (Equation S1). This also indicated that the PCN concentrations in the air could be estimated from the PCN concentrations in lichens (Equation 7).

$$log\ (K_{LA}/f_{OC}) = 0.28 logK_{OA} + 1.18 \qquad \text{(Equation 6)}$$

$$logK_{LA} = 0.28 logK_{OA} + logf_{OC} + 1.18 \qquad \text{(S1)}$$

$$logC_{air} = log\ C_{lichen} - 0.28 logK_{OA} - logf_{OC} - 1.18 \qquad \text{(Equation 7)}$$

Consequently, we calculated the predicted log K_{LA} values and predicted atmospheric PCN concentrations based on the log K_{OA} (Table S2) of the PCNs, lipid content of the lichens and PCN concentrations in lichens. Results were shown in Figure 3 (d and e). Measurements and predictions of the average PCN distributions were conducted using lichen and air samples that were collected at the same sites (N14, N17, and N20). Predicted and measured log K_{LA} values or PCN concentrations were compared for the PCN congeners with log K_{OA} values reported in Table S2.

- **Estimation of the PCN concentrations in lichens with altitude**

The relationship between the altitude and PCN congener concentrations in the lichens normalized to lipid content were shown in Table S3 and Equation S2. In this equation, a and b are constants (Table S3). i represents the PCN congener i; x represents the sampling site; H_x is the altitude of the site x.

$$\log(C_{lichen_i}/f_{OC}) = a \times H_x + b \qquad \text{(S2)}$$

The significant linear correlation between altitude and PCN concentration in lichens relative

to lipid content allows for estimation of the PCN concentrations in lichens. This result was estimated assuming that the only source of PCNs in the Tibetan Plateau region was LRAT, and ignoring local climate change.

- **Influence of temperature on the atmospheric PCN concentrations**

The concentration of the PCNs in lichens relative to lipid content were estimated to be linearly correlated with the altitude (within the investigated range) (Equation S2). Temperature should be linearly correlated with the altitudes (Equation S3). T_x represents the temperature at the site x, T_0 represents the temperature at the site 0, ΔT represents the temperature range decreased when the altitude increases 1 km, ΔH_x is the relative altitude of site x to site 0 (Equation S4). T_0, H_0 and ΔT are the constants.

$$T_x = T_0 - \Delta T \times \Delta H_x \qquad \text{(S3)}$$

$$\Delta H_x = H_x - H_0 \qquad \text{(S4)}$$

Combined with the relationship between the lichen–air partition coefficients and log K_{OA} of the PCNs (linear correlation with $1/T$, Table S2) in Equation S1, the relationship between the atmospheric concentrations of specific PCN congeners with the temperature could be predicted using Equation S8. p_i, r_i, and m_i are constants affected by the physicochemical properties of the PCN congeners and the characteristics of the lichens. Therefore, the concentrations of PCNs at site x would be influenced by the surrounding temperature at this site, the physical properties of the PCN congener itself, and the characteristics of the lichens. Variation of $\frac{r_i}{T_x}$ for specific PCN congeners was small among the sampling sites, which meant that the relationship between temperature and atmospheric concentration could almost be regarded as a linear.

$$\log C_{air_{xi}} = p_i T_x + \frac{r_i}{T_x} + m_i \qquad \text{(Equation 8)}$$

Table S1. Basic information for the sampling sites.

Area	Site	Samples				Altitude (m)	Temperature (°C)	Air sampling rate (XAD samplers; m^3 d^{-1})	Longitude	Latitude
		Lichen		Moss	Soil	Air				
Shergyla	1	√	0.03[a]				2781	8.6		29°39'4.25"N	94°42'52.88"E
	2	√	0.06 [a]				4139	2.8		29°35'24.00"N	94°33'48.00"E
	3	√	0.06 [a]				3777	4.3		29°32'39.12"N	94°33'49.80"E
	4	√	0.14 [a]				3136	7.1		29°49'13.87"N	94°44'20.29"E
	5	√	0.13 [a]				4217	2.4		29°35'57.12"N	94°36'25.60"E
	6	√	0.06 [a]				4205	2.5		29°37'31.76"N	94°42'29.23"E
	7	√	0.11 [a]				3136	7.1		29°49'12.00"N	94°44'12.48"E
	8	√	0.07 [a]				3331	6.2		29°33'51.24"N	94°32'35.46"E
	9	√	0.03 [a]				3446	5.7		29°43'57.68"N	94°43'14.02"E
	10	√	0.08 [a]				3895	3.8		29°39'4.25"N	94°42'52.88"E
	11	√	0.10 [a]				2781	2.2		29°53'41.03"N	94°47'16.69"E
	12	√	0.05 [a]				2781	2.2		29°34'11.50"N	94°34'33.02"E
	13	√	0.12 [a]				3526	5.4		29°34'6.48"N	94°33'55.08"E
Medog	14	√	0.08 [a]	√		√	1989	12	1.88	29°18'21.70"N	95°21'12.61"E
	15	√	0.02 [a]	√	√		3046	7.5		29°43'5.49"N	95°37'46.36"E
	16	√	0.05 [a]	√	√		3708	4.6		29°47'1.69"N	95°41'54.66"E
	17	√	0.07 [a]	√	√	√	1661	13	1.78	29°19'56.55"N	95°21'23.50"E
	18	√	0.09 [a]	√	√		2934	7.9		29°52'13.02"N	95°43'37.37"E
	19	√	0.07 [a]	√	√		3488	5.6		29°48'25.50"N	95°42'4.55"E
	20	√	0.04 [a]	√		√	2707	8.9	2.11	29°42'6.52"N	95°33'58.10"E
	21			√	√	√	2113	12	1.92	29°39'51.12"N	95°29'31.39"E
	22			√	√	√	1754	13	1.81	29°38'18.51"N	95°29'9.43"E
	23				√	√	859	17	1.57	29°19'44.42"N	95°18'9.62"E
	24				√	√	1528	14	1.75	29°29'47.43"N	95°27'36.40"E
	25					√	1182	16	1.65	29°19'31.06"N	95°19'17.89"E
	26					√	3773	4.3	2.54	29°46'1.21"N	95°41'47.75"E
	27					√	3637	4.9	2.48	29°44'32.61"N	95°40'44.77"E
	28				√		1661	13		29°19'56.55"N	95°21'23.50"E

[a] Lipid content in the lichens (%);

Table S2. Homologue distributions of PCNs (%) in all the samples from Tibetan Plateau.

Llichen	**N1**	**N2**	**N3**	**N4**	**N5**	**N6**	**N7**	**N8**	**N9**	**N10**	**N11**	**N12**	**N13**	**N14**	**N15**	**N16**	**N17**	**N18**	**N19**	**N20**
MoCN	5.5	5.1	5.8	5.6	5.9	4.7	6.2	5.7	6.1	10.9	14.8	16.5	23.4	7.1	19.1	26.4	17.6	23.6	22.7	19.2
DiCN	23.1	18.9	19.5	23.7	14.6	15.6	20.5	18.4	11.3	27.9	21.9	21.9	19.2	25.9	24.6	47.1	37.2	44.7	38.0	43.8
TiCN	14.1	14.0	13.8	16.2	11.3	10.8	22.1	16.6	13.0	21.8	31.7	19.8	30.6	18.0	24.0	15.0	20.9	23.4	15.4	20.3
TeCN	16.9	17.0	18.3	17.5	21.4	38.6	25.8	21.4	19.8	15.3	15.5	17.8	13.8	17.7	12.6	5.3	8.9	5.3	7.7	7.3
PeCN	17.7	20.4	22.5	16.7	22.3	13.5	11.0	15.3	20.4	11.7	7.8	10.8	5.0	15.5	8.5	3.4	6.4	1.3	7.8	4.3
HeCN	14.4	17.7	13.7	13.5	16.1	11.1	9.3	15.5	19.2	8.2	5.5	8.3	5.7	10.6	7.5	1.8	5.8	0.9	6.9	3.6
HpCN	5.8	4.5	4.7	4.5	5.6	4.2	3.4	4.4	5.9	2.4	2.1	2.6	1.3	3.7	2.4	0.7	2.0	0.5	1.1	1.0
OCN	2.5	2.5	1.7	2.4	2.8	1.6	1.6	2.6	4.4	1.6	0.8	2.1	1.1	1.5	1.4	0.3	1.1	0.3	0.4	0.6
Moss	**N14**	**N15**	**N16**	**N17**	**N18**	**N19**	**N20**	**N21**	**N22**											
MoCN	23.1	18.7	21.2	23.6	18.5	19.0	31.6	23.1	29.2											
DiCN	45.4	31.1	46.9	47.5	50.4	54.5	34.8	46.4	51.9											
TiCN	23.8	33.8	24.4	23.0	26.5	21.5	22.6	24.1	15.1											
TeCN	5.7	11.9	5.7	4.6	3.6	4.0	7.5	5.1	2.8											
PeCN	0.7	1.4	0.5	0.5	0.5	0.3	1.4	0.5	0.3											
HeCN	0.6	1.7	0.6	0.4	0.2	0.4	0.8	0.4	0.4											
HpCN	0.4	0.9	0.3	0.2	0.1	0.2	0.7	0.3	0.3											
OCN	0.3	0.4	0.3	0.2	0.1	0.2	0.5	0.2	0.1											
Soil	**N15**	**N16**	**N17**	**N18**	**N19**	**N21**	**N22**	**N23**	**N24**	**N28**										
MoCN	42.5	58.9	34.9	47.8	48.1	54.3	31.7	60.2	60.7	57.3										
DiCN	32.6	26.7	38.5	36.0	29.6	30.4	39.0	29.4	28.9	30.3										
TiCN	18.9	9.2	19.2	11.0	14.4	10.7	14.6	8.5	8.5	10.0										
TeCN	4.5	2.5	5.9	3.9	3.9	2.6	6.1	1.0	1.2	1.7										
PeCN	0.6	1.2	0.8	0.7	1.4	1.1	2.3	0.3	0.2	0.3										
HeCN	0.5	0.9	0.4	0.2	1.9	0.6	1.9	0.4	0.3	0.2										
HpCN	0.1	0.6	0.2	0.4	0.6	0.2	2.9	0.1	0.1	0.1										
OCN	0.2	0.1	0.0	0.0	0.1	0.1	1.4	0.1	0.0	0.2										
Air	**N14**	**N17**	**N20**	**N21**	**N22**	**N23**	**N24**	**N25**	**N26**	**N27**										
MoCN	55.3	63.2	23.3	61.5	53.1	44.1	52.1	42.1	12.5	45.2										
DiCN	28.6	24.6	33.1	27.9	33.4	34.6	28.0	36.2	47.9	32.7										
TiCN	10.6	8.5	34.9	7.4	8.1	13.9	12.6	14.2	34.6	16.1										

TeCN	3.7	2.0	6.4	1.7	2.4	4.9	6.1	6.3	4.2	4.2
PeCN	1.2	0.5	1.6	0.3	0.7	1.0	0.7	0.6	0.3	0.8
HeCN	0.4	0.8	0.4	0.7	1.2	1.2	0.2	0.4	0.3	0.6
HpCN	0.2	0.3	0.2	0.3	0.8	0.3	0.1	0.1	0.1	0.3
OCN	0.1	0.1	0.1	0.1	0.4	0.1	0.1	0.1	0.0	0.1

Table S3. Logarithm of the octanol–air partition coefficient (log K_{OA}) values for the PCNs at 25 °C .

CN No.	Congener	log K_{WA} [a]	log K_{OA} [b]	CN No.	Congener	log K_{WA} [a]	log K_{OA} [b]
1	1-CN	2.05	5.81	39	1,2,6,7-tetraCN	2.56	8.35
2	2-CN	2.01	5.76	40	1,2,6,8-tetraCN	3.01	8.47
3	1,2-diCN	2.38	6.76	41	1,2,7,8-tetraCN	3.54	8.71
4	1,3-diCN	2.07		42	1,3,5,7-tetraCN	1.78	7.90
5	1,4-diCN	2.09	7.11	43	1,3,5,8-tetraCN	2.5	8.26
6	1,5-diCN	2.03	6.62	44	1,3,6,7-tetraCN	2.08	8.18
7	1,6-diCN	1.93	7.11	45	1,3,6,8-tetraCN	2.18	8.22
8	1,7-diCN	2.18		46	1,4,5,8-tetraCN	3.29	8.59
9	1,8-diCN	2.83		47	1,4,6,7-tetraCN	2.19	8.18
10	2,3-diCN	2.32		48	2,3,6,7-tetraCN	2.45	8.40
11	2,6-diCN	2.1		49	1,2,3,4,5-pentaCN	3.3	9.28
12	2,7-diCN	1.98	6.62	50	1,2,3,4,6-pentaCN	2.66	8.99
13	1,2,3-triCN	2.57	7.63	51	1,2,3,5,6-pentaCN	2.67	8.98
14	1,2,4-triCN	2.09	7.41	52	1,2,3,5,7-pentaCN	2.3	8.75
15	1,2,5-triCN	2.06	7.45	53	1,2,3,5,8-pentaCN	3.13	9.14
16	1,2,6-triCN	2.09	7.50	54	1,2,3,6,7-pentaCN	2.74	9.10
17	1,2,7-triCN	2.37	7.54	55	1,2,3,6,8-pentaCN	2.96	9.16
18	1,2,8-triCN	3.35		56	1,2,3,7,8-pentaCN	3.73	
19	1,3,5-triCN	1.92	7.32	57	1,2,4,5,6-pentaCN	2.87	9.14
20	1,3,6-triCN	1.68		58	1,2,4,5,7-pentaCN	2.61	8.89
21	1,3,7-triCN	2.01		59	1,2,4,5,8-pentaCN	3.32	9.25
22	1,3,8-triCN	2.63		60	1,2,4,6,7-pentaCN	2.4	8.75
23	1,4,5-triCN	2.71	7.70	61	1,2,4,6,8-pentaCN	2.8	8.94
24	1,4,6-triCN	2.02	7.41	62	1,2,4,7,8-pentaCN	3.09	9.15
25	1,6,7-triCN	2.54	7.54	63	1,2,3,4,5,6-hexaCN	3.71	9.99
26	2,3,6-triCN	2.23	7.54	64	1,2,3,4,5,7-hexaCN	3.31	9.72
27	1,2,3,4-tetraCN	2.72	8.33	65	1,2,3,4,5,8-hexaCN	4.13	10.1
28	1,2,3,5-tetraCN	2.4	8.26	66	1,2,3,4,6,7-hexaCN	3.04	9.66
29	1,2,3,6-tetraCN	2.31		67	1,2,3,5,6,7-hexaCN	3.04	9.66
30	1,2,3,7-tetraCN	2.68	8.33	68	1,2,3,5,6,8-hexaCN	3.36	9.72
31	1,2,3,8-tetraCN	3.45	8.60	69	1,2,3,5,7,8-hexaCN	3.5	9.79
32	1,2,4,5-tetraCN	2.72	8.33	70	1,2,3,6,7,8-hexaCN	3.91	10.1
33	1,2,4,6-tetraCN	2.02	8.04	71	1,2,4,5,6,8-hexaCN	3.6	10.7
34	1,2,4,7-tetraCN	2.08	8.04	72	1,2,4,5,7,8-hexaCN	3.64	10.7
35	1,2,4,8-tetraCN	2.83	8.35	73	1,2,3,4,5,6,7-heptaCN	4.11	10.6
36	1,2,5,6-tetraCN	2.26	8.22	74	1,2,3,4,5,6,8-heptaCN	4.34	10.7
37	1,2,5,7-tetraCN	2.03	8.04	75	OctaCN	5.21	11.5
38	1,2,5,8-tetraCN	2.66	8.47				

[a] reported byTomasz Puzyn and Jerzy Falandysza [2]; [b] reported by Su et al [3];

Calculation of log K_{OA} of PCNs at other temperature was based on the Equation S1.

$$Log\ K_{OA} = \Delta_{OA}U/(\ln 10 * R * T_x) + b \qquad \text{Equation (S8)}$$

$\Delta_{OA}U$ is the internal energy of air-octanol phase transfer in J*mol^{-1}, R is the ideal gas constant, T is the temperature (K), b is the intercept which is a constant. The values of $\Delta_{OA}U$ and b were reported by Su et al.[3]

Table S4. Regression curves of logarithms of the PCN congener concentrations in lichens (pg g^{-1}, normalised to the lipid content (%) of the lichens) with altitude (m).

Congener	Slope	Intercept	R^2	Congener	Slope	Intercept	R^2
2-MoCN	0.00051	-0.03	0.54	12457-PeCN	0.00133	-3.93	0.69
1-MoCN	0.00025	1.16	0.15	12468-PeCN	0.00065	-1.44	0.40
13-DiCN	0.00069	-0.84	0.68	12346-PeCN	0.00091	-1.64	0.74
14/16-DiCN	0.00053	0.31	0.60	12356-PeCN	0.00096	-1.81	0.68
15/27-DiCN	0.00055	-0.39	0.59	12367-PeCN	0.00114	-2.62	0.73
12-DiCN	0.00075	-0.84	0.62	12456-PeCN	0.00100	-2.45	0.54
135-TiCN	0.00058	-0.94	0.56	12478-PeCN	0.00106	-2.66	0.47
146/124-TiCN	0.00041	0.68	0.43	12358-PeCN	0.00113	-3.21	0.63
125-TiCN	0.0005	-0.50	0.69	12368-PeCN	0.00060	-1.55	0.35
126-TiCN	0.00073	-1.50	0.86	12458-PeCN	0.00079	-2.23	0.52
127/167/236-TiCN	0.00063	-0.79	0.70	12345-PeCN	0.00126	-3.70	0.57
123-TiCN	0.00065	-0.56	0.77	123467/123567-HxCN	0.00082	-0.89	0.74
145-TiCN	0.00045	-0.21	0.41	123457/123568-HxCN	0.00074	-0.95	0.71
1357-TeCN	0.00081	-2.18	0.80	123578-HxCN	0.00115	-2.81	0.86
1257/1246/1247-TeCN	0.00071	-0.83	0.60	124568/124578-HxCN	0.00079	-1.87	0.68
1367/1467-TeCN	0.00086	-1.00	0.46	123456-HxCN	0.00121	-2.91	0.55
1368/1256-TeCN	0.00056	-0.17	0.71	123458/123678-HxCN	0.00163	-5.08	0.81
1235/1358-TeCN	0.00056	-0.96	0.58	1234567-HpCN	0.00088	-1.36	0.77
1234/1237-TeCN	0.00075	-0.75	0.81	1234568-HpCN	0.00097	-2.49	0.58
1267-TeCN	0.00048	-0.49	0.15	12345678-OCN	0.00075	-1.07	0.77
1245-TeCN	0.00071	-1.44	0.83				
1248-TeCN	0.00085	-1.90	0.88				
2367-TeCN	0.00056	-1.24	0.70				
1258/1268-TeCN	0.00069	-1.25	0.38				
1458-TeCN	0.00093	-2.35	0.79				
1238-TeCN	0.00074	-1.85	0.79				
1278-TeCN	0.00091	-2.29	0.69				
12357/12467-PeCN	0.00075	-0.89	0.79				

Table S5. Ratios of CN45/36 to CN42, CN54 to CN53/55, CN66/67 to CN71/72, and CN73 to CN74 in samples from Shergyla Mountain, Medog County, and the Bangladesh.

	Soil		Lichen		Moss	Air	Stack gas
	Bangladesh	Medog	Shergyla Mountain	Medog	Medog	Medog	China
CN45/36 to CN42	22.4 ± 15.9	8.66 ± 3.98	14.8 ± 8.49	18.0 ± 11.9	13.5 ± 5.92	14.2 ± 9.93	7.76 ± 9.32
CN54 to CN53/55	1.88 ± 0.68	1.64 ± 2.37	3.89 ± 2.15	3.81 ± 1.46	1.28 ± 1.21	3.98 ± 3.41	2.64 ± 2.32
CN66/67 to CN71/72	9.40 ± 8.66	10.1 ± 8.90	14.0 ± 9.85	36.1 ± 30.4	11.3 ± 10.4	2.51 ± 2.92	21.9 ± 38.5
CN73 to CN74	3.95 ± 1.10	9.48 ± 7.61	6.14 ± 3.90	4.74 ± 1.16	7.53 ± 2.46	6.00 ± 1.43	4.10 ± 6.91

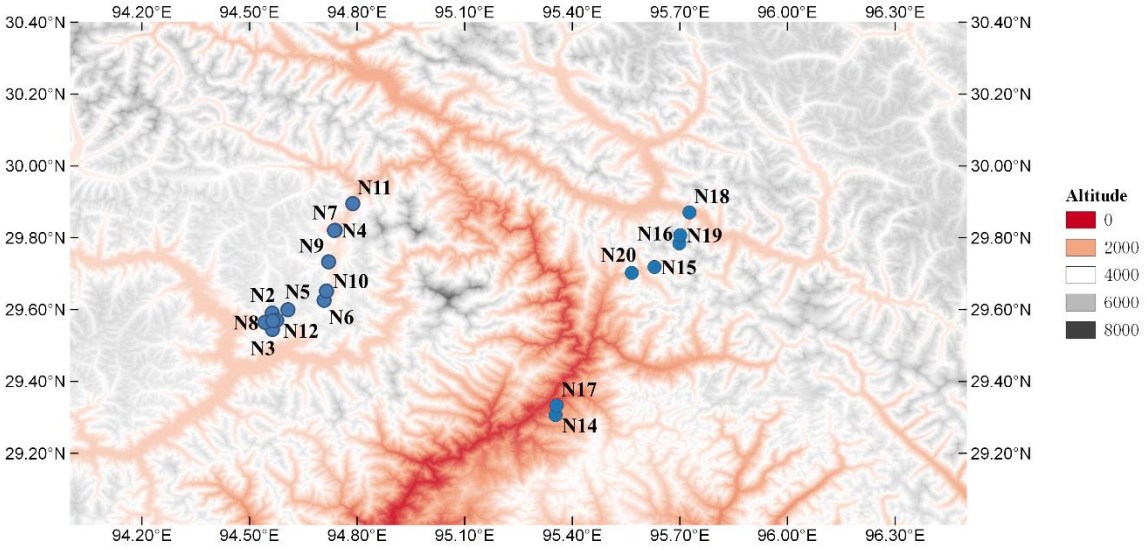

Figure S1. Sampling sites of lichens in Tibetan Plateau.

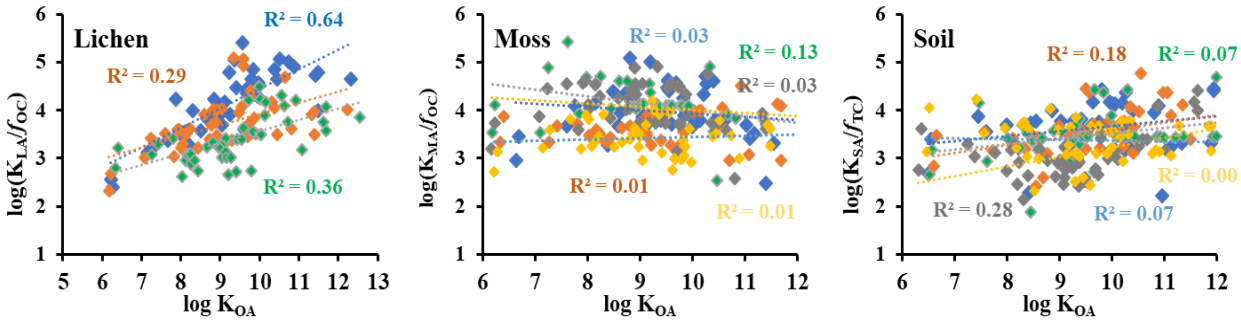

Figure 2. Correlations between log K_{OA} and log $K_{LA}/K_{MA}/K_{SA}$ for each paired sampling sites.

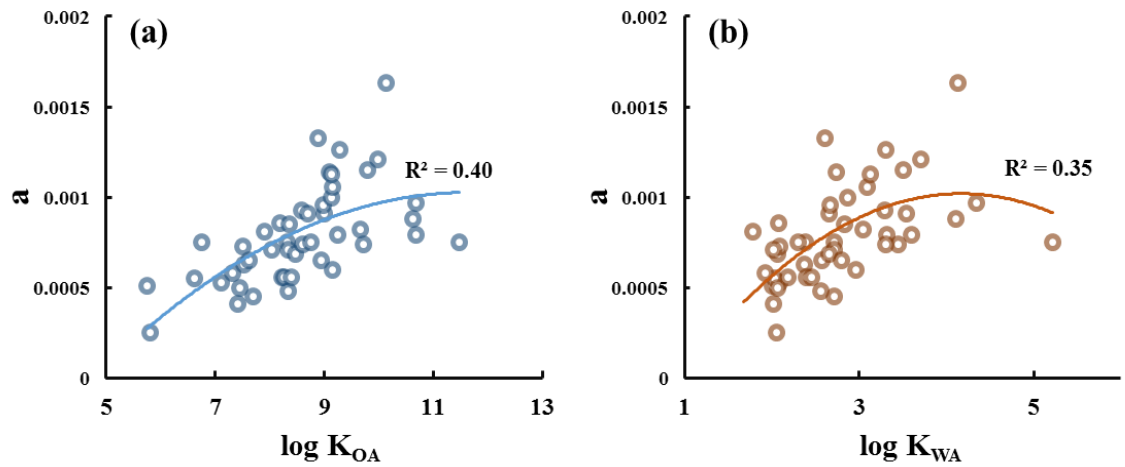

Figure S3. Relationship between log K_{OA} (a) or log K_{WA} (b) and the slope.

*a is the slope of regression curves of the log (PCN congener concentrations normalized to lipid content) to altitudes. Values were shown in Table S2 (Slop).

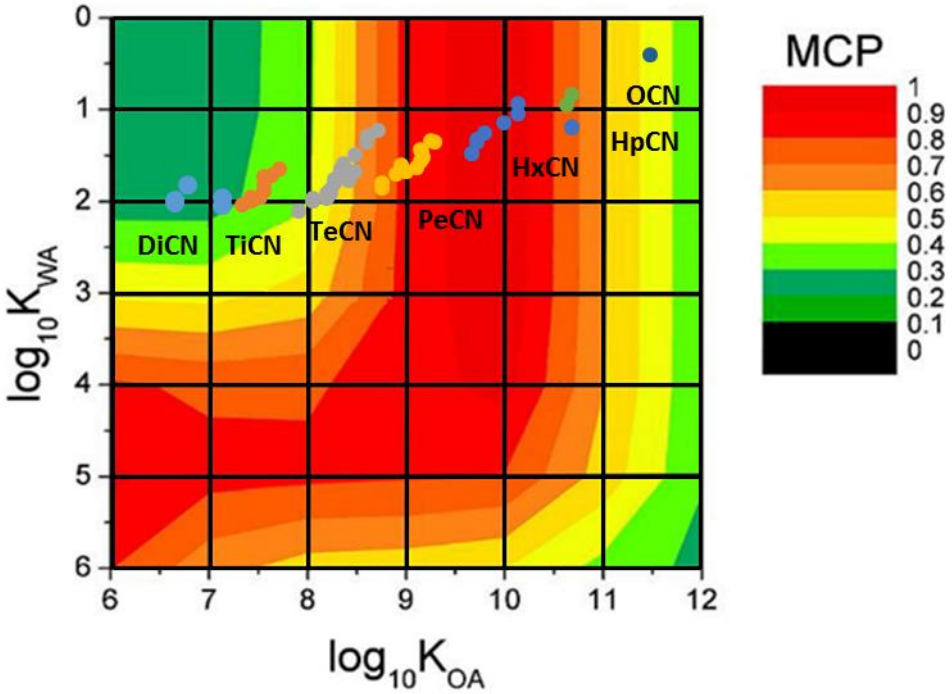

Figure S4. Application of log K_{OA} and log K_{WA} of the PCN homologues to the mountain contamination potential model developed by Wania et al.[1]

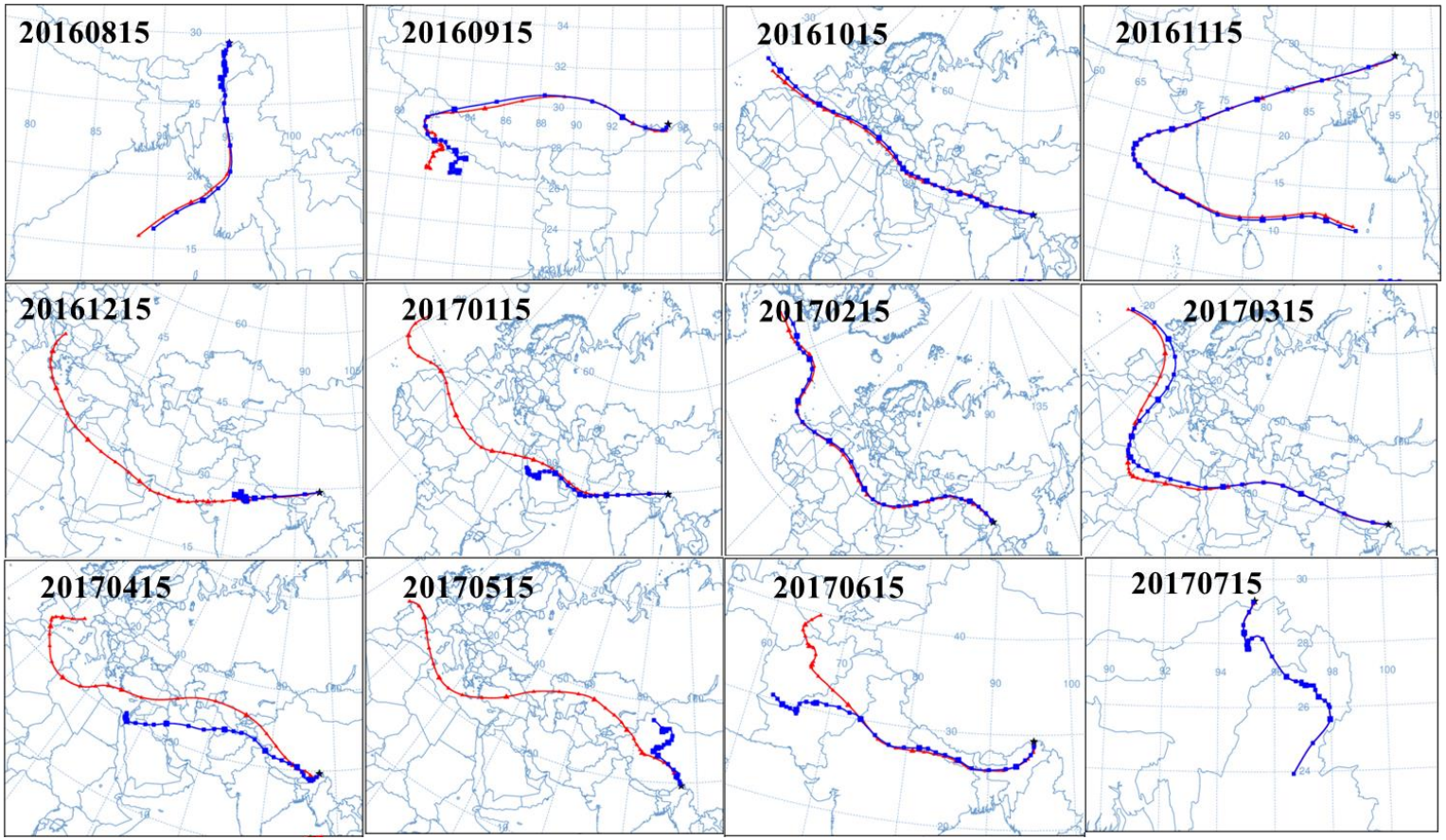

Figure S5. Hybrid Single Particle Lagrangian Integrated Trajectory Model results based on back trajectories from August 2016 to July 2017 in Medog County in Tibetan Plateau.

Reference

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2. Puzyn, T.; Falandysz, J., QSPR modeling of partition coefficients and henry's law constants for 75 chloronaphthalene congeners by means of six chemometric approaches—a comparative study. *J. Phys. Chem. Ref. Data* **2007,** *36*, (1), 203-214.
3. Su, Y. S.; Lei, Y. D.; Daly, G. L.; Wania, F., Determination of octanol-air partition coefficient (K-OA) values for chlorobenzenes and polychlorinated naphthalenes from gas chromatographic retention times. *J. Chem. Eng. Data* **2002,** *47*, (3), 449-455.