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Compositions of Upper Mantle Fluids Beneath Eastern China: Implications for Mantle Evolution

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Abstract The composition of gases trapped in olivine, orthopyroxene and clinopyroxene in lherzolite xenoliths collected from different locations in eastern China has been measured by the vacuum stepped-heating mass spectrometry. These xenoliths are hosted in alkali basalts and considered as residues of partial melting of the upper mantle, and may contain evidence of mantle evolution. The results show that various kinds of fluid inclusions in lherzolite xenoliths have been released at distinct times, which could be related to different stages of mantle evolution. In general, primitive fluids of the upper mantle (PFUM) beneath eastern China are dominated by H_2, CO_2 and CO, and are characterized by high contents of H_2 and reduced gases. The compositions of PFUM are highly variable and related to tectonic settings. CO, CO_2 and H_2 are the main components of the PFUM beneath cratons; the PFUM in the mantle enriched in potassic metasomatism in the northern part of northeastern China has a high content of H_2, while CO_2 and SO_2 are the dominant components of the PFUM in the Su-Lu-Wan (Jiangsu-Shandong-Anhui) region, where recycled crustal fluids were mixed with deeper mantle components. There are several fluids with distinct compositions beneath eastern China, such as primitive fluids of upper mantle (CO, CO_2 and H_2), partial melting fluids (CO_2 and CO) and metasomatic fluids mixed with recycled crustal fluids (CO_2, N_2, SO_2 and CH_4) etc. Fluids of the upper mantle beneath the North China craton are different from that of the South China craton in total gases and chemical compositions: the contents of the reduced gases of the PFUM in the NCC are higher than those in the SCC.

Key words: chemical composition, mantle fluids, lherzolite xenoliths, eastern China

1 Introduction

Mantle fluids are the most active components in the processes of mantle evolution and energy exchange in earth's interior, and they play a critical role in the processes of partial melting, mantle plume formation and mantle metasomatism. Mantle-derived ultramafic xenoliths contain abundant fluids, and the fluids in lherzolite xenoliths representing trapped samples of fluid phases present in the upper mantle are very important for understanding of the mantle evolution (Fan et al., 1989; Xia et al., 1990; Rosenbaum et al., 1996; Andersen et al., 2001). The lherzolite xenoliths hosted in alkali basalts in eastern China are considered to be residues of the upper mantle after partial melting. The compositions of fluids from different kind of minerals in lherzolite underwent distinct affects in various mantle events, which are inextricably linked to the mantle evolution and tectonic setting, can provide a direct approach to understanding the mantle evolution in eastern China (Chi, 1988; Wood et al., 1990; Fan and Yang, 1993; Du et al., 1996; Zhang et al., 1999a). In the present study, the fluid compositions within olivine, orthopyroxene and clinopyroxene in lherzolite xenoliths of alkali-basalts in eastern China are measured by on-line connection between a stepped-heating apparatus and a mass spectrometer, and the compositional characters of the mantle fluid associated with the tectonic setting are discussed.

2 Samples and Experiments

The investigated samples are fresh lherzolite xenoliths collected from Dixiasenlin (DS), Jiaohe (JH), Huinan (HN), Damaping (DM), Zhangjiakou (ZJ), Dafangshan (DF), Liuhefangshan (FS), Mingxi (MX), Dayangcao (DY) and Tazhuang (TZ) in eastern China. The fresh bulk samples of lherzolite xenoliths were crushed to a grain size of 20-80 mesh, and olivine, orthopyroxene and clinopyroxene mineral separates were carefully hand-picked under a binocular microscope. Samples were dipped into 3M HCl and CH_2Cl_2 to remove secondary carbonate and surficial organic matter respectively, followed by ultrasonic cleaning with distilled water, and drying in an oven at °C.

The stepped-heating, gas extraction, mass-spectrometry method for analysis of fluids has been widely used for analysis of mantle xenoliths (Nadeau et al, 1990; Zhang et al, 1999a, b, 2003), and was employed to analyze the fluid compositions of different minerals from lherzolite xenoliths in eastern China. The gas extracting apparatus is composed of a quartz tube, a cold trap, and a valve directly connected to the MAT-271 mass spectrometer. The cleaned sample is loaded in the quartz tube and heated for 1 hour under high-purity O_2 at 200°C to oxidize the surficial organic matter, the adsorbed gases on the mineral surface are pumped away for 4 hours at a pressure of 1×10^{-3} Pa. The mass spectrometer is then isolated from the gas extraction line by closing the valve, and the quartz tube is heated from 200°C to 1200°C with a increment of 100°C. Each temperature increment is held for 1 hour, and the released gases are condensed in the cool trap with liquid nitrogen. The valve is then opened to the mass spectrometer and the liquid nitrogen is replaced with an alcohol-liquid nitrogen mixture to release all of the condensed gases except for H_2O. The gas compositions are measured by the mass spectrometer (Finnigan MAT 271), and the standard deviation for this method was reported by Liu et al. (1997).

3 Gases Compositions of Different Minerals from Lherzolite Xenoliths

3.1 The fluid-degassing features of lherzolite xenoliths in eastern China and their significances

The fluids in olivine, orthopyroxene and clinopyroxene from lherzolite xenoliths have been released stage by stage and formed two releasing peaks at about 400°C (named the low temperature peaks hereafter) and 800°C (named the high temperature peaks hereafter) in vacuum stepped-heating experiments. As shown in Fig. 1, the total amounts of the gases released with the temperature increase in olivine (olv), orthopyroxene (opx) and clinopyroxene (cpx) of lherzolite xenoliths in DM, eastern China have two main peaks at about 400°C and 800°C. Because efficient sample pretreatments were employed to remove the gases adsorbed

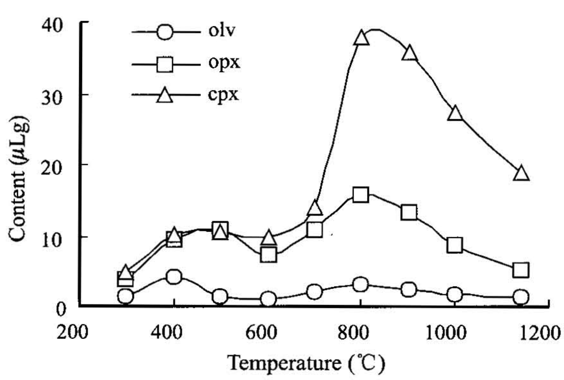

Fig. 1. Amounts of gases released at different heating temperatures in olivine (olv), orthopyroxene (opx) and clinopyroxene (cpx) from lherzolite xenoliths in Damaping, eastern China.

on sample surfaces and filled in cracks, and the vacuum cooling method was adopted in the experiment to rapidly trap such gases as H_2O and CO_2, and high-weight hydrocarbons released from the sample and to reduce the possibility for new species (H_2 and CO etc.) to be formed as they were reacted with other gases in the high temperature area, the gases released from the sample should be derived from different fluid inclusions (Nadeau et al, 1990; Zhang et al., 2003).

Fluid inclusions in mantle xenoliths are formed by decompression exsolution of gaseous components dissolved in pyrolite at high temperature and high pressure during mantle uplift, trapping-pressures that form fluid inclusions are so high that the bursting temperature of the fluid inclusion is lower than its homogenization temperature (Du et al, 1996; Zhang et al., 1999b; Andersen, et al, 2001). There are three kinds of fluid inclusions in lherzolite xenoliths in eastern China with significant differences in size and shape. The first type refers to the late-stage fluid inclusions, which have irregular shapes like tree branches or bead strings, ranging from 0.01 to 0.1 mm in size, and have micro-cracks occurring at the edges of the inclusions, suggesting that they are in poorly closed systems. The second type, the early-stage fluid inclusions have regular shapes like balls, eggs or negative crystals with the sizes ranging from 0.005 to 0.02 mm. These inclusions are in closed systems. The third type is ultra-microscopic fluid inclusions with the sizes less than 0.001 mm, which are very abundant in lherzolite xenoliths in eastern China. The formation temperatures of the fluid inclusions in lherzolite xenoliths from eastern China range from 987 to 1296°C, and the homogenization temperatures are from 861 to 1074°C (Liu et al, 1981; Xia, 1984,1990; Fan et al., 1989; Peng et al., 1994). Consequently, gases that make up high-temperature peaks that are released at higher temperatures of 700–1200°C (close to the homogenization temperature range) are likely formed by the bursting of the early-stage fluid inclusions and some ultra-microscopic fluid inclusions. However, gases that comprise low-temperature peaks occurring at lower temperatures from 300 to 600°C were possibly derived from the bursting of the late-stage fluid inclusions and some early-stage fluid inclusions occurring at the sample surfaces. This has been demonstrated by the similarity of the fluid compositions to those of single fluid inclusions obtained by using the Laser Raman probe (Xia, 1984). The total amounts of gases comprising low-temperature peaks are lower than those of the corresponding high-temperature peaks, possibly indicating great leakage that the late-stage fluid inclusions had suffered. Based on the measurement results of the stepped-heating mass spectrometer, fluid compositions of two released peaks in lherzolites in

Table 1 Volatile compositions of olive (olv), orthopyroxene (opx), clinopyroxene (cpx) and whole-rock (wr) of lherzolite xenoliths beneath eastern China (μL/g)$_{STP}$

No		Low temperature peaks (300–600°C)							High temperature peaks (700–1200°C)						
		H_2	CH_4	H_2S	CO	N_2	CO_2	SO_2	H_2	CH_4	H_2S	CO	N_2	CO_2	SO_2
JH01	olv	0.28	0.13	0.00	0.49	0.04	3.83	0.46	0.75	0.08	0.00	0.16	0.18	3.98	0.10
HN01		0.07	0.09	0.00	0.72	0.05	3.92	0.07	1.89	0.15	0.00	1.42	0.11	1.52	0.00
MP01		0.24	0.16	0.01	2.77	0.01	4.91	0.01	0.88	0.02	0.01	8.92	0.07	0.79	0.00
ZJ01		1.19	0.26	0.00	1.88	0.95	4.95	0.00	8.96	0.26	0.00	12.23	1.34	4.58	0.00
DF01		0.45	0.38	0.01	2.79	0.00	3.12	0.00	1.33	0.09	0.02	16.13	0.34	4.31	0.00
MX01		0.04	0.12	0.00	0.47	0.11	1.45	0.35	0.06	0.08	0.00	1.07	0.11	2.09	0.22
DY01		0.28	0.19	0.00	0.75	0.06	5.37	1.16	0.18	0.09	0.00	0.51	0.18	7.68	2.25
TZ01		1.16	0.68	0.00	2.18	0.06	9.25	0.02	10.73	0.39	0.00	13.18	0.25	12.70	0.00
JH02	opx	4.03	0.70	0.00	2.55	0.09	5.71	0.00	79.52	0.39	0.01	8.95	0.49	7.79	0.00
HN02		0.18	0.30	0.02	2.51	0.22	6.09	0.42	90.52	0.75	0.73	21.72	0.62	1.94	0.00
MP02		0.30	0.53	0.02	11.62	0.09	19.05	0.06	6.99	0.32	0.11	33.40	1.97	11.10	0.00
ZJ02		0.76	1.01	0.03	3.86	0.08	11.39	1.73	21.57	0.78	0.40	11.27	0.38	18.12	0.05
DF02		3.55	0.18	0.02	4.07	0.17	1.82	0.00	88.35	0.37	0.94	39.27	0.36	4.91	0.41
MX02		1.51	0.36	0.00	2.71	0.12	4.40	0.23	145.10	0.49	0.02	17.87	0.54	1.53	0.00
DY02		0.70	0.41	0.00	1.34	0.17	2.58	0.00	107.79	0.46	0.00	11.66	0.31	5.60	0.00
TZ02		1.54	0.73	0.00	4.33	0.00	16.20	0.06	99.91	0.88	0.05	22.01	0.00	18.16	0.01
JH03	cpx	0.27	8.65	0.13	2.16	1.95	8.79	0.00	0.45	12.37	0.00	130.26	117.35	174.65	0.00
HN03		0.12	6.25	0.12	0.40	1.91	7.32	0.00	2.13	2.56	0.00	28.04	133.52	201.96	0.00
MP03		0.27	15.82	0.01	2.79	3.19	23.52	0.00	2.28	8.21	0.09	27.01	31.13	116.02	0.00
ZJ03		0.19	12.32	0.24	2.14	2.09	6.76	0.00	0.54	11.10	0.00	43.72	42.87	107.57	0.00
MX03		0.09	3.65	0.52	0.19	0.28	2.80	0.00	0.63	1.87	0.00	57.34	84.32	164.55	0.00
DY03		1.96	2.38	0.00	7.92	1.14	13.98	0.00	176.90	2.03	0.02	86.33	4.29	15.23	0.00
TZ03		1.95	0.70	0.00	6.25	0.18	20.87	0.00	155.56	0.86	0.10	40.68	0.28	25.52	0.00
DS20	wr	1.53	0.25	0.00	1.65	0.18	4.50	0.00	18.73	0.20	0.00	4.60	0.38	6.26	0.18
FS20		0.17	0.49	0.00	1.80	0.12	17.54	0.02	0.12	0.03	0.00	0.00	1.01	24.66	2.61

different localities have been calculated and listed in Table 1.

3.2 Fluid compositions of minerals in lherzolite xenoliths

Fluid compositions in different minerals from lherzolite xenoliths are significantly variable (Table 1). The total amount of fluid in olivine is the lowest among the minerals from lherzolite, averaging 22.54 (μL/g)$_{STP}$; fluids for high-temperature peaks are composed mainly of CO_2, CO and H_2, whose contents are 37.4%, 37.5% and 17.05% respectively, and secondarily of SO_2 and CH_4, and the total amount of gases averages 15.31(μL/g)$_{STP}$; for low-temperature peaks, CO_2 and CO with contents of 63.26% and 21.15% respectively are the major components, and H_2, SO_2, CH_4 and N_2 are the minor ones. The total amount of fluid in clinopyroxene is the highest among the three minerals with an average of 355.54 (μL/g)$_{STP}$. CO_2 is the main gas for high-temperature peaks with a content of 35.19%, and the others are N_2, CO, SO_2 and H_2 with contents of 16.99%, 16.82%, 16.17% and 13.01% respectively. For low-temperature peaks, CO_2 and CH_4 are the major constituents with contents of 47.61% and 29.02% respectively, and CO, N_2 and SO_2 are the minor components. Although the total amount of fluid in orthopyroxene is lower than that in clinopyroxene, with an average value of 125.93 (μL/g)$_{STP}$, H_2 is the dominant component for high-temperature peaks with concentrations as high as 72.13%, and CO_2 and CO are the secondary components with contents of 18.73% and 7.80% respectively. For low-temperature peaks, CO_2 is the main component with the content as high as 55.77%, and the contents of CO and H_2 are 27.37% and 10.42% respectively. The content of SO_2 both in orthopyroxene and olivine is higher for the low-temperature peaks than for the high-temperature peaks.

4 Fluid Composition Features of the Upper Mantle beneath Eastern China

4.1 Evolution features of the upper-mantle fluids beneath eastern China

Primitive fluids of the upper mantle (PFUM) were sealed in the minerals during crystallization of pyrolite, and their original characters have been affected by the allochthonous fluids in the processes of upper-mantle partial melting and metasomatism. Only the early-stage fluid inclusions in the interior of such refractory minerals as olivine have kept intact and are representative of the original compositions of the PFUM. Fluids in other minerals from lherzolite may retain information pertaining to the evolution of Earth's mantle. Based on the fluid compositions in olivine, orthopyroxene and clinopyroxene of lherzolite in eastern China, some different kinds of mantle fluids can be distinguished as follows.

(1) PFUM, referring to fluids at a primary stage of the upper-mantle crystallization, are characterized by gases of high-temperature peaks in olivine, which were derived from the bursting of the closed early-stage fluid inclusions and ultra-microscopic fluid inclusions. The PFUM beneath eastern China are characterized by high concentrations of H_2, CO_2 and CO and SO_2, and H_2S, N_2 and CH_4 are the minor constituents. Relevant studies show that the contents of CO_2 and H_2O in the whole rock of lherzolite are higher than those of olive, which implies that CO_2 and H_2O are not mainly preserved in olive (Du et al., 1996); CO_2-related LREE-rich fluids are preserved mainly in mineral interstices (Fan et al., 1993; Xu, 2000), suggesting that the concentrations of H_2O and CO_2 in the PFUM are not high.

(2) Late-stage fluids of the upper mantle, referring to fluids in the late stage of the upper-mantle crystallization, are characterized by gases released from orthopyroxene for high-temperature peaks since orthopyroxene has a lower crystallization temperature than olivine but a higher temperature than clinopyroxene. The gas composition is predominantly H_2 with relatively small amounts of CO_2 and CO (Fig. 2, opx$^+$). Figure 2 shows the PFUM-normalized patterns of the fluid compositions in different minerals from lherzolite in the North China craton (NCC) and the South China craton (SCC). The high content of H_2, which has been proved little derived from OH^- in the orthopyroxene structure (Bell et al., 1995; Skogby et al., 1989, Zhang et al., 2003), is related to the high level of H_2 enrichment during the late-stages upper-mantle crystallization.

(3) Partial-melting fluids of the upper mantle refer to the fluids related to partial melting of the upper mantle. Partial melting of the upper mantle occurred in various degrees beneath eastern China, resulting in the formation of melt inclusions and late-stage fluid inclusions in the interior of minerals and the growth of spongy partial-melt belts on mineral marginal rims and in cracks (Chi, 1988; Xia, 1990; Peng et al, 1994). The fluids of the low temperature peaks in olivine and orthopyroxene show similar compositions (Fig.2, olv$^-$, opx$^-$), which are derived from the late-stage fluid inclusions and considered as the fluids related to the partial melting of the upper mantle. CO_2 is the main component, while CO, H_2, SO_2, CH_4 and N_2 are the minor components. Higher contents of CO_2 and SO_2 indicate that partial melting of the upper mantle may have occurred under oxidizing conditions.

(4) Metasomatic fluids mixed with recycled crustal fluids refer to those that have been involved due to the the mixing of mantle fluids and recycled crustal fluids. Extensive and strong metasomatism occurring in eastern China has affected the fluids in clinopyroxene, an easily melting mineral, so fluid compositions in clinopyroxene are different from those in paragenetic olivine and orthopyroxene as well as the host alkali basalts (Zhang et al, 1999b). The fluid of the high-temperature peak in clinopyroxene is composed mainly of CO_2 (Fig. 2, cpx$^+$); while that of the low temperature peak, mainly of CO_2 and CH_4 (Fig 2, cpx$^-$). N_2 and CH_4 are enriched compared to the PFUM, and CO_2 is likely related to LREE-rich metasomatic fluids, which are enriched in N_2, SO_2, CO_2 and CH_4 as a result of the contribution of recycled crustal fluids (Peacock, 1990; Wood et al., 1990; Tatsumoto et al., 1992; Alt et al., 1993; Fan et al., 1993).

4.2 Compositional features of the upper-mantle fluids beneath eastern China

Differences in tectonic and geochemical geneses of the

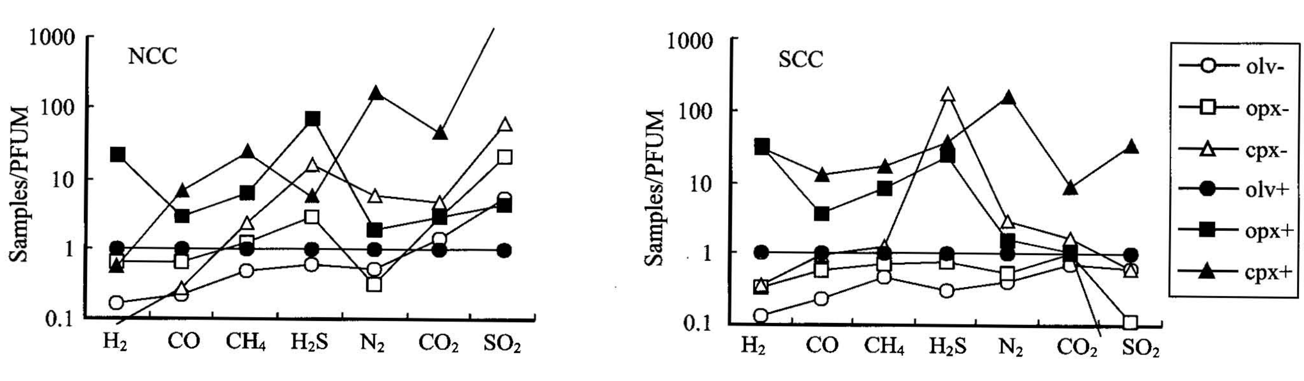

Fig. 2. PFUM-normalized patterns for the fluid compositions of different minerals in lherzolite from the NCC and SCC.
PFUM is based on fluid compositions of high temperature peaks in olivine (olv+).

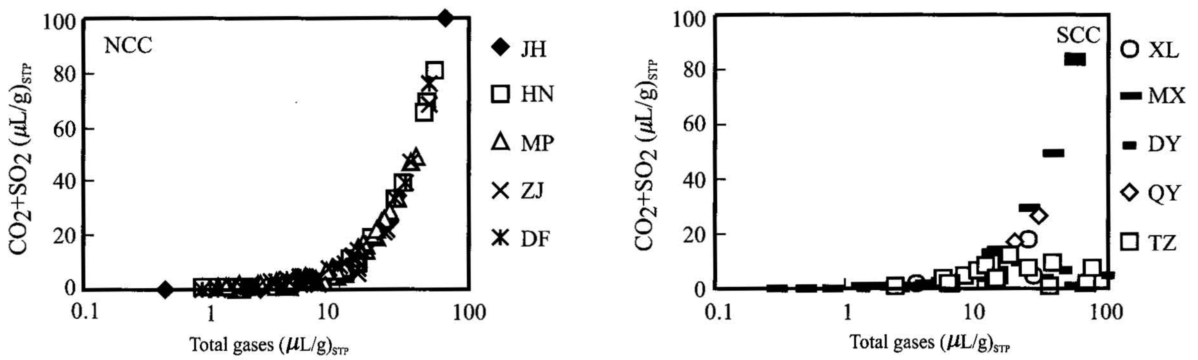

Fig. 3. Relationships between the total gases and CO_2+SO_2 from lherzolite in the NCC and SCC.

upper mantle between the NCC and the SCC result in different fluid compositions observed in lherzolite xenoliths (Chi, 1988; Du et al., 1996; Xu et al., 1996; Zheng et al, 1999; Zhi et al., 2001). The total amount of gases in the SCC is slightly higher than that of the NCC, and the relationships between the total amounts of oxidizing gases such as CO_2+SO_2 also show some differences between the NCC and the SCC (Fig. 3). In eastern China, the compositions of mantle sources are different in different tectonic settings and the compositions of the PFUM are also varied with different mantle reservoirs in various tectonic settings (Fan et al., 1989; Tatsumoto et al., 1992; Zhang et al., 1999a). The PFUM compositions in different tectonic settings in eastern China are summarized in Table 2. The PFUM beneath cratons are composed mainly of CO, CO_2 and H_2. The PFUM in the NCC associated with a primitive mantle-like reservoir is composed mainly of CO and secondary of CO_2 and H_2. The PFUM beneath the SCC, which is related to a depleted mantle reservoir, is dominated by CO_2 with minor amounts of CO and H_2. The total amount of reduced gases of the PFUM in the NCC (75.46%) is obviously higher than that of the SCC (50.80%). The PFUM in the northern part of northeastern China with enriched mantle of potassic metasomatism is characterized by the highest total gases and obviously enriched in H_2 compared with the PFUM in the NCC, showing that metasomatic fluids with a high content of H_2 exist in the upper mantle. The PFUM of the Jiangsu-Shandong-Anhui region with a mixed mantle reservoir is composed primarily of CO_2, and secondly of SO_2, suggesting that recycled crustal metasomatic fluids could be involved (Alt et al., 1993; Peacock, 1990; Fan et al., 1993). Partial-melting fluids in the upper mantle in the NCC are rich in CO_2 and SO_2, while those in the SCC rich in CO_2 compared with the PFUM. Metasomatic fluids mixed by recycled crustal fluids in the NCC are dominated by CO_2, and those in the SCC are composed mainly of H_2, CO_2 and CO.

Table 2 Compositions of PFUM in different settings beneath eastern China

District	Type of mantle	H_2	CH_4	H_2S	CO	N_2	CO_2	SO_2	Total
NCC	Primitive Mantle	2.76	0.12	0.01	7.77	0.41	3.04	0.02	14.13
SCC	Depleted Mantle	3.66	0.19	0.00	4.92	0.18	7.49	0.82	17.26
NPNC	Enriched Mantle	18.73	0.20	0.00	4.60	0.38	6.26	0.18	30.35
SLW	Mixed Mantle	0.12	0.03	0.00	0.00	1.01	24.66	2.61	28.43

Note: NCC – Northern China craton; SCC – South China craton; NPNC – northern part of northeastern China; SLW – Jiangsu-Shandong-Anhui region. Data of NPNC and SLW are calculated based on fluid compositions of lherzolite.

5 Conclusions

(1) The total amount of gases released with increasing temperature in lherzolite xenoliths have been identified, which formed two main peaks at about 400°C and 800°C and different minerals have obvious different fluid compositions and total gases.

(2) The primitive fluids of the upper mantle beneath eastern China are dominated by H_2, CO_2 and CO, and characterized by high contents of H_2 and reduced gases. The compositions show a close relation to the tectonic setting and the fluids beneath cratons are composed mainly of CO, CO_2 and H_2.

(3) Four types of fluids in lherzolite xenoliths have been identified, i.e. primitive fluids of the upper mantle, late-stage fluids of the upper mantle, partial-melting fluids and metasomatic fluids mixed with recycled crustal fluids.

(4) This is the first published report relating distinct differences in compositions and total gas contents of the fluids in the upper mantle between the NCC and the SCC.

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