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Sulfur-containing particles emitted by concealed sulfide ore deposits: an unknown source of sulfur-containing particles in the atmosphere

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Supplementary Information

For 16 ore deposits, in which we have studied particles carried by ascending geogas, a large number of sulfur-containing and Pb- and As-containing particles were found. There are oxidative ore bodies in many concealed sulfide ore deposits. As sulfide minerals change into oxide minerals, sulfide was released from these minerals. There are some sulfide concentration data for ascending geogas. Yuan et al. (China University of Geosciences, Beijing, China, 2014) analyzed sulfide concentrations of ascending geogas in soil at the Sunit deposit (the Inner Mongolia Autonomous Region, China), using plasma mass spectrographic analysis. Their sampling method allowed the flow of geogas in the soil through liquid collector slowly using a pump. The particles carried by the ascending geogas flow were adsorbed in the liquid collector. The volume of the geogas extracted per hole was 5 liters. The geogas extracted from 3 holes (15 liters) was combined to make one sample. The liquid collector was made with high purity nitric acid and Mini-Q ultra pure water. The liquid collector was placed in a 25 ml polyethylene bottle. The analysis results from 1054 samples showed that the average sulfur content of the liquid collector was 26.4571 μg ml^{-1}. The maximum value was 35.33 μg ml^{-1} and the minimum value was 16.89 μg ml^{-1}. A concentration of 26.4571 μg ml^{-1} in the liquid collector may be translated into 44.095 mg per cubic meter of geogas flow. We know that sulfur-containing substances carried by geogas flow may be not completely adsorbed in the liquid collector. Therefore, the average sulfur content of the ascending geogas flow may have been higher than 44.095 mg per cubic meter. We analyzed the sulfide concentration of

ascending geogas in the soil at the Kangjiawan deposit in the Hunan Province, China. Our sampling method is similarly to the method used by Yuan et al. (2014). The main difference is that our liquid collector was made with high purity aqua regia and tri-distilled water. The volume of the liquid collector was 100 ml. The volume of the geogas extracted from a hole was 9 liters. Therefore, the volume of the geogas extracted from 3 holes was 27 liters. The sulfide concentration of the liquid collector was analyzed using the plasma spectrum method. We analyzed the samples along 3 sections (sample numbers were 31, 74, and 20). The results showed that the average sulfur contents of the 3 sections were 0.27, 1.40, and 32.81 μg ml^{-1} respectively (Tables S1–3), which may be translated into 1.00, 5.19, and 121.50 mg per cubic meter of geogas flow, respectively. Wang et al. (2008) collected particles carried by ascending geogas in soil over the Jiaolongzhang Pb-Zn-Cu-Ag deposit, Eastern Gansu, China using a liquid collector. Analysis results from 14 samples showed that the mean content of Cu, Pb, and Zn was 844 ng/ml (gas volume), 107 ng/ml, and 1751 ng/ml, respectively.

There is earth degassing phenomena in metallic and nonmetallic deposits. The giant gold deposits, such as the Porcupine gold deposit in Canada, the Witwatersrand gold deposit in South Africa, and the Muruntau gold deposit in Uzbekistan, exhibit upward vertical movement of hydrocarbon gas. The Witwatersrand gold deposit has significant upward gas flow. In one day, 36700 m^3 of hydrocarbon gases degas from underground gold mining vents and 5×10^8 m^3 of hydrocarbon gases degas from 3000m or deeper mines every year. The Azerbaijan oil and gas region is strongly

degassed, with 4×10^8 m^3 of gases degassed every year (Du, 2009). The ascending gas flow rates were measured to be between 60×10^{-4} and 4 cm^3 min^{-1} m^{-2} horizontally projected borehole area at three different sites by Malmqvist & Kristiansson (1984). Carbon dioxide concentrations above sulfide mineralizations are often enhanced. Hidden sulfide mineralizations at a depth of 200 m have been located in quartzite in areas such as Brittany, and sulphide ores have been located in granite in Cornwall. Above mineralizations, carbon dioxide in the soil gas has been found to increase to 10% from the normal concentration of 1%. The carbon dioxide flow may be as large as 0.2 l m^{-2} h^{-1} (Hermansson et al. 1991). The Dongshengmiao deposit lies in a seismically active zone. The Langshan Mountain-front fault, in which minor earthquake activity frequently takes place and where M=6 earthquakes have taken place three times in the twentieth century, passes through the deposit. The release of geogas in active tectonic areas is widespread and occurs at a significant level (Judd et al., 1997; Etiope, 1999; Mörner and Etiope, 2002). The CO_2 emission flux of the Siena Graben Faults (Italy), Siena G. Arbia Fault (Italy), Ustica Arso Fault (Italy), and San Andreas Fault (California) were 0.83–1123, 12.4–74.4, 77.3, and 0.4–23 kg m^{-2} year^{-1} respectively (Etiope, 1995; 1999; Mörner and Etiope, 2002; Lewicki and Brantley, 2000). These equate, respectively, to 0.02–26.94, 0.3–1.78, 1.85, and 0.01–0.55 cm^3 m^{-2} s^{-1} if CO_2 density is assumed to be 1.3401 kg m^{-3}. The area of the Dongshengmiao deposit is 4.65 km^2. The emission flux estimation of the Dongshengmiao deposit was 0.5 cm^3 m^{-2} s^{-1} according to the emission fluxes of the above-mentioned faults and deposits. Therefore, the estimated degassing rate for the

Dongshengmiao deposit was 2.325 $m^3 s^{-1}$.

The distribution areas of concealed sulfur ore deposits are different. The ore deposits with the distribution areas of 1–12 km^2 may have more deposits than other areas. Concealed metal deposits containing sulfide minerals can be very extensive, such as the Killik massive sulfide deposit in northeastern Turkey (Çiftçi et al., 2005), the Masa Valverde blind massive sulfide deposit in Spain (Ruiz et al., 2002), and the Huize carbonate-hosted Zn–Pb–(Ag) District in South China (Han et al., 2007). Concealed sulfur nonmetallic deposits, such as gypsum and barite, are also widely distributed. The number of concealed sulfide deposits is far greater than those of active volcanoes. Under the climate-warming conditions, oxidation of sulfur-containing minerals is particularly accelerated.

Table S1 Plasma spectrum S results for liquid collectors along the 1st section (μg/mL)

Number	S	Number	S	Number	S	Number	S
K1-1	0.22	K1-9	0.08	K1-17	0.12	K1-25	0.43
K1-2	0.20	K1-10	0.18	K1-18	0.13	K1-26	0.33
K1-3	0.13	K1-11	0.15	K1-19	0.26	K1-27	0.83
K1-4	0.12	K1-12	0.12	K1-20	0.27	K1-28	0.15
K1-5	0.12	K1-13	0.75	K1-21	0.68	K1-29	0.48
K1-6	0.12	K1-14	0.13	K1-22	0.37	K1-30	0.09
K1-7	0.35	K1-15	0.14	K1-23	0.91	K1-31	0.09
K1-8	0.13	K1-16	0.20	K1-24	0.11		

Table S2 Plasma spectrum S results for liquid collectors along the 2nd section (μg/mL)

Number	S	Number	S	Number	S	Number	S
K2-1	1.74	K2-20	3.81	K2-39	0.6	K2-58	0.31
K2-2	1.21	K2-21	1.52	K2-40	0.9	K2-59	0.58
K2-3	1.46	K2-22	4.44	K2-41	1.08	K2-60	0.42
K2-4	0.27	K2-23	0.72	K2-42	0.26	K2-61	0.59
K2-5	1.68	K2-24	1.07	K2-43	2.03	K2-62	3.86
K2-6	0.97	K2-25	0.57	K2-44	1.05	K2-63	0.51
K2-7	0.31	K2-26	0.43	K2-45	0.48	K2-64	0.57
K2-8	1.35	K2-27	0.61	K2-46	2.46	K2-65	0.2
K2-9	0.93	K2-28	0.11	K2-47	0.45	K2-66	0.2
K2-10	1.51	K2-29	0.39	K2-48	0.8	K2-67	0.49
K2-11	0.27	K2-30	1.39	K2-49	0.28	K2-68	0.29
K2-12	0.52	K2-31	0.88	K2-50	0.24	K2-69	0.87
K2-13	2.55	K2-32	0.6	K2-51	4.73	K2-70	0.65
K2-14	0.48	K2-33	4.63	K2-52	0.29	K2-71	0.3
K2-15	1.97	K2-34	1.84	K2-53	6.85	K2-72	8.28
K2-16	1.21	K2-35	4.1	K2-54	0.57	K2-73	0.48
K2-17	2.73	K2-36	1.92	K2-55	0.69	K2-74	1.84
K2-18	1.27	K2-37	1.18	K2-56	5.85		
K2-19	0.22	K2-38	0.38	K2-57	0.61		

Table S3 Plasma spectrum S results for liquid collectors along the 3rd section (μg/mL)

Number	S	Number	S	Number	S	Number	S
K3-1	34.90	K3-6	19.43	K3-11	4.08	K3-16	76.28
K3-2	2.35	K3-7	1.00	K3-12	16.88	K3-17	77.21
K3-3	4.89	K3-8	1.38	K3-13	74.51	K3-18	79.81
K3-4	0.52	K3-9	1.43	K3-14	51.57	K3-19	81.52
K3-5	2.65	K3-10	0.10	K3-15	49.66	K3-20	76.07

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