Determination of carbon-bonded sulfur in soils by hydriodic acid reduction and hydrogen peroxide oxidation

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Abstract. A sequential extraction method has been developed for the determination of carbon-bonded sulfur in soils. The soil sample has been sequentially reduced with HI and oxidized with hydrogen peroxide, and finally the residue has been digested with a mixture of nitric acid and perchloric acid. All inorganic sulfur components and ester sulfur has been reduced to H_2S by HI except the un-reducible sulfur including pyritic sulfur, carbon-bonded sulfur and a previously unidentified sulfur fraction. Whereas a part of the carbon-bonded sulfur has been dissolved in the HI reducing solution another part of carbon-bonded sulfur was removed by hydrogen peroxide oxidation. The total carbon-bonded sulfur compose for oxic soils of the HI-dissolved sulfur and the H_2O_2 -oxidized sulfur. However, because the pyritic sulfur can be completely decomposed by H_2O_2 , this form of sulfur should be subtracted from the sum of the two sulfur fractions in case of anoxic soils. Unidentified sulfur components were also detected in the residue after the sequential extraction.

Introduction

Sulfur occurs in soils mainly in organic forms, which can be divided into two distinct fractions: ester sulfur (organic sulfates containing C-O-S linkages, e.g. choline sulfate, phenolic sulfates, sulfated polysaccharides) and carbon-bonded sulfur, in which sulfur is directly bonded to carbon, consisting mainly of sulfur containing amino acids, such as methionine and cysteine.

To determine carbon-bonded sulfur in soils, Frenzy et al. [1] investigated Raney nickel, which reduced hydrocarbon-bonded sulfur to inorganic sulfide, but as the sulfur in many inorganic compounds and sulfur in organic compounds, such as cysteine-S-sulfonate is not directly bonded to carbon, the Raney nickel method was not specific for sulfur bonded to carbon. Recently, a $Sn-H_3PO_4$ method was reported for the reduction of organic sulfur in soils [2], this method was not specific for carbon-bonded sulfur either and would attack a variety of organic and inorganic sulfur compounds.

Since no specific and direct methods are available, carbon-bonded sulfur is estimated in soil and sediment samples as difference between total sulfur and all the other sulfur fractions. Nriagu and Soon [3] subdivided the sulfur fractions in unpolluted and highly polluted sediments into acid volatile sulfur, HCl-soluble sulfur, elemental sulfur, pyrite sulfur, ester sulfur and carbon-bonded sulfur as difference between total sulfur and all of the other fractions. David et al. [4] classified the sulfur into total sulfur, hydriodic acid-reducible sulfur (HI-S) and Zn-HCl reducible sulfur (Zn-HCl-S). HI-S includes all inorganic sulfur and non-carbon-bonded sulfur, while Zn-HCl-S includes most of the non-sulfate sulfur, i.e. sulfides, sulfites, elemental sulfur and thiosulfates. Here the carbon-bonded sulfur was determined as the difference between total sulfur and HI-S. Shan et al. [5] divided sulfur in soils into water soluble sulfur, adsorbed sulfur, HCl-volatile sulfur, dilute HCl soluble sulfur, pyritic sulfur, ester sulfur and carbon-bonded sulfur as difference between the total sulfur and all the other sulfur fractions. In general, quite similar methods were used by numerous researchers [6–12]. The main drawback of these methods is the error, which is the sum of all errors of each fraction and can therefore be large especially for small values of carbon-bonded sulfur.

Another approach to determine the carbon-bonded sulfur in soils is based on the linkage of sulfur to carbon and the quantitative removal or destruction of organic matter in soils.

For this purpose hydrogen peroxide is often used under different experimental conditions [13]. Obviously Sequi and Aringhieri [14] found that the presence of $0.1 \text{ mol} \cdot \text{l}^{-1} \text{ Na}_4\text{P}_2\text{O}_7$ at pH 7 substantially improved the effectiveness of H_2O_2 in removing organic matter from soil due to the chelating action of pyrophosphate, but the procedure was tedious and time-consuming. In some of the sequential extractions, H_2O_2 in an acidic solution was adopted to oxidize organic matter in soil and sediment [15–17] at a higher temperature ($85 \pm 2^\circ\text{C}$). But the oxidation of organic matter was incomplete, about 10–20% organic carbon remained in the residue.

Therefore a method for the determination of carbon-bonded sulfur in soils is developed. A previously unidentified sulfur fraction is found, thus a more accurate determination of carbon-bonded sulfur is warranted.

Experimental section

Apparatus

A Jarrell-Ash model 1155V AtomComp polychromator (Jarrell-Ash Division, Fisher Scientific Company, Waltham MA, USA) was used for the determination of sulfur at SI 180.74 nm. The spectral interferences were corrected [5]. The detection limit (2σ of 10 replicate measurements) was found to be $11 \mu\text{g L}^{-1}$.

The distillation/reflux apparatus described by Landers et al. [6] was modified. The details of the apparatus were described previously [5].

Reagents

The HI reducing solution was prepared according to Landers et al. [6]. All other chemical used in this study were of analytical reagent grade.

Soils

The four surface soils (0–15 cm) used were collected from different provinces of China. The zones encompass an environmental gradient ranging from frigid through temperate to subtropics resulting in soils that vary in nature and amounts of the soil organic matter present as well as in other chemical and physical properties. Table 1 shows some of the soil characteristics.

The soils were air-dried at room temperature, ground to pass a 2 mm sieve, and stored at room temperature. Before analysis and fractionation the soils were subsampled, larger undecomposed plant material removed, and finely ground to < 100 mesh.

Organic carbon measurement

Organic carbon was determined by a routine analysis method [18].

Sulfur fractionation

Total sulfur. An accurately weighed soil sample (0.50 g) was placed into a 50 mL conical flask, and a minimum volume of $0.1 \text{ mol} \cdot \text{L}^{-1}$ HNO_3 was added to moisten the sample thoroughly, then 10 mL of conc. HNO_3 (67%) and 2 mL of conc. HClO_4 (70%) were added. The flask was

covered with a small funnel in order to avoid rapid loss of the acids during the digestion. The sample was refluxed on a hot plate until white silica appeared. Then, the small cover was removed, and the content was heated at about 220 °C until the volume was reduced to about 0.5 mL. The flask was cooled down to the room temperature and the interior surface of the flask was rinsed with 10 mL of $0.1 \text{ mol} \cdot \text{L}^{-1}$ HNO_3 . The solution was heated for a few minutes and filtered with a filter paper. This procedure was repeated for several times. The filtrates were combined and diluted to 50 mL with distilled water for the final determination.

Sequential extraction

According to the present sequential extraction procedure the sulfur in soils was divided into four fractions: HI-reducible sulfur (previously defined as all inorganic sulfur except for pyritic sulfur and ester sulfur in the literature), HI-dissolved sulfur (carbon-bonded sulfur), H_2O_2 -oxidized sulfur (carbon-bonded sulfur) and the sulfur in the residue (previously unidentified sulfur fraction in the literature).

HI-reducible sulfur. An accurately weighed soil sample (1.00 g) was transferred to the boiling flask and connected to the J-N apparatus. The HI-reducing solution (20 mL) was added dropwise from the dropping funnel. The suspension was refluxed by gently boiling for 1 h under N_2 . The H_2S gas was trapped in 10 mL of $0.05 \text{ mol} \cdot \text{L}^{-1}$ NaOH by a stream of N_2 and the sulfide was oxidized with H_2O_2 to sulfate. The solution was shaken for a few seconds and allowed to stand for 10 min to complete the oxidation.

HI-dissolved sulfur. The digest in the boiling flask was filtered with a filter paper into a 50 mL of volumetric flask. All solid residue was washed thoroughly with distilled water, the washings were combined with the filtrate and the solution diluted to the mark with distilled water.

H_2O_2 -oxidized sulfur. The solid residue on the filter paper was transferred into a 200 mL conical flask and 20 mL of H_2O_2 (30%) was added. The flask was covered with a little funnel and heated gently on a hot plate, the reaction was vigorous and the temperature of the suspension rose to 100–103 °C within 10 min. After heating for 1 h, the funnel was removed and the solution was evaporated near dryness. Then, another 20 mL of H_2O_2 (30%) was added and the digestion was repeated. The residue was rinsed with 10 mL of $0.10 \text{ mol} \cdot \text{L}^{-1}$ HNO_3 , the digest was filtered into a 50 mL volumetric flask, the procedure was repeated for several times. Finally, the solution was diluted to the mark with $0.1 \text{ mol} \cdot \text{L}^{-1}$ HNO_3 .

Residue sulfur. The solid residue on the filter paper was transferred into a 100-mL conical flask, and a quite similar procedure was performed using 20 mL of HNO_3 and 4 mL of HClO_4 .

Pyritic sulfur. A subsample of the soil (0.5 g) was treated as for HI-reducible sulfur and HI-dissolved sulfur. To the solid residue on the filter paper transferred back to the boiling flask, 3.0 g of granular zinc was added. The flask was attached to the J-N apparatus and purged with N_2 at 2 bubbles/s. HCl (20 mL, $6 \text{ mol} \cdot \text{L}^{-1}$) was added in three

Table 1. Some characteristics of soil samples

Soil	Order	Family	Texture (%)		
			Clay	Silt	Sand
Mudanjiang	Mollisol	Sandy-silty, mixed	17.8	57.1	25.1
Changchun	Mollisol	Sandy-silty, mixed	10.6	49.8	39.6
Linyi	Entisol	Silty-clayed, mixed	33.5	59.2	7.3
Hangzhou	Ultisol	Fine-silty, mixed	23.4	65.0	11.6

portions from the dropping funnel. After the initial vigorous reaction the flask was boiled gently for 1 h. The H_2S gas was trapped in 10 mL of $0.05 \text{ mol} \cdot \text{l}^{-1}$ NaOH and then oxidized to sulfate with H_2O_2 as described above.

Optimization of H_2O_2 oxidation procedures

To optimize the digestion of soil organic matter, several H_2O_2 oxidation methods reported in the literature were tested (Table 2).

Results and discussion

Effectiveness of H_2O_2 in destroying organic matter in soil

Because sulfur is directly bonded to carbon in the carbon-bonded sulfur compounds, the main assumption for the final determination of sulfur is to selectively reduce the carbon-bonded sulfur to H_2S gas or to quantitatively destroy or remove organic matter from the soils. As no specific reduction reaction seems to be available to meet the former requirement emphasis is put on the latter. For this purpose a comparison was carried out between various pretreatment procedures for soils (Table 2) and their effectiveness in releasing organic carbon (Table 3)

The organic carbon content in untreated soils are in the range of 0.80–3.74%. When soil samples were treated by HI reducing solution, part of the organic carbon was removed. If diluted H_2O_2 solutions ($\text{H}_2\text{O}_2\text{-I}$ and $\text{H}_2\text{O}_2\text{-II}$) or a mixture of $\text{HNO}_3\text{-H}_2\text{O}_2$ were used more but not all organic carbon was recovered. Almost all the organic carbon was transferred to the solution by $\text{H}_2\text{O}_2\text{-Na}_4\text{P}_2\text{O}_7$, and the organic carbon remaining in the residue was only in the range of 1.9–4.5%, but this method is time-consuming and the strong color of the extract may cause troubles in the final ICP-AES determination. Using the method developed here a more efficient oxidation was achieved, with small organic carbon remaining in the residue which was close to the detection limit of the routine method of organic carbon determination. In addition, the procedure is time-saving and the extract solution exhibits only a very slight color and is therefore more suitable for ICP-AES determinations.

To examine whether carbon-bonded sulfur compounds could be destroyed and/or oxidized to sulfate by H_2O_2 -oxidation, several typical carbon-bonded sulfur compounds such as methionine, cystine, taurine and thiamine were added to soil samples. Then, the total sulfur concentrations were determined by ICP-AES and ion chromatography (IC) [3], and the recovery of carbon-bonded sulfur was determined (Table 4). Quantitative recoveries of carbon-bonded sulfur compounds were obtained and a very good agreement was achieved between recoveries determined by ICP-AES and by IC leading to the conclusion that all the carbon-bonded sulfur compounds were destroyed and oxidized to sulfate by H_2O_2 -oxidation.

As there are numerous reports on the performance of HI-reduction in the literature [5] and it is well recognized that all inorganic sulfur compounds except for pyritic sulfur and ester sulfur can be reduced to H_2S by HI

Table 2. Comparison of digestion methods from literature

Method	Reference	Sample amount	Oxidant	Oxidation time	Oxidation temperature	Other conditions
H_2O_2 (I)	[13]	5 g	10 mL water + 1 mL H_2O_2 (30%)	15 min	60–70 °C	5 fold repetition
H_2O_2 (II)	[13]	10 g	20 mL water + 5 mL H_2O_2 (30%)	3 h	60–70 °C	5 mL addition of H_2O_2 after first vigorous reaction
$\text{H}_2\text{O}_2 + \text{Na}_4\text{P}_2\text{O}_7$	[14]	10 g	80 mL 0.10 mol/L $\text{Na}_4\text{P}_2\text{O}_7$ + 20 mL H_2O_2 (30%)	end of reaction	70–80 °C	further H_2O_2 addition until end of reaction
$\text{H}_2\text{O}_2 + \text{HNO}_3$	[15–17]	1 g	5 mL 0.02 mol/L HNO_3 + 5 mL H_2O_2 (30%) (pH 2)	2 h	85 ± 2 °C	two steps
			2 mL H_2O_2 (30%)	3 h	85 ± 2 °C	

Table 3. Organic carbon in soils and in residues after various treatment^a (conditions see Table 2)

Soil	Organic carbon (%)						
	Untreated soil	HI reduction	$\text{H}_2\text{O}_2\text{-I}$	$\text{H}_2\text{O}_2\text{-II}$	$\text{H}_2\text{O}_2 + \text{HNO}_3$	$\text{H}_2\text{O}_2 + \text{Na}_4\text{P}_2\text{O}_7$	Present method
Mudanjiang	3.58	1.86	0.91	0.55	0.22	0.06	0.028
	3.62	1.88	0.89	0.55	0.21	0.08	0.024
Changchun	1.74	1.43	0.80	0.37	0.19	0.07	0.030
	1.70	1.46	0.76	0.35	0.21	0.09	0.027
Linyi	0.80	0.57	0.31	0.20	0.13	0.03	0.010
	0.80	0.55	0.29	0.21	0.11	0.03	0.013
Hangzhou	3.71	2.71	1.58	0.93	0.35	0.12	0.023
	3.77	2.61	1.54	0.90	0.35	0.11	0.025

^a H_2O_2 treatment was performed on the residue after HI reduction

Table 4. Recovery of carbon-bonded sulfur added to soil after H_2O_2 oxidation

Soil	Soil weight (mg)	Compound	Sulfur species	Sulfur (μg)				Recovery (%)	
				Soil	Added ^a	Total sulfur		ICP-AES	IC
						ICP-AES	IC		
Mudanjiang	500	Methionine	$-\text{C}-\text{S}-\text{C}-$	332	200	517.2	499.0	97.2	93.8
						513.8	505.4	96.6	95.0
Mudanjiang	500	Cystine	$-\text{C}-\text{S}-\text{S}-\text{C}-$	332	200	526.5	512.8	99.0	96.4
						533.5	518.0	100.3	97.4
Mudanjiang	500	Taurine	$-\text{C}-\text{SO}_2\text{OH}$	332	200	512.6	490.2	96.3	92.1
						506.6	491.8	95.2	92.4
Mudanjiang	500	Thiamine		332	200	529.3	515.7	99.5	96.9
						524.1	508.9	98.5	95.6

^a Added as carbon-bonded sulfur**Table 5.** Carbon-bonded sulfur reduced and dissolved by HI reduction solution (% of total sulfur)

Compound	Sulfur added as carbon-bonded sulfur (μg)	Sulfur found (%)			
		HI-reducible sulfur		In HI reducing solution	
Methionine	1000	0.42	0.46	96.0	95.2
Cystine	1000	0.25	0.23	100.9	103.1
Taurine	1000	0.11	0.11	99.8	99.3
Thiamine	1000	0.63	0.65	97.1	97.9

reduction, one can infer that the combination of HI-reduction and H_2O_2 -oxidation probably offers a sequential extraction procedure for the determination of carbon-bonded sulfur in soils.

Validity of the sequential extraction procedure for the determination of carbon-bonded sulfur

To examine the sequential extraction procedure of HI-reduction followed by H_2O_2 oxidation for the determination of carbon-bonded sulfur in soils, a series of experiments were carried out. Several typical carbon-bonded sulfur compounds such as methionine, cystine, taurine and thiamine were chosen to verify whether HI is able to reduce these compounds to H_2S . The results are shown in Table 5. It indicated that the carbon-bonded sulfur compounds were not reduced to H_2S by HI and almost completely remained in the HI reducing solution. Additionally, some of inorganic minerals present in the soil such as montmorillonite, illite, K-feldspar and pyrite containing no organic matter were also investigated. The minerals were subjected to HI-reduction followed by H_2O_2 -oxidation, and the sulfur remaining in the residue after the above sequential treatments was determined after the digestion with a mixture of HNO_3 - HClO_4 . The organic carbon contents in the samples were also measured. All the results are summarized in Table 6. Sulfur in

the montmorillonite, illite and K-feldspar can be reduced to H_2S by HI. No sulfur was detected either in the HI-reducing solution nor after H_2O_2 oxidation. Although pyritic sulfur cannot be reduced to H_2S by HI-reduction, it can be decomposed by H_2O_2 oxidation. The reason for 4% of sulfur in the pyrite being reduced to H_2S by HI was probably due to impurities like other sulfur compounds present.

Residual sulfur fractions were found after the HI reduction/ H_2O_2 oxidation, not being identified in the literature.

Humic substances complex and poor-defined polydisperse mixtures of heterogeneous organic polyelectrolytes [19] are the major component of soil organic matter and contain the bulk of the total sulfur in many soils, particularly in humid regions. Purified commercial and soil humic acids were subjected to HI-reduction and H_2O_2 oxidation, the sulfur was divided into HI-reducible sulfur, HI-dissolved sulfur and H_2O_2 -oxidized sulfur fractions (Table 7). Since all humic acids were destroyed by H_2O_2 oxidation no residue remained. The HI-reducible sulfur fraction includes ester sulfur and all inorganic sulfur fractions except pyritic sulfur and unidentified residue sulfur. The HI-dissolved sulfur fraction can be related to the carbon-bonded sulfur, H_2O_2 -oxidized sulfur includes carbon-bonded sulfur and pyritic sulfur. If the humic acids were directly destroyed by H_2O_2 without the sequential pretreatment or by the mixture of HNO_3 and HClO_4 the total sulfur concentrations were quite similar, no matter what method was used. This finding further verified that all organic matter was quantitatively destroyed by H_2O_2 oxidation.

Based on the above study a sequential extraction procedure was proposed for the determination of carbon-bonded sulfur in soils as given in the experimental section.

Determination of carbon-bonded sulfur in soils

The recommended sequential procedure was applied to the determination of carbon-bonded sulfur in soils. The results are given in Table 8.

Table 6. Sulfur fractions of several natural minerals

Mineral	Sample weight (g)	Sulfur fraction ($\mu\text{g/g}$)						Organic carbon content
		HI-reducible S		HI-dissolved S	H_2O_2 -removed S		Residue S	
Montmorillonite	2.00	216.0	211.0	ND ^a	ND		163.2 167.9	0.01
Illite	2.00	143.7	145.9	ND	ND		150.5 153.5	ND
K-feldspar	2.00	141.3	147.1	ND	ND		134.3 128.5	ND
Pyrite	0.05	22.2 ^b	22.6 ^b	ND	532.0 ^b	527.4 ^b	ND	ND

^a ND – Not detected^b [mg/g].**Table 7.** Sulfur in humic acid [$\mu\text{g/g}$]

Humic acid	Sequential extraction						Total sulfur			
	HI-reducible S		HI-dissolved S		H_2O_2 -oxidized S		After H_2O_2 oxidation		After digestion with $\text{HNO}_3/\text{HClO}_4$	
Shanghai	269	263	585	571	670	696	1528	1552	1568	1592
Beijing-1	578	562	2162	2196	2080	2060	4845	4821	4982	4948
Beijing-2	1335	1367	1759	1765	2372	2342	5551	5589	5621	5645

Table 8. Carbon-bonded sulfur in soils [$\mu\text{g/g}$]

Soil	Sulfur fraction								Carbon-bonded sulfur		Total sulfur	
	HI-reducible S		HI-dissolved S		H_2O_2 -removed S		Residue S		HI-dissolved + H_2O_2 -removed S			
Mudanjiang	272.5	260.3	194.0	190.0	130.0	138.0	89.0	83.0	324.0	328.0	672.0	656.0
Changchun	566.0	559.8	291.5	302.5	155.5	148.9	147.0	139.0	447.0	451.4	1220.0	1186.0
Linyi	120.4	131.2	52.2	56.8	75.0	69.0	44.5	38.1	127.2	125.8	302.2	317.6
Hangzhou	553.5	566.2	380.0	369.0	258.0	264.0	101.2	105.0	638.0	633.0	1290.0	1306.0

Since all these soils are oxic agricultural soils in which no pyritic sulfur was detectable, carbon-bonded sulfur was determined by summing HI-dissolved sulfur and H_2O_2 -oxidized sulfur. Organic carbon content in these soils and in the soil residues after sequential extraction were determined (Table 3). As can be seen from Table 8, total sulfur in these soils differed drastically (from the minimum value of 309.9 $\mu\text{g/g}$ in Linyi soil to the maximum value of 1298 $\mu\text{g/g}$ in Hangzhou soil). But these total sulfur values were in significant correlation with the organic carbon values ($r = 0.729$), indicating most of the total sulfur are organic-bound. HI-reducible sulfur accounted for about 40% of the total sulfur (40.1–46.8%), ester sulfur would be smaller. HI-dissolved carbon-bonded sulfur values were higher than that obtained by H_2O_2 oxidation except Linyi soil. But only a small portion of organic carbon was removed by HI (see Table 3), suggesting the organic matter containing sulfur was dissolved by HI. This was likely to be fulvic acid which is dissolved in acid medium.

In the case of anoxic soils there are various amounts of pyritic sulfur present, which can be recovered by H_2O_2 -oxidation treatment. Therefore, a subtraction of the py-

ritic sulfur is required from the sum of HI-dissolved sulfur and H_2O_2 -oxidized sulfur fractions.

Residue sulfur fractions

Traditionally, many researchers believe that all inorganic sulfur fractions and ester sulfur can be reduced to H_2S by HI-reduction except for pyritic sulfur and carbon-bonded sulfur fractions. Therefore, subtraction method is frequently used for the estimation of carbon-bonded sulfur after the determination of the above mentioned sulfur fractions. However, residual sulfur fractions were detected in the present study after the sequential treatment and the fractions have not been identified previously in the literature. Although the chemical nature of these fractions are not certain at present, these unidentified sulfur fractions are more likely to be inorganic sulfur because the natural minerals of montmorillonite, illite and K-feldspar contain no organic matter and quite significant contents of residue sulfur have been detected after the sequential treatment. The explanation for the chemical nature of the unidentified sulfur fractions may be quite complicated. David et al. [4] suggested that an unknown proportion of

inorganic sulfate maybe exist in an occluded form. The discovery of the residue sulfur gave direct evidence for this postulation. Usually, trace metals in soils and sediments have been divided into various fractions by sequential extractions, such as exchangeable, bound to carbonates, bound to Fe-Mn oxides, bound to organic matter and residue. The residual form were thought to be metals being occluded within the crystal structure of the residual solid containing mainly primary and secondary minerals after other fractions have been removed. This may be the same case for residue sulfur.

Conclusion

Based on the present study the following conclusions can be drawn:

(1) All organic matters in soils can be destroyed and typical carbon-bonded sulfur compounds can be oxidized to sulfate by undiluted H_2O_2 -oxidation at higher temperatures than that used by other researchers. Pyritic sulfur is also recovered by the H_2O_2 treatment. Certain parts of carbon-bonded sulfur are dissolved in HI-reducing solution. Thus, a sequential procedure has been suggested for the determination of carbon-bonded sulfur in soils by HI-reduction followed by H_2O_2 -oxidation. In the case of oxic soil the carbon-bonded sulfur is the sum of HI-dissolved sulfur and H_2O_2 -oxidized sulfur. For anoxic soil the pyritic sulfur fraction should be subtracted from the above sum.

(2) A previously unidentified sulfur fraction, which is experimentally defined as residue sulfur, is found in soil samples subjected to the above sequential treatment. The residue sulfur is great in comparison to the total sulfur in the soil samples. Therefore, a large error would be introduced in the estimation of carbon-bonded sulfur by the different methods widely used in the literature. This mistake can be avoided by the method recommended.

(3) Further study is still required to develop a better analytical technique for the determination of carbon-bonded sulfur. The chemical nature of the residual sulfur needs to be ascertained. Although these questions have not been satisfactorily answered, a certain progress has

been made in the determination of carbon-bonded sulfur. The sequential procedure recommended has advantages over methods reported in the literature. The most important aspects of the developed sequential procedure are more accurate determination of carbon-bonded sulfur and discovery of the unidentified sulfur fractions. This will stimulate the research on the chemical characteristics of sulfur.

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