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Rapid effects of terrestrial alteration on highly siderophile elements in the Sutter's Mill meteorite

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Abstract

The ^{187}Re - ^{187}Os isotopic systematics of many bulk chondrites plot well beyond analytical uncertainties of a primordial isochron. Limited variations in $^{187}\text{Os}/^{188}\text{Os}$, coupled with large variations in Re/Os ratios among chondrites, suggest that this apparently open-system behavior is a result of the comparatively recent gain or loss of Re and/or Os. In order to assess whether or not rapid alteration in the terrestrial environment could be responsible for open-system behavior in chondrites, four pieces of the Sutter's Mill meteorite were examined for Os isotopic systematics and abundances of highly siderophile elements. Pieces SM1 and SM2 were collected prior to a rain event, within 2 days of the fall. Pieces SM51 and SM53 were collected after a rain event. There are significant but minor relative and absolute variations in the abundances of the highly siderophile elements, as well as $^{187}\text{Os}/^{188}\text{Os}$ among the four pieces. Rhenium-Os isotopic data for SM1 and SM2 plot within analytical uncertainties of a primordial isochron, while powders made from SM51 and SM53 do not. These results suggest that interactions with rain caused some redistribution of Re, and to a lesser extent Os, within small pieces of the meteorite. Thus, Re-Os isotopic systematics of <dm-size pieces of chondrites must be considered susceptible to modification after only a short time on the surface, where exposed to rain.

INTRODUCTION

Differences in the relative abundances of the highly siderophile elements (HSE: including Re, Os, Ir, Ru, Pt, and Pd), as well as $^{187}\text{Os}/^{188}\text{Os}$ ratios (reflecting longterm Re/Os), of bulk samples of chondrites have commonly been interpreted to reflect primary fractionations resulting from high-temperature nebular condensation or evaporation processes in precursor components (Horan et al. 2003). Such fractionations may prove to be diagnostic in refining

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SUPPORTING INFORMATION

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our understanding of the high-temperature processing of different chondritic precursor components, although the mechanisms causing the HSE fractionations are still poorly understood (Mason and Taylor 1982; Horan et al. 2009). Osmium isotopic compositions, and the relative abundances of the HSE, have also proved useful for “fingerprinting” the dominant types of planetesimals added to planetary mantles by late accretion (Becker et al. 2006; Fischer-Gödde et al. 2011), as well as in identifying impactor types involved in the generation of impact melt rocks associated with the lunar basins and terrestrial impact craters (Norman et al. 2002; Tagle and Berlin 2008; Fischer-Gödde and Becker 2012; Liu et al. 2015; Gleißner and Becker 2017). Despite the importance of these applications, it remains unclear to what extent HSE fractionations reflect secondary processes on the parent bodies of chondrites, are a result of comparatively recent asteroidal (within the past 2 Ga), or possibly terrestrial alteration processes. This uncertainty presently limits the utility of these elements to study nebular processes and fingerprint genetically distinct materials.

Definitive evidence for the effects of late-stage secondary processes on HSE ratios has mainly come from several Re-Os isotopic studies of bulk chondrites ($^{187}\text{Re} \rightarrow ^{187}\text{Os} + \beta^-$; $\lambda = 1.677 \times 10^{-11} \text{ yr}^{-1}$) (e.g., Walker et al. 2002; Fischer-Gödde et al. 2010). These studies have shown that isotopic data for many bulk chondrites plot significantly beyond analytical uncertainties of a primordial isochron, along which chondrites should plot if formed early in solar system history from a reservoir with a common initial $^{187}\text{Os}/^{188}\text{Os}$ (Fig. 1). In some cases, deviations from the primordial isochron are so large that it would be challenging to explain them as a consequence of open-system behavior prior to about two billion years ago. For example, a negative 3% offset in $^{187}\text{Os}/^{188}\text{Os}$ from a present-day primordial isochron would have required a nearly 16% reduction in Re/Os, if a modification event occurred at 3 Ga. Such a large change in Re/Os would place a sample outside the range in Re/Os in bulk chondrites, which is not observed. Thus, the Os isotopic data most likely indicate late-stage movement of Re and/or Os in bulk samples, as has also been noted for calcium-aluminum-rich inclusions (CAI) and chondrules present in carbonaceous and ordinary chondrites (Becker et al. 2001; Horan et al. 2009; Archer et al. 2014). Such late-stage alteration of bulk chondrites has rarely been previously recognized with regard to other elements or isotope systems, although some early Rb-Sr isotopic studies of bulk chondrites also hinted at comparatively recent alteration (e.g., Kaushal and Wetherill 1970).

Sutter’s Mill is a meteorite that entered Earth’s atmosphere on April 22, 2012. Its fall resulted in the collection of >75 pieces of the meteorite in northern California. The masses of the pieces range from 0.1 to 205 g. Sutter’s Mill is a regolith breccia composed mainly of CM type carbonaceous chondrite material, and also includes some enstatite chondrite or achondrite material (Jenniskens et al. 2012; Zolensky et al. 2014). It is characterized by substantial isotopic and mineralogic heterogeneity (Jenniskens et al. 2012; Yamakawa and Yin 2014). The heterogeneities likely result from a complex formation history on the parent body surface. For the purposes here, the Sutter’s Mill meteorite enables the comparison of materials collected almost immediately after entering the terrestrial atmosphere, with materials affected by subsequent terrestrial alteration. This is possible because several pieces of Sutter’s Mill were collected before they were exposed to rain, while most pieces were collected after a major rain event. The pre-rain specimens contain the calcium sulfide oldhamite, a water-soluble mineral, that is absent in any of the studied specimens recovered

after exposure to rainwater (Jenniskens et al. 2012). This was one of the first and most obvious observations of terrestrial alteration in Sutter's Mill. Consequently, Sutter's Mill provides a rare opportunity to assess whether or not short-term terrestrial alteration, in the form of rain, can affect the relative abundances of the HSE (especially Re/Os) in a chondrite. Although it may seem unlikely that such limited exposure of a meteorite to rain could affect elemental abundances in the interiors of the collected pieces, several studies of presolar, Os-bearing components in chondrites have shown that substantial Os can be easily removed from a meteorite by very gentle leaching (e.g., Reisberg et al. 2009; Yokoyama et al. 2010).

ANALYTICAL METHODS

Two ~100 mg interior fragments of the SM1 and SM2 pieces were acquired from the Field Museum (ME 6051) and Sutter's Mill Meteorite Consortium (Jenniskens et al. 2012), respectively. These pieces were recovered on April 24, 2012, 2 days after the meteorite fall. Both were collected prior to rain in the area, and serve as the most pristine pieces of Sutter's Mill that exist. A total of 5.6 and 4.0 g of SM1 and SM2 were collected, respectively. After collection, the pieces were stored in desiccators and split dried with a rock splitter. Pieces were characterized using scanning electron microscopy and X-ray μ CT (see supporting information). Each piece was ground into a fine powder for analysis, using an agate mortar and pestle. Piece SM51 consisted of 12.3 g total. The piece was recovered on May 2, 2012, 10 days after the meteorite fall, and following a significant rain event. It was found on a driveway of a private house underneath a tree on dirt. Approximately 0.5 g of the interior of SM51 (the UC Davis Collection, see table S3 in Jenniskens et al. 2012) was powdered and homogenized using an agate mortar and pestle. Two ~50 mg aliquots were taken from this powder, SM51-A and SM51-B, and were separately analyzed. Piece SM53 (also from the UC Davis Collection) was found on May 11, 2012. It is the main mass (205 g) recovered thus far (Jenniskens et al. 2012) on a public land on grass. Similar to SM51, an interior fragment of this piece consisting of ~0.6 g was powdered and homogenized using an agate mortar and pestle. Two ~200 mg aliquots of the powder, SM53-A and SM53-B, were taken and analyzed. The latitude and longitude at which each piece of Sutter's Mill was collected are provided in table S3 in Jenniskens et al. (2012).

Both X-ray μ CT scan and SEM/EDS analysis of SM1 reveal a uniform lithology for this piece (see supporting information for μ CT image stack). By contrast, X-ray μ CT scans of portions of SM51 and SM53 show some (1–2 mm) clasts (see supporting information for μ CT image stack). Consequently, it must be concluded that the lithologies of SM1 and SM51 and SM53 are somewhat different.

Samples were analyzed at the University of Maryland for HSE concentrations (by isotope dilution) and $^{187}\text{Os}/^{188}\text{Os}$ isotopic composition, using standard high-temperature acid digestion, chemical purification, and mass spectrometric techniques, most recently described in Archer et al. (2014). In brief, HSE spikes (^{185}Re , ^{190}Os , ^{191}Ir , ^{99}Ru , ^{194}Pt , and ^{105}Pd) and sample powders were mixed and digested inside sealed Pyrex® Carius tubes for >24 h at 260 °C, in a 1:2 mixture of concentrated hydrochloric and nitric acids. Osmium was separated by solvent extraction into carbon tetrachloride, and back-extracted into concentrated hydrobromic acid. Osmium fractions were further purified by microdistillation

into concentrated hydrobromic acid. The remaining HSE were purified by anion exchange chromatography. Total analytical blanks for the HSE for the period during which the meteorite was processed averaged 1 pg (Re), 0.7 pg (Os), 0.3 pg (Ir), 4 pg (Ru), 75 pg (Pt), and 11 pg (Pd). The quantities of the HSE present in each aliquot of sample powder were at least 1000 times greater than the blanks and the blanks had no impact on the measurements. Osmium isotopic compositions were measured as OsO_3^- by thermal ionization mass spectrometry, using a Thermo Fisher Triton mass spectrometer. For this measurement, the purified Os fraction was dried onto Pt filaments and coated with barium hydroxide. Ionization was achieved by resistively heating the filament to 800–900 °C. Ion beam intensities were measured by a static collection routine in Faraday cups. The isotopic compositions of the remaining HSE were measured in Faraday cups using a Nu Plasma, multicollector inductively coupled plasma–mass spectrometer (SM1, SM2, SM51), or in an electron multiplier using an Thermo Element 2, single collector inductively coupled plasma–mass spectrometer (SM53). The chemical blanks did not vary with the instrument used. An Aldrich W standard was added to all Re fractions measured so that instrumental fractionation could be monitored and corrected for using the natural $^{184}\text{W}/^{186}\text{W}$ ratio of the added W. For all analyses, uncertainties for Ir, Ru, Pt, and Pd concentrations are estimated to be 1%. Uncertainties are estimated to be 0.15% for Re and 0.1% for Os. Reproducibility of $^{187}\text{Os}/^{188}\text{Os}$ for these quantities of Os is $\sim\pm 0.05\%$.

RESULTS

The absolute and relative abundances of HSE in all of the Sutter's Mill materials examined here are within the range of compositions previously reported for carbonaceous chondrites. The $^{187}\text{Os}/^{188}\text{Os}$ ratios of SM1 and SM2 are 0.1260 and 0.1270 (Table 1), respectively, which are also within the range of carbonaceous chondrites. The $\sim 0.8\%$ difference between the two pieces is well beyond the analytical uncertainty of 0.05%, and spans about half of the range of variance in the Os isotopic compositions of carbonaceous chondrites (Fig. 1). Piece SM2 has higher Re, Os, Pt, and Pd absolute abundances than SM1, and correspondingly lower Ir and Ru (Fig. 2). Absolute abundances of the HSE in SM1 and SM2 differ by as much as 14% (for Pd). The relative abundances of HSE vary by as much as 20% (Pd/Ir) between the two pieces. These differences reflect moderate chemical heterogeneity of HSE within comparatively small (~ 50 mg) pieces of the meteorite examined. Heterogeneities of similar magnitude have been reported for duplicate analyses of small, bulk pieces of other chondrites (Horan et al. 2003).

The Os isotopic compositions of the two aliquots of SM51 powder are identical within the uncertainties of $\pm 0.05\%$, averaging 0.1261, and within the range of carbonaceous chondrites. Most of the HSE concentrations for the two aliquots are in agreement within $\pm 0.4\%$ of one another, consistent with homogeneity of the finely ground powder (Fig. 2). However, Re and Os concentrations differ by 4 and 2%, respectively. Furthermore, the resulting $^{187}\text{Re}/^{188}\text{Os}$ ratios differ by $\sim 6\%$. These variations are well above the 0.15% analytical reproducibility of Re and Os for standards, and indicate moderate heterogeneity, for these elements, between the two powder aliquots.

The Os isotopic compositions of the two aliquots of homogenized SM53 powder differ by 0.13%, and average 0.1279. This ratio is substantially higher than that measured for the other pieces, and is higher than known carbonaceous chondrites. The ratio is more typical of ordinary chondrites. Most of the HSE concentrations for the two aliquots are in agreement within $\pm 0.5\%$ of one another; however, as with SM51, Re and Os concentrations differ considerably more, with variations of $\sim 6\%$ for both elements. In contrast to SM51, however, $^{187}\text{Re}/^{188}\text{Os}$ ratios differ by only $\sim 0.3\%$. The abundances of Ir and Ru are similar to that of SM51, but Pd and especially Pt are as much as 15% higher than for SM51.

DISCUSSION

Sutter's Mill pieces SM1 and SM2, collected soon after the fall, have Re-Os isotopic characteristics consistent with closed-system behavior since formation in the nascent solar system. This is evidenced by the observation that both samples plot within uncertainties of a 4.57 Ga reference isochron, with ϵ_{Os} values (parts per thousand deviation of the sample from the isochron; Table 1) of +1.9 (SM1) and +0.3 (SM2), given uncertainties of $\sim \pm 2$ units. Although the two pieces appear to have maintained long-term closed-system isotopic evolution, the relative abundances of the HSE vary between the two pieces. These variations in the relative abundances of HSE, especially Re/Os, likely reflect minor differences in the distribution of the host phases for HSE in these rocks, probably reflecting minor differences in lithology, rather than postformation open-system behavior. This interpretation is consistent with the observation that Sutter's Mill contains CM chondrite and other lithologies, which were exposed to different degrees of thermal metamorphism and aqueous alteration, presumably soon after formation (Jenniskens et al. 2012; Zolensky et al. 2014). Given the differences between these two pre-rain pieces exhibiting long-term closed-system behavior of the HSE, neither piece can be considered to be representative of the bulk composition of the meteorite.

In contrast to SM1 and SM2, the powder aliquots of SM51 exhibit strong isotopic evidence for open-system behavior of the Re-Os system. Neither aliquot plots within uncertainty of the 4.57 Ga reference isochron with ϵ_{Os} values of -8.9 and -30.7 . Both aliquots have essentially identical $^{187}\text{Os}/^{188}\text{Os}$, yet considerably different Re/Os. Both plot beneath the isochron, consistent with the gain of Re, loss of Os, or a combination of both. Uniform $^{187}\text{Os}/^{188}\text{Os}$ but variable Re/Os have been observed for replicate analyses of some other small pieces of bulk ordinary and carbonaceous chondrite samples (Walker et al. 2002; Fischer-Gödde et al. 2010). The difference in Re/Ir between the two powder aliquots is much greater than the difference in Os/Ir, two elements with similar geochemical behaviors. This provides an indication that Re was more mobile than Os (Fig. 2). Thus, Re addition was the dominant aspect of the open-system behavior for this piece of Sutter's Mill. Furthermore, the observation suggests that the phase(s) from which the Re was mobilized was not homogenized by the fine powdering of the material, yet other HSE were. This, in turn, suggests that the excess Re was incorporated in a phase unique to it among the HSE.

The two aliquots of SM53 also show evidence for open-system behavior, although the effects are more muted than SM51, with ϵ_{Os} values of +6.6 and +5.8 that are significantly offset from the reference isochron, yet overlap with one another within analytical

uncertainties. The smaller deviations of the isotopic data for this piece compared with SM51 may reflect the larger mass of the recovered pieces. Alternatively, it is possible that the larger size of the powder aliquots analyzed averaged out some elemental/isotopic heterogeneity.

Differences in the $^{187}\text{Os}/^{188}\text{Os}$ ratios among the four pieces are significant. SM1 and SM2 have isotopic compositions that are consistent with the dominant CM2 lithology as observed with SEM (see supporting information for SM1 and Jenniskens et al. [2012] for SM2), although xenolithic enstatite material was observed in SM2 (Jenniskens et al. 2012). The higher ratios measured in SM51 and especially SM53 are consistent with them containing an even higher proportion of the enstatite chondrite (or achondrite) lithology that has been previously identified (Jenniskens et al. 2012). Unfortunately, no petrologic observations were made of the small pieces of SM51 and SM53 from which powders were made, so there is no supporting evidence for the surmised petrologic difference.

The long-lived Re-Os isotope system cannot be used to directly determine the timing of open-system behavior (e.g., Archer et al. 2014). Nonetheless, the two most pristine samples of Sutter's Mill, SM1 and SM2, are characterized by evident closed-system Re-Os isotopic system behavior, while aliquots of powders from two pieces exposed to rain (SM51 and SM53) are characterized by open-system behavior. These observations lead to the supposition that the rain led to a modest redistribution of Re, and possibly to a lesser extent Os, within pieces exposed to the rain. If the Re movement was simply redistribution from one portion of the meteorite to another, it might be expected that small portions of individual pieces of the meteorite should be characterized by both Re enrichments and depletions. It is worthy of note that fragments from the SM51 and SM53 pieces plot on either side of the primordial isochron, indicating that addition and removal of Re is possible. It is also possible that terrestrial Re was added to the piece of SM51, as rainwater passed into the interior through the comparatively porous matrix. The addition of terrestrial Re (and Os) has previously been suggested for some desert meteorites, most notably for shergottites (Brandon et al. 2012).

These results highlight the likelihood that even very short-term, modest interactions between small pieces of chondritic meteorites and the terrestrial environment, especially rain, can lead to significant modifications to the abundances of at least some of the HSE. Rhenium appears to be the most vulnerable of the HSE. Thus, HSE data for meteorite falls with even very short-term histories on Earth's surface, prior to collection, must be carefully scrutinized. This in turn leads to the tentative suggestion that much of the variability about the primordial isochron in the Re-Os isochron plot (Fig. 1) is the result of recent Re mobility, as has previously been shown for ureilites (Rankenburg et al. 2007). This is somewhat surprising in that many of the ordinary (e.g., Cenicerros, Forest City, Forest Vale, Chico) and carbonaceous (e.g., Vigarano, Lancé) chondrites characterized by largest deviations from a primordial isochron were falls, including some that were collected from dry desert environments.

Fortunately, the evidence for Re mobility relative to Os suggests that $^{187}\text{Os}/^{188}\text{Os}$ ratios for bulk meteorites primarily reflect the long-term Re/Os of the meteorite, and that $^{187}\text{Os}/^{188}\text{Os}$

remains a strong tracer of early solar system processes, among chondrites. Nevertheless, it must also be concluded that Os isotopic compositions of bulk chondrites reflect both large-scale, i.e., differences between chondrite classes, and small-scale processes, i.e., isotopic heterogeneity within a meteorite, such as between SM1 and SM2.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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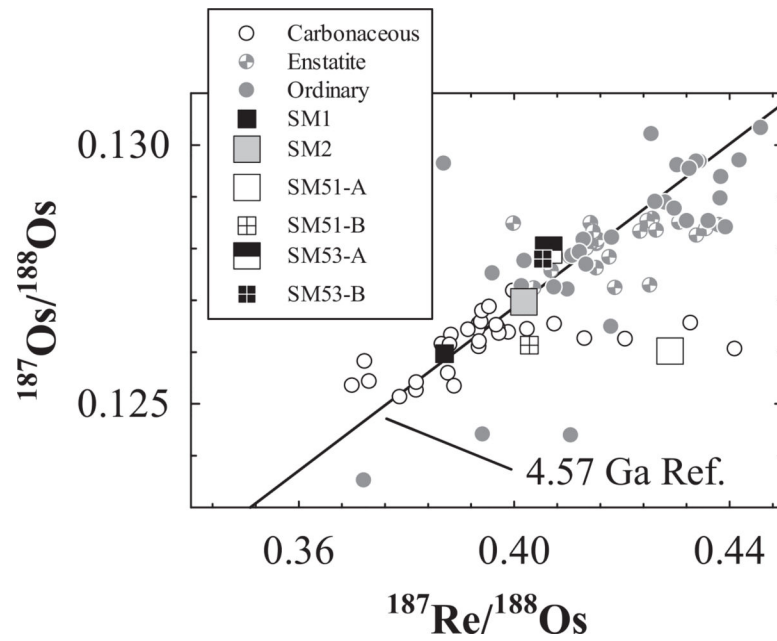


Fig. 1. Rhenium-Os isochron diagram for whole rock chondrites, including the four samples of Sutter's Mill examined here. Note that a large proportion of the bulk chondrite data plot beyond analytical uncertainties (smaller than symbol sizes) of a primordial reference isochron. Comparison data for carbonaceous, ordinary, and enstatite chondrites are from Walker et al. (2002) and Fischer-Gödde et al. (2010).

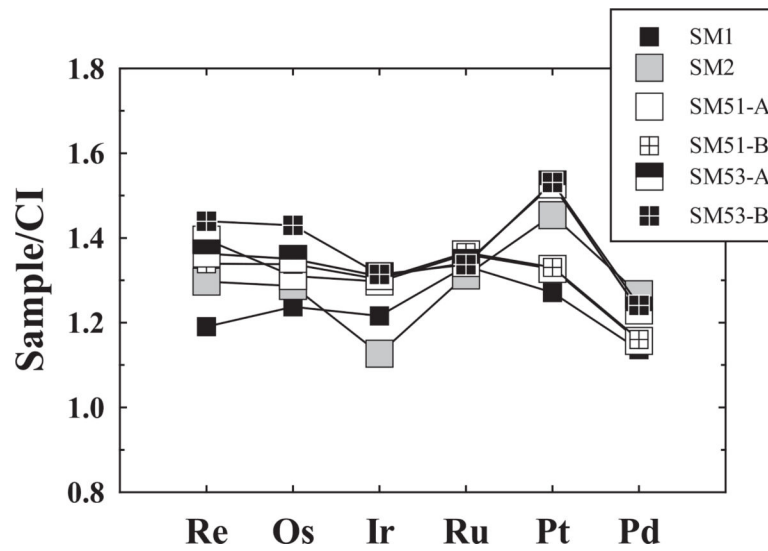


Fig. 2.

CI chondrite-normalized plot of highly siderophile element data for different pieces of the Sutter's Mill meteorite, and two different powder aliquots for the pieces SM51 and SM53. Note that the vertical scale is linear and highlights small variations in abundances relative to one another, and relative to the CI reference (CI data for Orgueil are from Horan et al. 2003). Uncertainties are the same or smaller than symbol sizes.

Table 1.

Highly siderophile element concentrations (in ng g^{-1}) and ^{187}Re - ^{187}Os isotopic data for bulk subsamples of the Sutter's Mill meteorite. Data for SM51-A and -B were previously published in Jenniskens et al. (2012). For all analyses, uncertainties for Ir, Ru, Pt, and Pd concentrations are estimated to be 1%. Uncertainties are estimated to be 0.15% for Re, and 0.1% for Os. δ_{Os} is the deviation in parts per thousand of Re-Os isotopic data for a sample from a primordial 4.568 Ga isochron established for calcium-aluminum-rich inclusions (Archer et al. 2014) where: $\delta_{\text{Os}} = 10^4 \times (^{187}\text{Os}/^{188}\text{Os}_{\text{sample}} - (0.9517 + 0.7908 \times ^{187}\text{Re}/^{188}\text{Os}_{\text{sample}}))$.

Sample	Wt. (mg)	Re	Os	Ir	Ru	Pt	Pd	$^{187}\text{Os}/^{188}\text{Os}$	2σ	$^{187}\text{Re}/^{188}\text{Os}$	2σ	δ_{Os}	2σ
SM1	51.2	45.60	567.5	553.8	866.4	1092	639.6	0.12597	0.00010	0.3871	0.00006	1.9	1.0
SM2	50.1	49.21	590.0	513.3	852.0	1249	713.5	0.12697	0.00010	0.4018	0.00006	0.3	1.0
SM51-A	50.0	53.49	600.7	591.0	884.2	1140	651.2	0.12602	0.00010	0.4289	0.00006	-30.7	1.0
SM51-B	55.8	51.31	613.5	593.4	887.0	1143	653.0	0.12613	0.00010	0.4028	0.00006	-8.9	1.0
SM53-A	203.2	52.22	619.1	596.6	870.9	1311	691.5	0.12797	0.00010	0.4064	0.00006	6.6	1.0
SM53-B	194.9	55.15	655.6	598.7	868.5	1315	698.6	0.12780	0.00010	0.4053	0.00006	5.8	1.0