Comment on "Carbon content drives high temperature superconductivity in a carbonaceous sulfur hydride below 100 GPa" by G. A. Smith, Ines. E. Collings, E. Snider, D. Smith, J. S. Smith, M. White, E. Jones, P. Ellison, K. V. Lawler, R. P. Dias and A. Salamat, Chem. Commun, 2022, 58, 9064

J. E. Hirsch
Department of Physics, University of California, San Diego, La Jolla, CA 92093-0319, USA

Experimental data supporting the claim that a carbonaceous sulfur hydride (CSH) under pressure is a high temperature superconductor were presented in Refs. [1] and [2]. Here we report results of a mathematical analysis that indicates that with probability larger than $1 - 10^{-338}$ some of those data were not measured in a laboratory, contrary to what the papers claim. This finding undermines confidence in the claim that *any* of the experimental evidence reported in those papers reflects the properties of real physical samples of CSH.

PACS numbers:

I. INTRODUCTION

Ref. [1] reported the discovery that CSH is a high temperature superconductor above pressures of 138 GPa, and the first room temperature superconductor at pressure 267 GPa. Ref. [2] reports further measurements on CSH using the same sample preparation procedure and the same measurement methods used in Ref. [1], and the finding that CSH is also superconducting at high temperatures and much lower pressures, below 100 GPa. If these findings reflect reality, they represent important discoveries of high relevance to the understanding of superconductivity in nature. Conversely, if these findings do not reflect reality, it is important to establish that to avoid being misled by a false understanding of such significant issues. References [1] and [2] have 4 authors in common including the corresponding authors (Ranga P. Dias for Ref. [1], Ashkan Salamat for Ref. [2]).

Fig. 1, reproduced from Fig. S14 of Ref. [2], shows results for critical temperatures versus pressure for all the CSH samples studied. In order to understand the significance of the results presented in Ref. [2], it is necessary to have confidence that all the results shown in its Fig. S14 of Ref. [2] (Fig. 1 here) reflect data obtained through actual measurements of the physical samples properly processed, as the papers claim. In fact however, Ref. [1] has recently been retracted by the journal [4], without the authors' agreement [5], due to concerns of the journal's editors about the processing steps used in handing what was reported as "raw data". Adding to these concerns, in this Comment we present clear evidence that the raw data underlying one of the points shown in Fig. 1, indicated by the arrow, were not obtained through actual physical measurements on a physical sample. If factual, this undermines the credibility of all the data shown in Fig. 1, and as a consequence of all the results presented in Ref. [2]. Our analysis shows that this can be established with mathematical certainty.

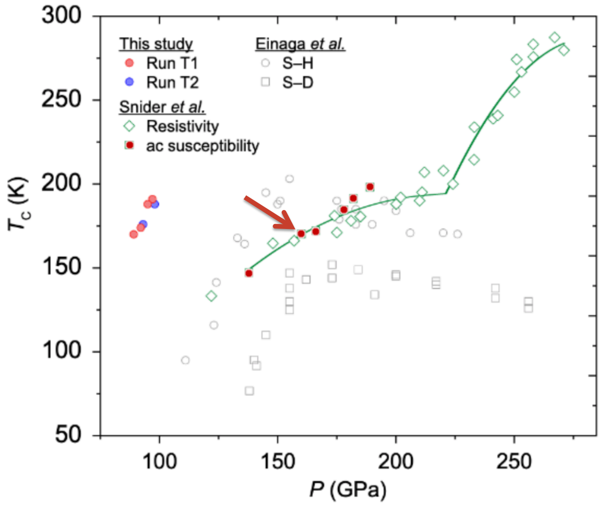

FIG. 1: Figure reproduced from Fig. S14 of Ref. [2], with arrow added. Critical temperature versus pressure for CSH, from experiments reported in Refs. [1, 2] and for SH_3 from Einaga et al, Ref. [3]. The arrow shows the point we are focusing on in this Comment.

The red points in Fig. 1 are derived from ac magnetic susceptibility measurements [1, 6]. Ac magnetic susceptibility is a useful measurement to detect the existence of superconductivity in materials under high pressure [7–12]. Because of the smallness of the sample required by the geometry of the diamond anvil cell, the detected signal is typically a small drop in a large signal (measured as a voltage in a pickup coil) coming from the superposition of the sample and the background signals, according to the relation

$$\textit{Supercond. Signal} = \textit{raw data} - \textit{background signal}. \tag{1}$$

Customarily, the background signal is obtained in a separate measurement at a lower pressure where no superconducting transition is expected [7–12].

In Ref. [6], numerical values for the raw data (called "Measured Voltage") and the superconducting signal detecting superconductivity for a CSH sample [1] at several pressures were reported [13]. We focus on the data for

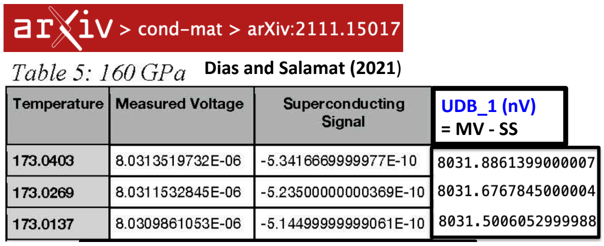

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Table 5: 160 GPa **Dias and Salamat (2021)**

Temperature	Measured Voltage	Superconducting Signal	UDB_1 (nV) = MV - SS
173.0403	8.0313519732E-06	-5.3416669999977E-10	8031.8861399000007
173.0269	8.0311532845E-06	-5.23500000000369E-10	8031.6767845000004
173.0137	8.0309861053E-06	-5.14499999999061E-10	8031.5006052999988

FIG. 2: A portion of Table V of Ref. [6] giving the "Measured Voltage" and "Superconducting Signal" (in V) for 160 GPa, supplemented with the corresponding entries for the background signal UDB_1 (in nV) (third column), obtained by subtracting the second from the first column.

160 GPa, which show remarkable features [14–16]. From those data, the point in Fig. 1 indicated by the arrow resulted. An image of the first lines of the table reporting those data in Ref. [6] is shown in Fig. 2. The background signal that was subtracted from the raw data to yield the superconducting signal was called UDB_1 (standing for "user defined background 1") in Ref. [17]. The numerical values of UDB_1 can be inferred from the data given in Table V of Ref. [6] by simple subtraction, as shown in Fig. 2, 4th column.

In this Comment we show that the quantities reported as "Measured Voltage" for 160 GPa, from which the point in Fig. 1 towards which the arrow is pointing was obtained, can be obtained through a mathematical calculation, hence provide no information on physical properties of the sample.

II. BACKGROUND SIGNAL UDB_1

Contrary to the standard procedure [7–12], the background signal UDB_1 was not obtained from a separate measurement. Figure 3 shows Fig. 2 from Ref. [17] that the authors used to explain how they constructed UDB_1. UDB_1 is the blue line near the top in Fig. 3 (b). Ref. [17] states *"the background can be approximated as linear in the region of the transition, and the susceptibility of the sample extracted after the background signal is subtracted from the raw data"*. And *"We use the temperature dependence of the measured voltage above and below the T_c of each pressure measurement and scale to determine a user defined background (Fig. 2a)... the subtracted background isolates the signal due to the sample."* and *"we use the profiles from the same dataset, before and after the superconducting transition to generate a user defined background profile"* and *"The user defined background for subtraction is qualitative in nature and does not represent a physical quantity"*.

The *"region of the transition"* is the region enclosed in the oval in Fig. 3a, bounded by the vertical dotted lines. Quantitatively, it is defined in Ref. [17] as the temperature range $T_a \leq T \leq T_b$, with $T_a = 169.5824K$, $T_b = 170.311K$. It is apparent from Fig. 3b that in that region the background is approximately linear, however it is not a straight line but has noise superimposed: Fig. 4 shows an amplified image. Ref. [17] does not explain how that noise was generated.

We asked the lead author of Ref. [17], also an author of Ref. [2], to clarify how the background signal UDB_1 in the transition region was obtained, given that it shows noise, in contradiction to the statement in Ref. [17] that it can be approximated as linear in the region of the transition. The response was [18] that it was interpolated, based on how it behaves in the upper and lower branches. We asked specifically whether the data from the signal itself (red curves in Figs. 3 and 4) were used in order to construct the background signal in the region of the transition where the signal is changing rapidly. The response was that they were not used [18]. This is consistent with the statements in Ref. [17] that only the part of the signal in the regions highlighted in blue in the left panel of Fig. 3, which do not include the transition region $169.5824K < T < 170.311K$, were used in constructing the "user defined background" UDB_1.

In the following we show that the "Measured Voltage" in the transition region can be *calculated*, without doing any measurement, starting from the background signal UDB_1 in the transition region.

III. OBTENTION OF MEASURED VOLTAGE STARTING FROM UDB_1

It has been shown in Refs. [15, 16] that the reported superconducting signal for 160 GPa [6] can be decomposed as

$$Supercond.\ Signal(T) = q(T) + P(T) \tag{2}$$

where $q(T)$ was called the "quantized component", and $P(T)$ the "unwrapped curve". The "quantized component" is a sequence of discrete jumps, all being integer multiples of the quantity 0.16555. Mathematically it is given by

$$q(T_{j+1}) = q(T_j) + 1.6555\Delta n_j. \tag{3}$$

Here, T_j is the $j-th$ value of the temperature, $1 \leq j \leq 438$, given in Table V of Ref. [6], with T_j decreasing as j increases, $T_1 = 173.0403K$, $T_{438} = 166.9137$, and Δn_j are integers ranging from -4 to 0.

What determines *uniquely* the quantity 0.16555 and the values of the Δn_j's? The condition that upon subtracting $q(T)$ from the reported superconducting signal, a *perfectly continuous* curve $P(T)$ results, the "unwrapped component". Any choices other than 0.16555 for the elementary jump and for the particular set of Δn_j found will *not* yield a perfectly continuous $P(T)$ [19]. The term "unwrapped" refers to the fact that the continuous curve $P(T)$ is hidden in the superconducting signal and only becomes visible after subtraction of the quantified component.

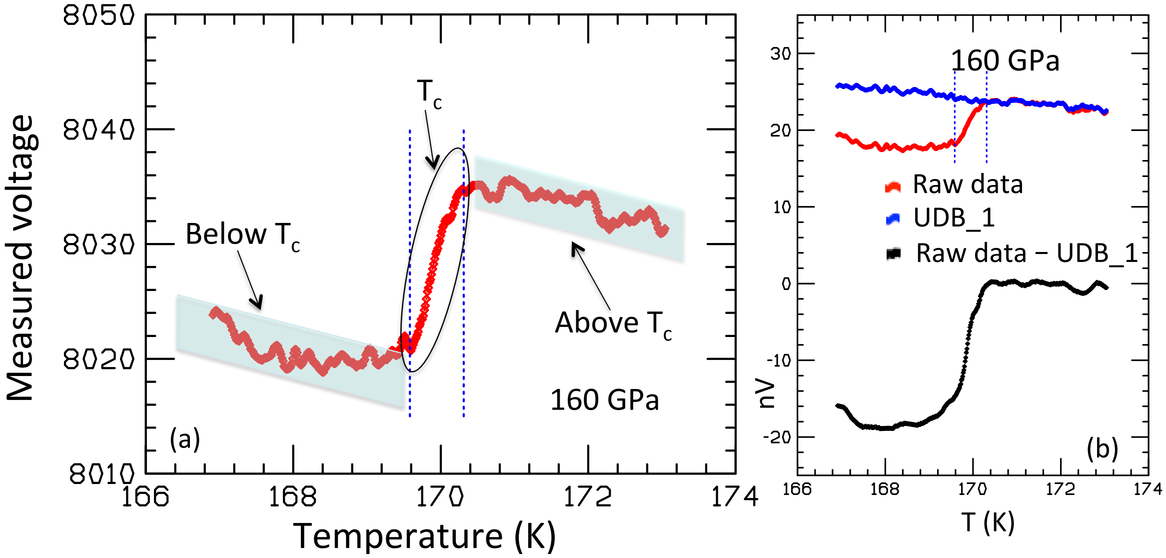

FIG. 3: Figure 2 of Ref. [17] (redrawn using the data of Ref. [6] Table V). We added the vertical dotted lines indicating the transition region. The caption in Ref. [17] reads: *"AC susceptibility data. (a) Raw data measured at 160 GPa. The profile of the regions highlighted in blue are used as part of the UDB_1. (b) Measured voltage from the susceptibility measurement explained in experimental details section in Refs. 1 and 2 for 160 GPa. Raw data (red), UDB_1 (blue) and raw data - UDB_1 (black)."* (Note that Refs. 1 and 2 in the caption are Refs. [1] and [6] here.)

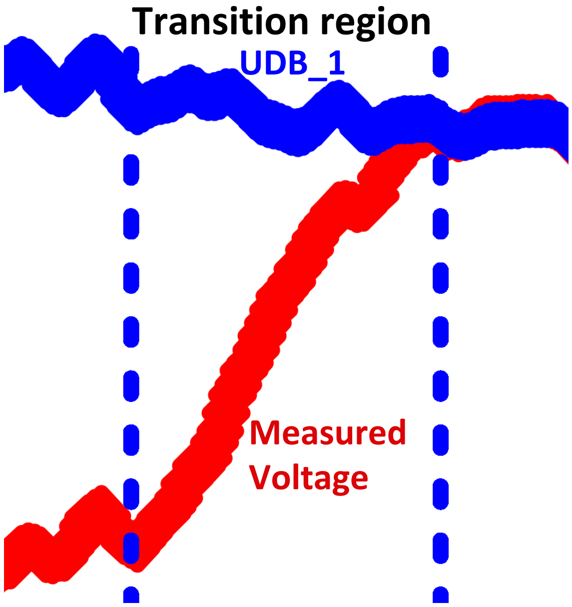

FIG. 4: Amplification of a portion of the upper part of Fig. 3 (b), showing the background signal in blue (UDB_1) and the raw data (Measured Voltage) in red in the transition region bounded by the vertical dashed lines.

Furthermore, it was found [16] that $P(T)$ can be accurately represented by a set of 14 third degree polynomials, with coefficients $a(n), b(n), c(n), d(n))$ given in Table I of Ref. [16].

From Eqs. (1) and (2), a 'non-measured voltage" NMV can be constructed as

$$NMV(T) = UDB\_1(T) + q(T) + P(T) \tag{4}$$

for the transition region $T_a \leq T \leq T_b$, which comprises 53 temperature values. Recall that according to Refs. [17] and [18] the measured voltage in the transition region was not used to construct UDB_1, so there is no reason to expect that the 53 numerical values of $NMV(T)$ in that region would have any relation with the measured voltage values.

Fig. 5 shows the results for NMV compared with the "Measured Voltage" (called $\chi_{mv}(T)$) given in Table V of Ref. [6], in the transition region $T_a \leq T \leq T_b$. The results are indistinguishable from one another for each of the 53 points. In other words, the right-hand side of Eq. (4) that supposedly contains no information on the measured voltage in the transition region [17, 18] reproduces perfectly the measured voltage in the transition region, whose drop of about $15nV$ was interpreted as signaling a superconducting transition in the sample in Refs. [1, 6, 17]. The inset in Fig. 5 shows the difference between $NMV(T)$ and $\chi_{mv}(T)$ in the transition region. They differ on average by $6.6 \times 10^{-5}nV$ for the 53 data points in the transition region, the maximum difference is $2 \times 10^{-4}nV$. In other words, they coincide to 8 significant figures.

As discussed in Appendix A, the amount of information needed to construct $NMV(T)$ using Eq. (4) in the transition region can be represented by approximately 86 digits. The calculated $NMV(T)$ reproduces the measured voltage for 53 temperatures to 8 digit accuracy, which is information contained in 424 (=53x8) digits.

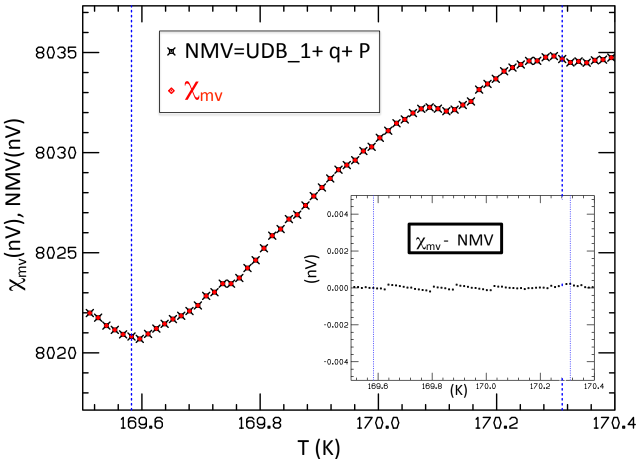

FIG. 5: Comparison of “non-measured voltage” NMV obtained from Eq. (4) with the “Measured Voltage” χ_{mv} obtained from the data in table V of Ref. [6], in the transition region between the dotted blue lines. They are indistinguishable on the scale of this figure. The inset shows the difference of the two quantities.

This establishes beyond a shadow of a doubt that the “Measured Voltage” was not measured but instead calculated using Eq. (4).

IV. CONCLUSION

Refs. [17] and [18] stated that the superconducting signal for ac susceptibility used to construct the point highlighted by the arrow in Fig. 1 (Fig. S14 of Ref. [2]) was obtained from a measured voltage and a constructed background signal UDB_1, with UDB_1 in the transition region having been constructed completely independently of the measured voltage in the transition region.

We have shown here that that statement is contradicted by facts. The measured voltage in the transition region can be obtained quantitatively to 8 digits accuracy through a mathematical calculation starting from UDB_1 through Eq. (4), hence without measuring a voltage. Hence it gives no information about the physical system, and in particular gives no information about superconductivity in the physical system. How the so-called “background signal” UDB_1 was constructed is unknown, its structure is consistent with it being the result of a measurement [16]. It shows no hint of a superconducting transition, hence provides no basis for drawing the point in Fig. 1 highlighted by the arrow.

In summary, according to the analysis in this paper, the probability that the voltages reported as “Measured Voltage” [6], or “raw data”, in the transition region for 160 GPa, from which the point indicated by the arrow in Fig. 1 (Fig. S14 of Ref. [2]) resulted, are truly measured voltages, is smaller than 10^{-338} (338=424-86). The authors continue to assert that those voltages are measured [4, 5]. This undermines the credibility of all the experimental data reportedly measured in Ref. [2]. Moreover, the extended analysis of Ref. [16] indicates that in fact none of the reported “Measured Voltage” data for any of the pressures for which ac susceptibility results for CSH were reported [6], with inferred transition temperatures shown by the six red points in Fig. 1 (Fig. S14 of Ref. [2]), was actually measured.

In conclusion, there are two separate issues raised by the analysis in this paper. One is, whether CSH is or is not a high temperature superconductor. We argue that the analysis in this paper, together with the analysis in Refs. [16, 20, 21], establish that there is currently no evidence supporting the claim that it is. To support that claim, new evidence by the authors of [1] or by other groups is needed. The second issue is whether there could be an alternative explanation to the analysis in this paper that indicates that the quantities reported as “Measured Voltage” underlying the susceptibility results in Fig. 1 were not measured in an experiment. If there is, it should be provided by the authors of Ref. [2] and/or Ref. [1]. Namely, they should describe the procedure by which the 53 values of UDB_1 in the transition region were obtained, and more generally the procedure by which the entire UDB_1 was constructed for the 438 temperature values in the measurement interval reported. The description should be with sufficient detail so that readers can themselves reproduce it.

Acknowledgments

I am grateful to the organizers of the International Workshop “Challenges in Designing Room Temperature Superconductors”, (CDRTS 2022), L’Aquila, Italy. 26-29 July 2022, for the opportunity to attend the meeting and ask questions to the speakers. This work was performed in collaboration with D. van der Marel.

Conflicts of interest: There are no conflicts of interest to declare

Appendix A: Information needed to construct $q(T)$ and $P(T)$ in the transition region

The values of the temperature (438 values in total) are given in the first column of Table V of Ref. [6], and in Ref. [13]. In order of decreasing temperature T_j, the transition region (in K) goes from $T_{201} = 170.3110$ to $T_{253} = 169.5824$. These are 53 temperature values. To construct $q(T_j)$ in that range we need the starting value $q(T_{201}) = -2.81435$ and 52 integers, that are given by 0,-1,-1, 0,-1,-1,-1,-2,-1,-1,-3,-1,-2,-2,-1,-1,-1,-1,-1, 0,-1,-1,-2,-1,-3,-2,-2,-3,-3,-3,-4,-4,-3,-4,-3,-4,-3,-2,-3,-2,-1,-3,-1,-2,-1,-1,-1,-1,-1,-1,-1, 0.

TABLE I: Coefficients of Eq. (A1) for $P(T)$ at 160 GPa in the transition region (from Ref. [16]).

n	$T(n)$	a_n	b_n	c_n	d_n
9	170.9066	2.04588	−1.86481	−2.15322	−1.00999
10	170.2829	2.6164	−0.352704	−0.0984839	−0.126134

There are 14 segments to $P(T)$ over the entire temperature range [16]. Almost all the temperatures in the transition region, from $T_{203} = 170.2829$ to T_{253}, i.e 51 temperature values, are in the 10th segment of $P(T)$. The first three temperature values, from T_{201} to T_{203} are in the 9-th segment. The segments are third order polynomials of the form

$$P(T) = a_n + b_n(T - T(n)) + c_n(T - T(n))^2 + d_n(T - T(n))^3. \tag{A1}$$

The values of $T(n)$ corresponding to the beginning of the segments needed are $T(n = 9) = T_{158} = 170.9066$ and $T(n = 10) = T_{203} = 170.2829$. The values of the coefficients for these segments are given in Table I.

Note that the information conveyed in the above quantities can be expressed by not more than 86 digits (26 for Δn_j's, 6 for $q(T_{201})$, 3 and 3 for the j values 158 and 203, 48 for the coefficients in Table I). With this information, the T_j values and the UDB_1 values (which are independent of χ_{mv}) we construct 53 values of NMV(T), eq. (4), that coincide with the reportedly measured 53 voltage values of $\chi_{mv}(T)$ to 8 digits. To express those values we need 53x8=424 digits. The fact that 86 << 424 shows unequivocally that $\chi_{mv}(T)$ was not measured but instead was constructed in this way.

Appendix B: Numerical values of measured voltages and calculated voltages

With the information given in Appendix A and the numerical data from Table V of Ref. [6], also given in Ref. [13], readers can easily calculate themselves $NMV(T)$ and verify that it coincides with the reported "Measured Voltage"to 8 digits. We provide the numbers in table II below and explain the calculation in detail here.

[1] "Measured Voltage" (MV), 7th column of Table II, from Ref. [6] table V, 2nd column.

[2] "Superconducting Signal" (SS), 5th column of Table II, from Ref. [6] table V, 3rd column.

[3] UDB_1, 6th column of Table II, obtained by subtracting SS from MV.

[4] To obtain the non-measured voltage NMV, 8th column in Table II, for the j-th row:

(i) Calculate

$$q(T_j) = n_j \times 0.16555, \tag{B1}$$

with n_j given in the 2nd column of Table II.

(ii) Calculate $P(T_j)$ using Eq. (A1), with the parameters in the first line of Table I for $j = 201$ to $j = 203$, or with the parameters in the second line of Table I for $j = 203$ to $j = 253$.

(iii) Calculate $NMV(T_j) = UDB\_1(T_j) + q(T_j) + P(T_j)$. It gives the value in the j-th row in the 8th column of Table II.

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[19] See Appendix C of Ref. [15].

[20] J. J. Hamlin, “Vector graphics extraction and analysis of electrical resistance data in Nature volume 586, pages 373-377 (2020)”, arXiv:2210.10766 (2022).

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TABLE II: Columns 4, 5 and 7 give temperature, “Superconducting Signal” and “Measured Voltage” in the transition region taken from Table V of Ref. [6] columns 1, 3 and 2 respectively, lines 201 to 253 in the table. Column 6 gives background UDB_1 computed by subtracting column 5 (SS) from column 7 (MV). Column 2 gives the integers needed to compute $q(T_j)$, Eq. (B1), column 3 gives the increments $\Delta n_j = n_{j+1} - n_j$. Column 8 gives the Calculated (Nonmeasured) Voltage NMV(T) from Eq. (4). Note that the last two columns of the table (Measured Voltage and Nonmeasured (Calculated) Voltage) agree to within 0.0001 nV (8 (occasionally 7) digits).

line in list (j)	n_j	Δn_j	Temperature (K)	Supercond. Signal (nV)	UDB_1 (nV) =MV-SS	Measured Voltage (nV)	NMV (nV) (Calculated V)
201	-17	0	170.3110	-0.2080000	8034.8811792	8034.6731792	8034.6729516
202	-17	-1	170.2970	-0.2028333	8035.0207468	8034.8179135	8034.8177001
203	-18	-1	170.2829	-0.3635000	8035.1248506	8034.7613506	8034.7613505
204	-19	0	170.2688	-0.5240000	8035.1105503	8034.5865503	8034.5864448
205	-19	-1	170.2548	-0.5191667	8035.1002594	8034.5810927	8034.5810256
206	-20	-1	170.2409	-0.6798333	8035.0796619	8034.3998286	8034.3996837
207	-21	-1	170.2268	-0.8406667	8035.0865330	8034.2458663	8034.2458496
208	-22	-2	170.2126	-1.0013333	8035.0647955	8034.0634622	8034.0634129
209	-24	-1	170.1986	-1.3276667	8035.0121698	8033.6845031	8033.6844438
210	-25	-1	170.1846	-1.4885000	8034.9166430	8033.4281430	8033.4280998
211	-26	-3	170.1705	-1.6493333	8034.7976650	8033.1483317	8033.1483165
212	-29	-1	170.1566	-2.1413333	8034.6975628	8032.5562295	8032.5562221
213	-30	-2	170.1428	-2.3023334	8034.6786871	8032.3763537	8032.3764034
214	-32	-2	170.1289	-2.6287666	8034.7753537	8032.1465871	8032.1465948
215	-34	-1	170.1149	-2.9552500	8035.0264701	8032.0712201	8032.0712556
216	-35	-1	170.1008	-3.1161333	8035.3101886	8032.1940553	8032.1940898
217	-36	-1	170.0867	-3.2770334	8035.5372720	8032.2602386	8032.2602788
218	-37	-1	170.0726	-3.4379333	8035.6282232	8032.1902899	8032.1903278
219	-38	-1	170.0586	-3.5988500	8035.5755974	8031.9767474	8031.9767610
220	-39	0	170.0446	-3.7598167	8035.4182940	8031.6584773	8031.6585127
221	-39	-1	170.0306	-3.7552000	8035.2192374	8031.4640374	8031.4640594
222	-40	-1	170.0166	-3.9164000	8035.0093081	8031.0929081	8031.0929086
223	-41	-2	170.0025	-4.0773333	8034.8153930	8030.7380597	8030.7380821
224	-43	-1	169.9886	-4.4038334	8034.6935613	8030.2897279	8030.2897272
225	-44	-3	169.9746	-4.5647666	8034.6437971	8030.0790305	8030.0790289
226	-47	-2	169.9607	-5.0568166	8034.6769732	8029.6201566	8029.6201460
227	-49	-2	169.9466	-5.3832667	8034.7622012	8029.3789345	8029.3789412
228	-51	-3	169.9325	-5.7096666	8034.8560062	8029.1463396	8029.1463257
229	-54	-3	169.9185	-6.2016500	8034.9103537	8028.7087037	8028.7086837
230	-57	-3	169.9046	-6.6936667	8034.9549685	8028.2613018	8028.2612916
231	-60	-4	169.8908	-7.1856667	8035.0121698	8027.8265031	8027.8264702
232	-64	-4	169.8768	-7.8434500	8035.2026493	8027.3591993	8027.3591889
233	-68	-3	169.8627	-8.5009000	8035.3994182	8026.8985182	8026.8985280
234	-71	-4	169.8487	-8.9927666	8035.6694103	8026.6766437	8026.6766306
235	-75	-3	169.8346	-9.6501500	8035.8352908	8026.1851408	8026.1851319
236	-78	-4	169.8205	-10.1419666	8035.9943081	8025.8523415	8025.8523484
237	-82	-3	169.8065	-10.7993166	8036.0206210	8025.2213044	8025.2213062
238	-85	-2	169.7925	-11.2913833	8035.9165172	8024.6251339	8024.6251543
239	-87	-3	169.7786	-11.6175833	8035.8547405	8024.2371572	8024.2371534
240	-90	-2	169.7647	-12.1093333	8035.8473034	8023.7379701	8023.7379775
241	-92	-1	169.7507	-12.4354500	8035.8896305	8023.4541805	8023.4541890
242	-93	-3	169.7367	-12.5959833	8036.0509355	8023.4549522	8023.4549683
243	-96	-1	169.7228	-13.0875666	8036.1161476	8023.0285810	8023.0285603
244	-97	-2	169.7087	-13.2480000	8036.0909748	8022.8429748	8022.8429840
245	-99	-1	169.6947	-13.5739333	8035.9399685	8022.3660352	8022.3660338
246	-100	-1	169.6806	-13.7342334	8035.8238522	8022.0896188	8022.0896088
247	-101	-1	169.6665	-13.8944833	8035.7477751	8021.8532918	8021.8532737
248	-102	-1	169.6524	-14.0547000	8035.7397641	8021.6850641	8021.6850573
249	-103	-1	169.6385	-14.2149334	8035.6625393	8021.4476059	8021.4476048
250	-104	-1	169.6245	-14.3753500	8035.5898977	8021.2145477	8021.2145545
251	-105	-1	169.6105	-14.5354000	8035.4812185	8020.9458185	8020.9457995
252	-106	0	169.5964	-14.6954000	8035.3965566	8020.7011566	8020.7011614
253	-106	-1	169.5824	-14.6898000	8035.5080974	8020.8182974	8020.8182986