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by

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INTRODUCTION

The lunar regolith has recorded the impingent solar corpuscular radiation since billions of years and thus provides, apart from the solar gas rich meteorites, the only possibility to study these characteristics of the sun in time. This record is difficult to read because it has been affected by secondary effects such as loss of trapped solar gases by diffusion, by sputtering and during agglutination. Among the various constituents in the lunar soil, mineral grains have a relatively short lifetime in a regolith surface layer with regard to destruction by meteorite impact. Therefore, their exposure history is comparatively simple and their record of solar wind and solar flare irradiation effects is the least distorted. In consequence of the short lifetime, minerals permit a comparatively high time resolution for changes in characteristics of the solar radiation, i.e. the antiquity of mineral grains is better defined than that of bulk samples. The term "antiquity" (cf. Kerridge, 1980) stands for a measure of how long ago a sample was exposed to the solar wind and the solar flares.

With the investigation reported here we attempt primarily to explore the mean particle flux in solar flares (SF) relative to that in the solar wind (SW) as a function of time over the past few billion years. An indication for a higher SF/SW ratio in the past was found on the basis of a comparatively high value of the SF-track density/SW-Ar concentration ratio in plagioclase crystals from the Apollo 15 deep drill core (Wieler et al., 1981). Recently, new information on solar flare particles in lunar samples became available in the form of solar flare implanted noble gases. (Yaniv and Marti, 1981; Etique et al., 1981; Nautiyal et al., 1982). In the latter two of these reports, SF-Ne with a $^{20}Ne/^{22}Ne$ ratio of $\leqslant 11.3$ and of 11.8 ± 0.3

respectively, was found in plagioclase grains which had been etched in order to remove surficially trapped SW-Ne. These values are roughly 10% lower than the respective ratio of $\sim$ 13.0 of SW-Ne retained in lunar ilmenites (e.g. Eberhardt et al., 1970; Signer et al., 1977). By the solar wind composition experiments a ^{20}Ne/^{22}Ne ratio of 13.7 ± 0.3 in the present day solar wind was determined (Geiss, 1973). The difference between this value and the ratio found in ilmenites strongly indicates that the solar Ne retained in lunar minerals is isotopically fractionated as a result of gas losses. However, the fact that the ^{20}Ne/^{22}Ne ratio of SF-Ne is low compared to the corresponding SW values cannot be explained by a fractionation after the implantation, because we will show here that SF-Ne is better retained than SW-Ne.//

P In this situation, we initiated a study of the elemental and isotopic composition of He, Ne, and Ar in mineral separates from additional regolith samples with high antiquity, to search for a possible SF-Ne contribution. By comparing these data with those obtained in previous studies, which had been carried out predominantly on recently irradiated surface soil minerals, the hypothesis of a variation of the average SF/SW intensity ratio with time is now further tested. In addition, the solar flare track densities, as an independent measure of the solar flare dose, were determined in several of the samples.

Because of limitations in the amounts of sample available, etching experiments on drill core soils were not feasible. The presence of SF-Ne had therefore to be inferred indirectly. This is possible, because a SF-Ne contribution that amounts to a significant fraction of the retained SW-Ne, will lower the ^{20}Ne/^{22}Ne ratio of the trapped solar gases in a sample. We assume thereby that the ^{20}Ne/^{22}Ne ratio of SF-Ne has been constant with time. In favor of this assumption speaks the fact that a secular variation of the same ratio in the solar wind is $< 2\%$ during the past 2-3 Ga., as we show in this paper. A constant isotopic composition of trapped SF-Ne in all lunar minerals does not necessarily contradict to the low $^{20}Ne/^{22}Ne$ ratios of $\sim 7.6 \pm 2$ detected by satellite borne instruments in a few contemporary flares (Dietrich and Simpson, 1979; Mewaldt et al., 1979) because SF particles measured by these experiments have considerably higher energies than those detectable in lunar samples and because the integration times of the two types of detectors differ many orders of magnitude. Moreover, we show below that the interpretation of the neon isotopic data which is based on the assumption of a constant composition of SF-Ne agrees with the conclusions inferred from the solar flare track data. The $^{20}Ne/^{22}Ne$ ratio in the solar wind as a function of time is studied by investigating the solar gases in the minerals ilmenite, olivine and pyroxene. All these minerals retain SW-Ne much better than plagioclase and a SF-Ne contribution is thus expected to be masked by the retained SW-Ne. The isotopic composition of Ne in the ancient solar wind is of interest also in the context of the large variations of the composition of trapped nitrogen in lunar soils (e.g. Kerridge et al., 1977; Clayton and Thiemens, 1980).

In the following discussion we will first present evidence that plagioclases from the lowest part of the Apollo 16 deep drill core - belonging to the earliest irradiated samples among the available lunar material - contain a significant SF-Ne contribution. In a next section, we will argue that SF-Ne is also detectable in plagioclase separates from other soils. To corroborate this claim, we then study the SW-Ne composition as a function of time. Finally, a temporal variation of the SF/SW intensity ratio is deduced from the Ne data of the plagioclases and independently from the SF track measurements.

SAMPLES, EXPERIMENTAL PROCEDURE AND RESULTS

Sample Preparation

Samples analyzed were predominantly mineral separates, a few agglutinate and glass separates, and some bulk soils. All separates were prepared by handpicking. Most data published in this work stem from samples that have been shielded from the solar radiation in recent times. These samples originate from deep drill core sections 60002 and 60003, from core sections 60009 and 60010, and from regolith breccias 79035 and 79135.

From each of the Apollo 16 deep drill core sections we investigated 5 samples from adjacent, about 5 mm thick layers (parent samples 60002,89; 92; 94; 96; 98 and 60003,138; 140; 143; 145; 147, respectively). The former belong to Major Petrology Unit A, the latter to Unit B in the classification of Vaniman et al. (1976). According to Allton et al. (1981), our 60002 samples were located about 192 - 195 cm and the 60003 samples about 151.5 - 154 cm below the regolith surface. From each of the ten sampled layers, a plagioclase separate in the grain size range 100 - 150 µm was prepared. In order to obtain sufficient noble gas amounts, the plagioclase grains of 150 - 200 µm size were combined into 2 samples for Unit A and into 1 sample for Unit B. Out of each Unit two pyroxene samples were prepared, one in the size range 100 - 150 µm and one in the range 150 - 200 µm.

Samples 60009,3089 and 60010,3008 were taken from 48 cm and 24,5 cm, respectively, below the regolith surface (Fruland et al., 1982). From each soil, a plagiocclase separate in the grain size range 150 - 200 µm was measured.

In contrast to the very friable regolith breccia 79035, breccia 79135 is rather coherent. In the crushing process of this sample, the original grain size of the minerals probably was not conserved. Plagioclases, pyroxenes, and from breccia 79035 also ilmenites, were separated.

Noble Gas Determinations

The He, Ne, and Ar results are listed in table 1. Included are the data of a few newly studied surface soil separates. The analytical procedures applied were described by Signer et al. (1977). The gas concentrations and isotope ratios are corrected for extraction blanks and gases released from the Al foils used to contain the samples. Typical values for these corrections are (in units of 10^{-10} cm^3 STP):

^{4}He : 80, ^{20}Ne : 2.2, ^{22}Ne : 0.4

^{36}Ar : 0.4, ^{38}Ar : 0.1, ^{40}Ar : 30 ± 15

These corrections are < 4 % of the amounts of gas in the sample, except for ^{40}Ar, where the corrections are generally < 10%, but may be as high as 30% for mineral separates with weights < 0.3 mg. For one sample set, the CO_2 background in the spectrometer was about 5 times higher than normally. For these samples, the blank corrections on m/e = 22 ranged between 8 and 20 %. The signal on m/e = 44 differed less than 6% between sample and corresponding blank runs, such that the respective additional corrections on the ^{22}Ne signal remained < 1.2%. The precision of the ^{20}Ne/^{22}Ne ratio of these samples is estimated in the Appendix to be around 1.5%.

^{4}He, ^{20}Ne, and ^{36}Ar concentrations reported in table 1 are accurate within 5 - 10%. The precision of the data is estimated as follows:

^{4}He; ^{20}Ne; ^{36}Ar : 2 %
^{4}He/^{3}He : 5 % (for ^{3}He < 100 . 10^{-8} cm^{3} STP/g)
1 % (for ^{3}He > 100 . 10^{-8} cm^{3} STP/g)
^{20}Ne/^{22}Ne; ^{21}Ne/^{22}Ne : 1 %
^{36}Ar/^{38}Ar : 0.5%
^{40}Ar/^{36}Ar : <20% (for sample weight < 0.3 mg)
5 % (for sample weight > 0.3 mg)

In Figures 3, 4, and 5 the calculated ^{20}Ne/^{22}Ne ratios of the solar gas retained in each sample are given (in the following called "(^{20}Ne/^{22}Ne)$_{sol}$)").For the evaluation, it was assumed that the Ne detected is a mixture of solar and spallogenic gas, whereby constant, but for each mineral species distinct isotopic compositions of the spallogenic component were selected. The values adopted were determined by Lugmair et al. (1976) on mineral separates of lunar rock 76535:

plagioclases: ^{20}Ne : ^{21}Ne : ^{22}Ne = 0.76 : 0.768 : 1
olivines, pyroxenes: ^{20}Ne : ^{21}Ne : ^{22}Ne = 0.84 : 0.94 : 1

The uncertainties of the (^{20}Ne/^{22}Ne)$_{sol}$ ratios are discussed in the Appendix.

We include in the following discussion data of mineral separates of all lunar soils measured in our laboratory, provided their solar gas concentrations were large compared to the spallogenic background. To be consistent, only samples in the 150 - 200 μm grain size range, complemented in some cases by 100 - 150 μm sized separates, are taken into account. The samples of soil 14163 (Signer et al., 1977) are omitted, because they were among the very first separates prepared and are, by present day standards, severely contaminated. Results presentec in the figures but not shown in table 1 were published earlier. (Frick et al., 1975; Signer et al., 1977; Wieler et al., 1981).

Track Determinations

The average of the track density in the centers of some 50 plagioclase grains (Mean central Track Density, MTD) was determined for 5 surface soils and 3 drill core soils. Whenever possible, aliquots of the plagioclases used for the noble gas analyses were investigated. The results are given in table 2, together with the percentage of track rich grains (% TRG), i.e. the fraction of grains with central track densities > 10^8 tracks per cm^2.

The tracks were revealed progressively by etching of the polished plagioclase grains between 2 and 6 minutes in a boiling solution of 40 cm^3 H_2O and 30 g NaOH. The grains were photographed by a scanning electron microscope at a magnification of 20'000. Because the track density within lunar soil grains is rather homogeneous at depths > 10 μm (Poupeau et al., 1975), the actual shape of the grain and the location of the area on the polished grain where the tracks are counted do not affect the results. The precision of the mean track densities is about 20%, including a possible systematic error arising from the fact that two different SEMs were used. A cross-calibration on one sample showed that this error is < 10%. From sample 60002,89-98, seven out of 53 grains were overetched after the first etching step, compared to 1 - 3 overetched grains out of about 50 in all other samples. Some of the overetched grains probably had high track densities, such that the true MTD of 60002,89 - 98 may be higher than the value given. This does, however, not affect our conclusions.

SOLAR FLARE NEON IN THE APOLLO 16 DEEP DRILL CORE

Vaniman et al. (1976) recognized four "Major Petrology Units" in this core, which were labelled A - D. In the lowest Unit A, the $^{20}Ne/^{36}Ar$ and $^{40}Ar/^{36}Ar$ ratios are considerably higher than in the rest of the core (Bogard and Hirsch, 1975; Heymann et al., 1978). Furthermore, I_S/FeO values and track densities indicate that this Unit is less mature than the overlaying soils (Gose and Morris, 1977; Blanford and Wood, 1978). Bogard and Hirsch (1975) postulate that Unit A was exposed to the solar radiation 0.5 - 1.5 Ga. ago, and concluded from their data that the Ne/Ar ratio in the solar wind was higher at that time than more recently. Based on this information we included samples from Units A and B in our search for SF implanted Ne and the studies of possible variations with time of the element and isotope ratios of the SW noble gases. In contrast to Bogard and Hirsch (1975), we thereby investigated mineral separates rather than bulk samples.

Our bulk soil data of 60002 (Unit A) and 60003 (Unit B) in table 1 agree well with the values given by Bogard and Hirsch (1975) and Heymann et al. (1978). In particular, the difference of the $^{20}Ne/^{36}Ar$ and the $^{40}Ar/^{36}Ar$ ratios between the two Units is confirmed. Furthermore, the concentrations of spallogenic ^{21}Ne and ^{38}Ar of the bulk soil as well as of all mineral separates in Unit A are roughly two times higher than the corresponding values in Unit B. Thus, the largely different irradiation histories of the two Units are confirmed.

For a detailed analysis of the Ne in the mineral separates, we turn to the three isotope correlation plot shown in Figure 1. The data points of the plagioclases from each Unit form a linear array, with a correlation coefficient > .99 for both cases. Taking the data arrays as mixing lines of two components, the solar ^{20}Ne/^{22}Ne and the spallogenic ^{21}Ne/^{22}Ne ratios have the values given in Figure 1. For the computation we assumed $(^{21}\mathrm{Ne}/^{22}\mathrm{Ne})_{sol} = .0312$ and $(^{20}\mathrm{Ne}/^{22}\mathrm{Ne})_{spal} = .76$. Uncertainties in these assumed ratios do not considerably influence the errors of the ratios indicated in Figure 1. In the Appendix, the two component interpretation of the figure is discussed in more detail. The ^{21}Ne/^{22}Ne ratios of the spallogenic component in plagioclases from both Units are close to the value of .768 given by Lugmair et al. (1976). Remarkable is the difference of the ^{20}Ne/^{22}Ne ratios of the solar component in the plagioclase samples from the two Units. In fact, plagioclases from Unit A have the lowest $(^{20}\mathrm{Ne}/^{22}\mathrm{Ne})_{sol}$ of all samples discussed in this paper. In contrast to plagioclase, pyroxenes from both core sections contain solar Ne with a similar isotopic composition, which is best illustrated by the two pyroxene data points that overlap in Figure 1.

We propose to explain the offset of the two mixing lines in Figure 1 by a comparatively large admixture of SF-Ne with a low ^{20}Ne/^{22}Ne ratio in the plagioclases of section 60002. In addition to the plagioclase data, this explains also the similar composition of the solar Ne in the pyroxenes from both Units. Underlaying this proposal is the hypothesis that SW-Ar is retained to at least 70% in minerals of the lunar regolith. The arguments for this claim are discussed by Signer et al. (1977) and by Wieler et al. (1981). Assuming SW-Ar to be well retained, the ^{20}Ne/^{36}Ar ratios of trapped solar gases show that plagioclase retains only some 3-5% of the SW-Ne, whereas olivine and pyroxene have a retentivity for this gas between about 15 and 60 %. SF-Ne in plagioclase, on the other hand, is expected to be much better retained than SW-Ne, due to its larger implantation depth. We show in the section "Isotopic composition of SF-Ne" that the ratio between the retentivities of SF-Ne and SW-Ne is considerably larger for plagioclase than for olivine and pyroxene. Therefore, a SF-Ne contribution becomes more prominent in the solar ^{20}Ne/^{22}Ne ratio of the plagioclase. Our view that all minerals retain SW-Ar well has been challenged by Frick and Pepin (1983) who deduce a SW-Ar retentivity in regolith minerals of some 10% only, whereas SW-Ne is retained to as few as 3% in ilmenite and pyroxene. The authors base their conclusions on stepwise degassing experiments on mg sized mineral separates whereby noble gases and nitrogen were measured simultaneously. This is clearly an important experimental breakthrough. Nevertheless, the correlation of the N and Ar data used to deduce the low retentivities of SW-Ar is, in our view, not beyond doubt, especially because in the experiments reported, combustion and pyrolysis steps followed each other in variable sequences and because different time/temperature schedules were used. The discrepancy concerning N and Ar concentrations released from ilmenites shows, as the authors state themselves, that more work is needed on this topic. For the time being, we consider therefore our arguments supporting the claim that SW-Ar is well retained in lunar soil minerals to be more conclusive than Frick and Pepins reasoning and we adhere to this claim.

The interpretation of Figure 1 given above requires that an admixture of SF-Ne large enough to influence the $^{20}Ne/^{22}Ne$ ratio of the solar Ne is also noticeable in the measured $^{20}Ne/^{36}Ar$ ratio. Consequently, plagioclases from Unit A (core section 60002) ought to have a higher $^{20}Ne/^{36}Ar$ ratio than plagioclases from Unit B (Section 60003). Again, the pyroxenes are expected to show no such difference. The $^{4}He/^{36}Ar$ versus the $^{20}Ne/^{36}Ar$ ratios are presented in Figure 2. The plagioclase separates from section 60002 have indeed an about 40% higher mean abscissa value than those of section 60003, whereas the pyroxenes of both sections have similar $^{20}Ne/^{36}Ar$ ratios. The data for the bulk samples exhibit the same element abundance pattern as the plagioclases. Note, however, that we interpret the higher $^{20}Ne/^{36}Ar$ ratios in the bulk soils - and in the plagioclases - of section 60002 to reflect a large SF-Ne contribution and not, as proposed by Bogard and Hirsch (1975), to result from a different Ne/Ar ratio in the ancient solar wind. Such a difference would emerge more prominently in the SW-Ne retentive pyroxenes than in plagioclase.

Because even spallogenic ^{3}He is very poorly retained in plagioclase (cf. Frick et al., 1975), also SF-He in this mineral is expected to be retained much less efficiently than SF-Ne. In fact, the $^{4}He/^{36}Ar$ ratios of the plagioclases of Units A and B are remarkably similar. In an earlier paper (Wieler et al., 1981) we have shown that the $^{4}He/^{36}Ar$ ratios of the plagioclases of Apollo 16 soils are similar within about ± 30% and nearly independent of the SW-Ar concentrations. Together with the fact that less than half a percent of the solar He is retained, this may imply that the solar He retained in plagioclases resides in lattice sites with high activation energy (cf. Fechtig and

Kalbitzer, 1966) produced by the solar irradiation, causing the abundance of such sites to be proportional to the exposure time of the sample.

Summarizing this section, we postulate that a large fraction of the solar Ne retained in the plagioclases from Major Petrology Unit A of the Apollo 16 deep drill core is of solar flare origin. The ratio of the concentrations of retained SF-Ne and SW-Ne is large enough to be reflected in the measured $^{20}Ne/^{36}Ar$ ratio and in the $^{20}Ne/^{22}Ne$ ratio of the solar component. SF-Ne may also be present in Unit B, but here the SF-Ne/SW-Ne ratio is definitely lower that in Unit A. The question now arises, whether plagioclase crystals from other soils also show evidence of a SF-Ne contribution.

SF-Ne IN PLAGIOCLASE SEPARATES

Plagioclase separates of all soils studied are examined in this section for the presence of SF-Ne. As proposed above, this is done by investigating the correlation between $(^{20}Ne/^{22}Ne)_{sol}$ and $^{20}Ne/^{36}Ar$, shown in Figure 3. A mixture of SW and SF gases in variable proportions in different samples leads in this figure to a trend of decreasing ordinate values with increasing abscissa values. Important is to note, that element and isotope fractionation caused by diffusive loss of SW gases would decrease the $(^{20}Ne/^{22}Ne)_{sol}$ with decreasing $^{20}Ne/^{36}Ar$. Losses of solar He and Ne from regolith minerals, leading to isotopic fractionation effects, only occur when the grains reside in the uppermost regolith layer, where daytime temperatures are high. During the hundreds of million years of residence below the surface, the minerals do not loose appreciable amounts of solar noble gases (Wieler et al., 1979). It is therefore meaningful to compare element and isotope ratios of solar gases in early and recently irradiated samples. The Apollo 16 plagioclase separates are less affected by mineral impurities than plagioclases from mare samples, because SW-Ne retentive minerals are scarce in highland soils. The data points of these samples (a-m) indeed show the trend expected if SF-Ne is present besides the SW-Ne. The broken line in Figure 3 is a mixing line that may roughly account for the data pattern of the Apollo 16 separates and is discussed in more detail in the section "Isotopic composition of SF-Ne". Besides the trend observed for the highland samples, also the four mare plagioclase separates with the lowest $(^{20}Ne/^{22}Ne)_{sol}$ (p, r, w, x) have the highest $^{20}Ne/^{36}Ar$ ratios. The position of these points cannot be due to contamination, because the $(^{20}Ne/^{22}Ne)_{sol}$ ratios of the mafic minerals of the samples in consideration (see Figure 4) are higher than the respective ratios of the plagioclases. Altogether, Figure 3 supports the hypothesis that low $(^{20}Ne/^{22}Ne)_{sol}$ ratios in plagioclase indicate a considerable SF-Ne contribution.

A remarkable consequence of this interpretation concerns the $^{20}Ne/^{36}Ar$ ratio of the retained solar wind. Despite Ne losses of about 96%, this ratio appears to be constant within ± 20% for all Apollo 16 plagioclase separates, as can be estimated from the position of their data points along the mixing line in Figure 3. (A possible exception is the separate from core soil 60009,3089; data point labelled "c"). Actually, the constancy of the $^{20}Ne/^{36}Ar$ ratios is in accord with the small variations of the $^{4}He/^{36}Ar$ values in these samples, which all agree within ± 30%, as was noted in the preceeding section. It is conceivable that in plagioclases not only He, but also SW-Ne is retained predominantly in lattice sites with high activation energy created in number proportional to the exposure time. Due to the remarkably constant retentivity, the trapped SW-Ne apparently is nearly uniformly fractionated in all Apollo 16 plagioclase separates, which forms the basis to detect SF-Ne contributions in these samples.

Another interesting feature of Figure 3 emerges below (refer to discussion of Figure 5), since we show there that all plagioclases with $(^{20}Ne/^{22}Ne)_{sol} < 12.4$ acquired their solar gases predominantly in ancient times. This leads us to examine in the following section the possibility that a secular variation of the Ne isotopic composition and of the Ne/Ar ratio in the solar wind could be responsible for the trend in this figure.

$^{20}Ne/^{22}Ne$ and $^{20}Ne/^{36}Ar$ IN THE ANCIENT AND MODERN SOLAR WIND

Olivines and Pyroxenes

Among the minerals retentive for SW-Ne, olivine and pyroxene are, in contrast to ilmenite, abundant in all mare samples. Olivine and pyroxene grains from recently exposed soils and from

samples exposed in ancient times are thus most suitable to detect temporal variations of the Ne isotopic composition and the Ne/Ar ratio in the solar wind. The isotopic composition of Ne in the ancient solar wind is of particular interest in context with the large variations of the $^{15}N/^{14}N$ ratio of nitrogen trapped in lunar samples (e.g. Kerridge et al., 1977; Clayton and Thiemens, 1980). These authors explain the observed variations by a secular change of the isotopic composition of N in the solar wind, whereas Geiss and Bochsler (1982) invoke a planetary nitrogen component with a low $^{15}N/^{14}N$ ratio. Since at least one theory attempting to explain an increase of the $^{15}N/^{14}N$ ratio in the solar wind also demands a secular increase of its $^{20}Ne/^{22}Ne$ ratio (Ray and Heymann, 1980), investigations of Ne retained in early irradiated minerals may contribute to the understanding of the nitrogen data.

Figure 4 shows the data of the SW-Ne retentive minerals analogous to Figure 3. It is shown in the next section that the separates represented by open symbols have antiquities as high as 2 - 3 Ga. Except for trench soil 15421 (r), all these samples are from drill cores or soil breccias. The filled symbols stand for separates from surface soils, presumably all with antiquities on the order of 100 Ma. only. The spread of the olivine and pyroxene data points along the abscissa in Figure 4 is quite large and precludes firm conclusions about the Ne/Ar abundance ratio in the solar wind as a function of time. The data do however not support the view that the high $^{20}Ne/^{36}Ar$ ratios in the bulk soil and the plagioclases of core section 60002 are the result of a higher Ne/Ar ratio in the ancient solar wind as was postulated by Bogard and Hirsch (1975). (The data point for the pyroxene of 60002 is labeled "a"). The main reason for the spread are probably mineral impurities, since it is more difficult to prepare a clean separate of olivine or pyroxene, than of colourless plagioclase.

In contrast to the plagioclase data in Figure 3, ordinate and abscissa values for olivines and pyroxenes in Figure 4 do not anticorrelate. Furthermore, the two domains encompassing the data points with low and high antiquity, respectively, overlap largely. The average $(^{20}Ne/^{22}Ne)_{sol}$ ratios of early and recently exposed olivines and pyroxenes differ only slightly. The data of the mafic minerals therefore give not clear-cut evidence for a difference between the $^{20}Ne/^{22}Ne$ ratio in the ancient and recent solar wind. A possible increase of this ratio during the past 2 - 3 Ga. is less than 2% and cannot account for the variations of the $(^{20}Ne/^{22}Ne)_{sol}$ in plagioclases. This is corroborated by the lack of a correlation between $(^{20}Ne/^{22}Ne)_{sol}$ of plagioclases and mafic minerals from the same soil (figure not shown).

The considerable spread of $(^{20}Ne/^{22}Ne)_{sol}$ of the olivines and pyroxenes is probably in part to be explained by impurities also. The concentration ratio between solar and spallogenic Ne in most of these samples is so large that the errors of the isotopic composition of the solar Ne depend predominantly on the precision of the measured $^{20}Ne/^{22}Ne$ ratio and are accordingly around 1%. An exception is the data point for the pyroxene of breccia 79135 (x). This point is discarded. Its $(^{20}Ne/^{22}Ne)_{sol}$ is thought to be unreliable, because such a low ratio should be reflected in the data from the bulk soils and from the plagioclases of this breccia too, which is not the case. Among the pyroxenes, the separate of 79135 is one of the very few samples where the value of $(^{21}Ne/^{22}Ne)_{spal}$ critically influences the computation of $(^{20}Ne/^{22}Ne)_{sol}$.

Ilmenites

Based in part on the comparison of the data of several grain size suites of lunar ilmenite (cf. Eberhardt et al., 1970; Eberhardt et al., 1972), Pepin (1980) argued that the $^{20}Ne/^{22}Ne$ ratio in the solar wind may have increased with time by several percent. Figure 4 shows that the ilmenites of breccia 79035 (w) have indeed about 2% lower $(^{20}Ne/^{22}Ne)_{sol}$ ratios than the ilmenites of surface soils 70271 (s) and 71501 (t). The opaque ilmenites, however, deserve special attention with respect to contamination. Examining all grain size suites reviewed by Pepin (1980), one notes that in three isotope correlation plots of Ne the linear best fit lines extrapolate towards unreasonably low $(^{21}Ne/^{22}Ne)_{spal}$ ratios between .09 and .14. The $(^{20}Ne/^{22}Ne)_{sol}$ ratios of the 10 - 40 μm sizes grains are consistently higher, by up to 4%, than the respective values of large (100-200 μm) grains. Furthermore, in all ilmenite suites the $^{4}He/^{36}Ar$ and $^{20}Ne/^{36}Ar$ ratios decrease with increasing grain size. Small grains of a few micron size have $^{20}Ne/^{36}Ar$ ratios between 30 and 35, comparable with the solar wind value of 45 $\pm$ 10 (Geiss and Bochsler, 1976), whereas in grains around 100 - 200 μm size, this ratio is about 20, in breccia 10046 even as low as 10. Apparently, small ilmenite grains contain solar wind Ne which is less fractionated, possibly because large ilmenites are frequently intergrown with glass (Grögler, pers. comm.). "Black Glass" (Table 1), which is rich in solar gases, may well consitute a contaminant in ilmenite separates. These observations lead us to doubt whether the data from the ilmenite grain size suites, particularly the $^{20}Ne/^{22}Ne$ ratios of the solar component obtained by ordinate intercept plots, really indicate a secular change of the isotopic composition of SW-Ne. Because of the possible contamination of large grains, we also hesitate to interpret the difference of 2% of $(^{20}Ne/^{22}Ne)_{sol}$ between the ilmenites from breccia 79035 (w) and from the surface soils 70271 (s) and 71501 (t) as an indication for a secular increase of the $^{20}Ne/^{22}Ne$ in the solar wind, although such a change is not inconsistent with the data pattern of olivines and pyroxenes.

In summary, the retentive minerals show that a possible increase of the $^{20}Ne/^{22}Ne$ ratio in the solar wind during the last 2 - 3 Ga. is below 2%. Such an effect is too small to explain the observed variations of the isotopic composition of solar Ne in the plagioclase separates.

TEMPORAL VARIATION OF THE SF-Ne/SW-Ne FLUX RATIO

From Figure 3 we concluded that plagioclase separates with a large SF-Ne contribution can be found not only in drill core samples and soil breccias but also in surface soils. This may suggest that the mean flux of SF-Ne normalized to the SW-Ne flux has fluctuated on a time scale comparable to the lifetime of minerals near the regolith surface, i.e. some 10^8 years (see next paragraph). Alternatively, if the several surface soils with low $(^{20}Ne/^{22}Ne)_{sol}$ acquired their solar gases predominantly in ancient times, a secular decrease of the mean SF/SW flux ratio is indicated. A measure for the antiquity of the samples is therefore needed.

Antiquities of the Samples

A sample provides a good time resolution for solar wind and solar flare studies if it has a well constrained antiquity, i.e. a relatively short interval between the first and the last surface exposure of its individual grains. Near the regolith surface, minerals have a limited lifetime, as a result of the destruction of grains by meteorite impact. Fresh minerals are constantly admixed to the regolith by erosion of rocks and pebbles. Wieler et al. (1981) estimated that minerals which are now in a regolith surface layer of a few cm thickness, were created as individual particles typically within the last 100 Ma. Due to the limited lifetime of minerals, they not only have better constrained, but also lower antiquities than their host soils as a whole. As a measure for the antiquity, Yaniv and Heymann (1972) proposed the ratio of trapped $^{40}Ar/^{36}Ar$. The $^{15}N/^{14}N$ ratio (cf. Clayton and Thiemens, 1980), the core deposition histories as derived from neutron fluence measurements and the GCR exposure ages may also be used to confine the antiquity of a sample. In the following, we rely on the $^{40}Ar/^{36}Ar$ "clock", although it is unclear whether the retrapping mechanism for ^{40}Ar proposed by Manka and Michel (1971) is solely responsible for excess Ar. Because even recently exposed mineral grains contain substantial amounts of excess Ar, the "Transient K" hypothesis, proposed by Baur et al. (1972) is, at least for such samples, not viable, because unreasonably high potassium concentrations on the grain surfaces would be required.

The correlation between $(^{20}Ne/^{22}Ne)_{sol}$ in plagioclases - as a measure for the SF-Ne/SW-Ne concentration ratio - and $^{40}Ar/^{36}Ar$ - as antiquity indicator - is presented in Figure 5. A logarithmic scale is used on the abscissa, to reflect the exponential decrease of ^{40}K in the moon. In plagioclases, the measured $^{40}Ar/^{36}Ar$ ratio differs from the trapped $^{40}Ar/^{36}Ar$, because these minerals contain relatively large amounts of in situ produced radiogenic Ar. Therefore, we used two different substitutes on the abscissa: In the upper part of the figure, the $^{40}Ar/^{36}Ar$ ratios of olivine or pyroxene separated from the same soils as the plagioclase are plotted. Because mafic minerals were not sufficiently abundant in all our samples, the lower part of the figure shows the $^{40}Ar/^{36}Ar$ ratios of the < 1000 μm bulk samples. The $^{40}Ar/^{36}Ar$ measured in these 2 types of samples are expected to be only slightly higher than the $^{40}Ar/^{36}Ar$ ratios of the really surface distributed gas.

Only for two soils reliable antiquity estimates exist to calibrate the $^{40}Ar/^{36}Ar$ clock: Black glasses from double drive tube 74001/2 have $^{40}Ar/^{36}Ar \sim 9$ and an antiquity of $\sim 3.8 \pm .5$ Ga. (Eugster et al.,1979). The other extreme of the scale is represented by the North Ray Crater soil 67601, which was exposed to the solar radiation for the first time about 50 Ma. ago (cf. Drozd et al., 1974; Arvidson et al., 1975). The bulk soil of 67601 has $^{40}Ar/^{36}Ar = 0.64$ (Kirsten et al., 1973). The semi-quantitative time scale in Figure 5 is based on these two values. The antiquities of the samples that are read from this scale are now to be compared with other antiquity estimates:

1) The $^{40}Ar/^{36}Ar$ ratios of the olivine and pyroxene separates of most surface soils are < 1. Such low values agree in essence with the statement that minerals from surface soils typically

have antiquities on the order of 100 Ma. only (Wieler et al., 1981). Furthermore, the $^{40}Ar/^{36}Ar$ ratios of the mafic minerals tend to be about half the values of the respective bulk soil, reflecting the short lifetime of minerals as individual particles.

2) Bulk samples and mineral separates from drill core soils and breccias mostly have $^{40}Ar/^{36}Ar > 1.5$, which implies an early exposure: i) The pyroxenes of core section 60002 (a) may have acquired their solar gases 2 - 3 Ga. ago, i.e. even earlier than the 1.5 Ga., estimated by Bogard and Hirsch (1976) as upper limit for the time when these samples were exposed to the solar wind. ii) The antiquities of breccias 79035 (w) and 79135 (x) may, according to Clayton and Thiemens (1980), be as high as 2.5 Ga., qualitatively consistent with our estimates. The same authors found in breccia 79035 the lowest $\delta\ ^{15}N$ value observed in lunar samples, which may imply a very early exposure to the sun and be in a certain contrast to our estimate of 1 - 1.5 Ga. for the antiquity of this breccia. iii) Lower limits of about 500 Ma. for the antiquities of the Apollo 15 and 16 core samples (o, p, a, b) are derived from the deposition histories of these cores (cf. Wieler et al., 1979; Gose and Morris, 1977).

3) Four surface soils (f, h, i, r) have $^{40}Ar/^{36}Ar$ ratios indicating an irradiation considerably earlier than 100 Ma. ago. For 3 of these samples, independent evidence for an early exposure is available: i) Sample 15421 (r) was collected on the rim of Spur Crater, which is probably only 2 Ma. old (Storzer et al., 1973). ^{36}Ar is much too abundant in the minerals of 15421 to have been

acquired in such a short time (cf. Wieler et al., 1981), indicating that this sample indeed collected a large fraction of its trapped gases in a previous exposure. The two Spur Crater soils investigated by Kaplan et al. (1976) have the lowest $^{15}N/^{14}N$ ratios of all Apollo 15 samples studied in that work, confirming a high antiquity of 15421. ii) An early exposure of rake soil 65501 (h) and its control sample 65511 (i), both collected on the inner rim of a 20 m sized crater, is also consistent with the nitrogen data: The $^{15}N/^{14}N$ ratio of 65500 is the lowest among all Apollo 16 surface soils reviewed by Kerridge et al. (1975).

In the samples irradiated in early times, the $^{40}Ar/^{36}Ar$ ratios of the bulk soils and the mafic minerals do generally not differ significantly. Here, the resolution of the $^{40}Ar/^{36}Ar$ clock is evidently insufficient to reflect the different antiquities of primary and secondary particles, even though agglutinates must have acquired part of their surface correlated species earlier than the minerals. Mineral separates in any case provide a better time resolution than bulk samples, as a consequence of the rapid destruction of primary particles near the regolith surface.

Solar Flare Ne in Early Exposed Samples

After having confirmed the usefulness of $^{40}Ar/^{36}Ar$ as an antiquity indicator, we turn to the interpretation of the trends in Figure 5. Both parts of the figure show decreasing $(^{20}Ne/^{22}Ne)_{sol}$ ratios with increasing antiquity. As discussed in previous sections, we rule out a secular variation of the isotopic composition of SW-Ne as well as differences in the diffusive Ne losses out of early and recently irradiated samples, respectively, as causes for the trends in Figure 5. This last point is corroborated by the $(^{20}Ne/^{22}Ne)_{sol}$ and the $^{40}Ar/^{36}Ar$ ratios of olivines and pyroxenes. We therefore take the trend in Figure 5 to testify that in recent times, i.e. during about the last 100 Ma., the mean SF-Ne/SW-Ne flux ratio was lower than the average of this ratio during the past 2-3 Ga.. This flux ratio may well have steadily decreased with time. In view of the considerable uncertainties involved in the calculation of $(^{20}Ne/^{22}Ne)_{so}$ the data point "k", representing North Ray Crater soil 67601, the only sample with low antiquity that also has a low $(^{20}Ne/^{22}Ne)_{sol}$, should not be taken to indicate a short time fluctuation of the solar flare/ solar wind flux ratio.

TRACK DENSITIES AND TEMPORAL VARIATIONS OF THE SF/SW INTENSITY RATIO

Solar flare tracks in plagioclase grains represent another important record of the high energy solar corpuscular radiation. For plagioclase separates from surface soils, Etique et al. (1978) and Wieler et al. (1981) reported a linear correlation between SW-Ar concentrations and percentage of track rich grains (i.e. the fraction of grains with a track density > 10^8 tracks/cm^2 in the center of the crystals). A more quantitative measure for the solar flare dose to which a sample was exposed is its mean track density (MTD). Wieler et al. (1981) reported that the ratio between MTD and SW-Ar concentration in the plagioclase separate of core sample 15002,50 is about a factor of two higher than the average of four surface soils. Meanwhile, we determined further mean track density values: In Figure 6, the MTD/^{36}Ar ratios are plotted versus (^{20}Ne/^{22}Ne)$_{sol}$ for plagioclase separates of five surface and three drill core soils. Both these quantities are sensitive to the ratio of the fluxes of solar flare and solar wind particles. The five surface soils all have low antiquities while the three core samples were irradiated probably more than 1 Ga. ago (see Figure 5). One important feature of Figure 6 is displayed on its left hand side: The average MTD/^{36}Ar value is about twice as high for drill core soils than for surface samples. Apart from this, a trend with a negative slope may be inferred, this correlation being however by far not clear-cut. Although no monotonic decrease of the SF/SW intensity ratio can thus be deduced from Figure 6, the track data nevertheless indicate that this ratio on average was in recent times about half as high as 1 - 3 Ga. ago. This supports our conclusions deduced from the Ne data in previous sections. Note that the trend in Figure 6 conforms with the assumption of a time independent ^{20}Ne/^{22}Ne ratio in the solar flares. Thus, no change of the abundance ratio between Ne and the track producing elements must be invoked to explain the figure.

In view of the considerable uncertainties of the data points, the correlation in Figure 6 is hardly expected to be more pronounced. Also, remember that the measured mean track density of sample 60002,89 - 98 is a lower limit. A further discrepancy between the two SF/SW intensity measures might arise from the fact that solar flare tracks and SF-Ne sample the flare particle spectrum not only at different atomic numbers, but also at different energies. Track producing particles have Z > 20, and must penetrate at least 50 µm to create a track near the center of a crystal, while essentially all SF-Ne detected in our experiments resides in the outermost 30 µm of a grain. Despite this, the tracks, as an independent record of SF particles, in essence confirm the conclusions reached from the Ne data. In addition, the track data allow to quantify these conclusions.

ISOTOPIC COMPOSITION OF SF-Ne

We return now to Figure 3 to discuss the values of the $^{20}Ne/^{22}Ne$ ratio and the $^{20}Ne/^{36}Ar$ ratio of the retained flare component. The track data allow to constrain these values if we assume that the difference by a factor of 2 of the mean MTD/^{36}Ar ratio in drill core and surface samples indicates that the flux ratio of SF-Ne and SW-Ne differed by the same factor in recent and ancient times. Consequently, none of the plagioclase separates contain neither nearly pure SW-Ne nor nearly pure SF-Ne. With respect to the Apollo 16 data points in Figure 3, this means that the pure SF and SW components do not plot near the end points of the shown mixing line. In a diagram of $(^{22}Ne/^{20}Ne)_{sol}$ versus $^{36}Ar/^{20}Ne$ (cf. Wieler et al., 1982), where mixing of two components results in a linear array, values for $(^{20}Ne/^{22}Ne)_{SF}$ within the range 11.3 - 11.9 and $(^{20}Ne/^{36}Ar)_{SF}$ values larger than 3 are adequate to explain the data points of the Apollo 16 plagioclases as mixtures of SF-Ne and SW-Ne, if in all samples at least about a third of the solar Ne were of solar flare origin. This estimate for the $^{20}Ne/^{22}Ne$ ratio of the flare component can be compared to values reported in the literature: i) For SF-Ne retained in lunar plagioclases, $^{20}Ne/^{22}Ne = 11.3 \pm .2$ was deduced by Etique et al. (1981), assuming that the etched minerals did not contain SW-Ne. Otherwise this value is < 11.3. Nautiyal et al. (1982) give for the same ratio a value of 11.8 ± 0.3. ii) By satellite borne instruments, much lower $(^{20}Ne/^{22}Ne)_{SF}$ ratios around 7.6 ± 2. were measured (Mewaldt et al., 1979; Dietrich and Simpson, 1979). We conclude that our interpretation of Figure 3 is supported by the values reported for the $^{20}Ne/^{22}Ne$ ratio of SF-Ne retained in plagioclases. It appears that the satellite instruments and lunar mineral grains sampled solar flare Ne particles with different isotopic composition. The satellites recorded particles with energies between 11 and 43 MeV/nucleon, whereas the SF-Ne found in plagioclases was implanted predominantly with energies < 5MeV/nucleon (Etique, 1982).

The retentivity of SF-Ne in plagioclases can only be estimated very roughly. A lower limit for the $^{20}Ne/^{36}Ar$ ratio of the flare component of about 3 was deduced above. On the other hand, SF-Ne is most probably not retained better than spallogenic Ne. From the data in table 1 we conclude that the ratio $(^{21}Ne/^{38}Ar)_{spal}$ of the plagioclase separates varies with the samples antiquity. Using production rates for spallogenic noble gases (Hohenberg et al., 1978), we estimate that plagioclase populations with high antiquities retain 50 - 100% of the spallogenic Ne, whereas recently irradiated separates - residing near the regolith surface, where daytime temperatures are high - retain only 20 - 40% of their spallogenic Ne. Because SF-Ne is only acquired very near the regolith surface, we take 40% as an upper limit for its retained fraction. Under the assumptions that the $^{20}Ne/^{36}Ar$ ratio of the accelerated SF particles is equal to the solar abundance ratio of

$\sim$ 26 given by Cameron (1982), and that SF-Ar is quantitatively retained, this leads to an upper limit of about 10 for the ^{20}Ne/^{36}Ar ratio of the retained SF-gases. The estimates for this ratio range thus between 3 and 10. The ^{20}Ne/^{36}Ar ratio of the SW retained in Apollo 16 plagioclases is probably < 1. SF-Ne ought therefore to be retained at least between 3 and 10 times better than SW-Ne.

The plagioclase data do not allow to estimate the ratio of the Ne fluxes in the solar wind and the solar flares, because the Ne/Ar ratio of the retained solar wind and the fraction of the trapped Ne which originates in solar flares are not known well enough. Because plagioclase most probably lost part of its SF-Ne by diffusion, the ^{20}Ne/^{22}Ne ratio of the solar flare component retained in this mineral is a lower limit for the value in the flare radiation. The difference is nevertheless expected to be smaller than the corresponding difference for SW gases, as SF-Ne in plagioclase is considerably better retained than SW-Ne. It is thus safe to conclude that in the solar corpuscular radiation the SF component has a lower ^{20}Ne/^{22}Ne ratio than the solar wind.

For most, if not for all mafic mineral samples, the ratio between the retentivities of SF-Ne and SW-Ne must be lower than for plagioclase separates, because mafic minerals retain SW-Ne to between 15 and 60% already. This justifies the postulate used in an earlier section that solar Ne retained in olivine and pyroxene is well suited to investigate the isotopic composition of SW-Ne as a function of time. It is however possible that a SF contribution may slightly influence the isotopic composition of solar neon also in these minerals. This alternatively may explain the somewhat lower mean (^{20}Ne/^{22}Ne)$_{sol}$ of the early exposed olivines and pyroxenes (see Figure 4).

Concluding Remark and Conclusions

To a large extent, the conclusions presented in this study are based on the calculated values for the $^{20}Ne/^{22}Ne$ ratios of the solar gases retained in the mineral separates. The uncertainties associated with these values are comparable to the magnitude of the observed effects. However, a large number of samples were investigated. Therefore, the two trends of decreasing $(^{20}Ne/^{22}Ne)_{sol}$ both with increasing $^{20}Ne/^{36}Ar$ as well as with increasing $^{40}Ar/^{36}Ar$ ratios observed in Figures 3 and 5 are not artifacts. Besides the conclusions presented below, we studied and rejected other conceivable explanations for these trends, namely diffusive element and isotope fractionation, a secular variation of the solar wind Ne isotopic composition, different mean shielding relevant for GCR produced noble gases for early and recently irradiated samples, and a higher proportion of SCR produced noble gases in ancient samples. Two hypotheses are at the basis of our reasoning. These are the good retentivity of all regolith crystals for solar wind Ar and the constancy with time of the isotopic composition of solar flare Ne. The agreement between the solar flare track data and the Ne and Ar data appears to justify these hypotheses. Thus, we draw the following conclusions from our studies of solar flare tracks and of He, Ne, and Ar in mineral separates from lunar soils exposed to the solar corpuscular radiation at various times in the past:

1) During the last 2-3 billion years, a possible secular increase of the $^{20}Ne/^{22}Ne$ ratio in the solar wind is limited to < 2%, as is shown by the solar wind Ne retained in olivines, pyroxenes, and ilmenites. This is in contrast to the isotopic compositions of N in lunar soils.

2) In plagioclase separates, a considerable fraction of the retained Ne is of solar flare origin. Plagioclases from the lowest part of the Apollo 16 deep drill core, which possibly trapped their solar gases as early as 2-3 billion years ago, show the highest ratio of retained SF-Ne/SW-Ne of all plagioclase separates investigated. Our data are compatible with the values of 11.3 and 11.8 for the $^{20}Ne/^{22}Ne$ ratio of solar flare Ne retained in plagioclases, reported by Etique et al. (1981) and Nautiyal et al. (1982), respectively.

3) Plagioclases irradiated recently, i.e. during about the last 100 million years, have seen a lower average of the ratio of solar flare to solar wind Ne particle fluxes than samples exposed about 1 - 3 billion years ago. The solar flare track data independently confirm this variation and show that the mean solar flare/solar wind intensity ratios in past and recent times differ by about a factor of two.

APPENDIX

The Reliability of the $(^{20}Ne/^{22}Ne)_{sol}$ ratio of plagioclases

The error of $(^{20}Ne/^{22}Ne)_{sol}$ is determined by the precision of the measured $^{20}Ne/^{22}Ne$ ratio and by the uncertainty in the composition of the spallogenic Ne. Concerning the experimental error, an estimate can be made from the data of the 6 plagioclase separates from core section 60003 by assuming that the isotopic composition of the retained solar and spallogenic Ne is identical in all separates from within the 2.5 cm thick core layer. During the analyses of these samples, the contribution of CO_2^{++} on m/e = 22 was stable and comparable to the one observed for most samples discussed in this paper. Therefore, the precision of $^{20}Ne/^{22}Ne$ for separates measured in the same sample load is believed to be about 5‰, which is the relative error assigned to the solar $^{20}Ne/^{22}Ne$ ratio of the plagioclases from section 60003 (see Figure 1). An additional uncertainty of about 5‰ arises from the error in the determination of the mass fractionation in different loads. This reduces the precision of the Ne isotopic ratios to about 1%. An exception to this are the separates from core section 60002. In the load in which most of these samples were analyzed, the CO_2 level was about 5 times higher than usual. Additional corrections on the ^{22}Ne peak of up to 1.2% were therefore necessary (see "Experimental procedure"). This caused the error of about 1.5% of the solar $^{20}Ne/^{22}Ne$ ratio of the plagioclases from 60002 (see Figure 1). If we reject the data point with the largest CO_2^{++} correction, the solar $^{20}Ne/^{22}Ne$ ratio of these samples is lowered to 12.03 ± 0.10.

The plagioclases from core sections 60002 and 60003 also serve to judge the reliability of the $(^{21}Ne/^{22}Ne)_{spal}$ ratios used to deduce the $(^{20}Ne/^{22}Ne)_{sol}$ ratios. It is conceivable that samples with high antiquity acquired their spallogenic gases under a higher mean shielding than recently exposed soils. From the production rates given by Hohenberg et al. (1978), it follows that the $^{21}Ne/^{22}Ne$ ratio of galactic cosmic ray (GCR) produced Ne in plagioclase decreases with increasing shielding. Thus, because the true $^{21}Ne/^{22}Ne$ ratio is lower than that used in the computation, for well shielded plagioclase separates a $(^{20}Ne/^{22}Ne)_{sol}$ is computed which is lower than the true value. Figure 1 may indicate - although within the limits of error - a slightly lower $(^{21}\mathrm{Ne}/^{22}\mathrm{Ne})_{\mathrm{spal}}$ ratio for the plagioclases of 60002 than for those of 60003. These samples have among the lowest concentrations of SW-Ne but among the highest concentrations of spallogenic Ne of all plagioclases investigated. Inspite of such an unfavourable condition, the uncertainty of $(^{21}\mathrm{Ne}/^{22}\mathrm{Ne})_{\mathrm{spal}}$ influences the accuracy of the solar $^{20}\mathrm{Ne}/^{22}\mathrm{Ne}$ ratios only slightly: For 60002, the average $(^{20}\mathrm{Ne}/^{22}\mathrm{Ne})_{\mathrm{sol}}$ of the 7 plagioclase fractions is 11.96 ± 0.18, whereas from the regression line in Figure 1 we obtained a value of 12.08 ± 0.18. For 60003 the respective values are 12.60 ± 0.05 and 12.49 ± 0.10. The differences in both cases are below 1%. Adding another 1% to account for the precision of the measured $^{20}\mathrm{Ne}/^{22}\mathrm{Ne}$ ratio, the maximal error of $(^{20}\mathrm{Ne}/^{22}\mathrm{Ne})_{\mathrm{sol}}$ should not exceed 2% for the plagioclase separates. The mean error of 1.5% given in Figures 3 and 5 thus is justified. For the plagioclase separate from soil 67711 (cf. Wieler et al., 1981) we can also confine the $(^{21}\mathrm{Ne}/^{22}\mathrm{Ne})_{\mathrm{spal}}$ value, because these minerals have a very low concentration of solar Ne. Assuming $12.0 < {}^{20}\mathrm{Ne}/^{22}\mathrm{Ne} < 12.8$ for the solar gases in this sample, we compute $0.758 < (^{21}\mathrm{Ne}/^{22}\mathrm{Ne})_{\mathrm{spal}} < 0.774$, whereby an error of 1% of the measured $^{20}\mathrm{Ne}/^{22}\mathrm{Ne}$ ratio is included. The value of 0.768 (cf. Lugmair et al., 1976), used in this paper to compute $(^{20}\mathrm{Ne}/^{22}\mathrm{Ne})_{\mathrm{sol}}$ is well centered in this range.

To explain a shift of $(^{20}\mathrm{Ne}/^{22}\mathrm{Ne})_{\mathrm{sol}}$ from 12.7 to 12.2 for the plagioclases of soils 65501 and 65511 by an incorrectly assumed isotopic composition of the spallogenic Ne, the true $(^{21}\mathrm{Ne}/^{22}\mathrm{Ne})_{\mathrm{spal}}$ of these samples would have to be $\simeq 0.725$. This value requires a mean shielding of about 100g/cm^2 instead of 40g/cm^2. This cannot rigorously be excluded, but the much smaller differences between the assumed and the actually observed compositions of spallogenic Ne in the plagioclases of the two Apollo 16 deep drill core sections do not suggest that such large variations in the mean shielding are common. For the plagioclases from the mare samples with the lowest $(^{20}Ne/^{22}Ne)_{sol}$ (15421, 15002,50 and 79035), the situation is even more clear-cut. For these separates, the true $(^{21}Ne/^{22}Ne)_{spal}$ ratio would have to be between 0.54 and 0.59 to shift the $(^{20}Ne/^{22}Ne)_{sol}$ ratios up to 12.85. Such low $(^{21}Ne/^{22}Ne)_{spal}$ ratios due to a deep shielding are unreasonabl

Solar cosmic ray (SCR) produced Ne in plagioclase has a $^{21}Ne/^{22}Ne$ ratio around .4 only (cf. Hohenberg et al., 1978). Because of a possibly considerably stronger solar activity in the early history of the sun (cf. Newkirk, 1980, and references therein), it is thus conceivable that minerals with high antiquity contain spallogenic Ne with a lower $^{21}Ne/^{22}Ne$ ratio than is observed for recently irradiated crystals. The available data, however, show no evidence for such low ratios in an ancient sample: i) It is seen from Figure 1 that the spallogenic Ne in the plagioclases of drill core section 60002 has a very similar $^{21}Ne/^{22}Ne$ ratio to the value given by Lugmair et al. (1976). These minerals belong to the samples with the highest known antiquity. ii) The ratio of SCR/GCR produced spallogenic gas in the plagioclases of soil 61501 - also an early irradiated mineral population - is smaller than .05 as was shown by Etique (1982) by investigating etched minerals. This small SCR contribution lowers the $(^{21}Ne/^{22}Ne)_{spal}$ ratio less than about 2%. Note in this context that the Ne data of a grain size suite of the early exposed soil 74241 (Hübner et al., 1975) seem to indicate a ratio $(^{21}Ne/^{22}Ne)_{spal} \sim .4$. However, the solar Ne in this sample resides mainly in ilmenitic material, and it was discussed in a preceeding section that the data of all ilmenite grain size suites studied extrapolate to unreasonably low $(^{21}Ne/^{22}Ne)_{spal}$ ratios. iii) A substantially increased SW and SF activity during the exposure of a sample should lead to a higher solar wind gas loading of that sample. However, in comparison to recently exposed samples, neither the mineral separates nor the bulk soils with high antiquities have, on average, exceptionally large SW-Ar concentrations. None of the early exposed mineral separates is saturated with SW-Ar, as is seen from the saturation concentrations estimated by Wieler et al. (1981). If the sun really was considerably more active in the past, this was probably during the first few hundred million years of its history (Newkirk, 1980) whereas the antiquities of all mineral separates discussed here is below 3 Ga.

A final remark is to be made with respect to the interpretation of the best fit lines in Figure 1 in terms of a two component mixture. Etique et al. (1981) showed that the assumption of two Ne components in plagioclase grain size suites can lead to erroneous extrapolations of the isotopic composition of spallogenic and solar Ne, because in plagioclase samples a third Ne component, namely SF-Ne can be abundant. Only if the concentration ratio between the two solar components is identical in all samples of a suite, the two solar components appear as a single pseudo component in a three isotope plot. For samples with largely different grain sizes, this requirement is not necessarily fulfilled. The grain sizes of the plagioclase separates of the two Apollo 16 deep drill core sections, however, are all similar, and the extrapolated solar $^{20}Ne/^{22}Ne$ ratios can thus be assumed to be correct within the given limits of error.

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FIGURE CAPTIONS

Figure 1: Three isotope correlation plot of Ne in plagioclases and pyroxenes from drill core samples 60002,89-98 and 60003,138-147. A detailed description of the mineral separates is given in section "Samples". The extrapolated $^{20}Ne/^{22}Ne$ ratios of the solar components are given in the upper left hand corner of the figure, the extrapolated spallogenic $^{21}Ne/^{22}Ne$ ratios in the lower right hand corner. The direction towards $(^{21}Ne/^{22}Ne)_{spal} = 0.768$, (cf. Lugmair et al., 1976) is also indicated.

Figure 2: $^{4}He/^{36}Ar$ versus $^{20}Ne/^{36}Ar$ of mineral separates and bulk soils from drill core samples 60002,89-98 (filled symbols) and 60003,137-148 (open symbols). The average $^{20}Ne/^{36}Ar$ ratio of the plagioclase separates of both core sections are indicated.

Figure 3: $(^{20}Ne/^{22}Ne)_{sol}$ versus $^{20}Ne/^{36}Ar$ of all plagioclase separates in the grain size range 150-200 µm. Large symbols represent Apollo 16 highland samples. The ordinate values for the separates from the Apollo 16 core sections 60002 and 60003 are taken from Figure 1, all other $(^{20}Ne/^{22}Ne)_{sol}$ are calculated as described in section "Samples". The mean error on the ordinate, as indicated for one sample, is estimated in the Appendix. The broken line is explained in the text. Data point labelled (x) is off scale in the abscissa. Sample numbers are coded as follows:

a) 60002	b) 60003	c) 60009	d) 60010	e) 60051
f) 61501	g) 64421	h) 65501	i) 65511	k) 67601

l) 67941 m) 67960 n) 10084 0) 15002,89 p) 15002,50
q) 15021 r) 15421 s) 70271 t) 71501 u) 72261
v) 72501 w) 79035 x) 79135

Figure 4: $(^{20}Ne/^{22}Ne)_{sol}$ versus $^{20}Ne/^{36}Ar$ of olivines, pyroxenes and ilmenites in the grain size range 150-200 µm. For breccia 79035, the data point of the 100-150 µm ilmenite separate is also given. The mean error on the ordinate is indicated for one sample. For olivines and pyroxenes, the average ordinate values of samples with "low" and "high" antiquity, respectively, are indicated (refer to the text). Sample identifiers correspond to those given in Figure 3.

Figure 5: Upper part: $(^{20}Ne/^{22}Ne)_{sol}$ of plagioclases versus $^{40}Ar/^{36}Ar$ of olivines or pyroxenes of the same sample. Where mafic minerals in more than one grain size were measured, the average of the 100-150 µm and the 150-200 µm separates is given on the abscissa. Lower Part: $(^{20}Ne/^{22}Ne)_{sol}$ of plagioclases versus $^{40}Ar/^{36}Ar$ of the respective bulk sample < 1000 µm. The data for the bulk soils of samples 64421 (g) and 67601 (k) are taken from Kirsten et al. (1973), those ot the bulk soils of 67941 (l) and 67960 (m) are from Eberhardt et al. (1976). In both parts of the figure, open symbols represent drill core samples or soil breccias, filled symbols stand for surface soils. Sample identifiers correspond to those given in Figure 3. Linear best fit lines and correlation coefficients are indicated.

Figure 6: Ratio between the mean central track density (MTD) and the ^{36}Ar concentrations versus $(^{20}Ne/^{22}Ne)_{sol}$ for plagioclases. The letters refer to sample identifiers used in preceeding figures. The errors on the ordinate are determined by a 20 % uncertainty of the MTD and 10% for the reproducibility of the ^{36}Ar concentration in milligram sized aliquots of the same plagioclase separate. In the left part of the figure, the average ordinate values for surface and drill core samples

are given. The measured MTD of sample 60002,89-98 is a lower limit to its true value (cf. section "Results").

TABLE 1. Noble Gas Data

Sample	Grain size (µm)	Weight (mg)	^{4}He	^{20}Ne	^{36}Ar	$\frac{^{4}He}{^{3}He}$	$\frac{^{20}Ne}{^{22}Ne}$	$\frac{^{22}Ne}{^{21}Ne}$	$\frac{^{36}Ar}{^{38}Ar}$	$\frac{^{40}Ar}{^{36}Ar}$	Spallog.		Code
											^{21}Ne	^{38}Ar	
Bulk samples													
60002,94	0-1000	.747	3338000	119900	37610	2357	12.21	25.02	5.27	3.41	89		6,89
60002,94	150-200	.953	684700	24650	8674	1549	11.51	12.59	4.93	3.43	107		6,48
60003,145	0-1000	.283	4071000	87780	38560	2405	12.48	26.32	5.31	1.36	49		6,67
60003,143	150-200	.576	808600	19440	10060	1887	12.11	15.82	5.14	2.06	53		6,93
60009,3089	0-1000	1.049	3403000	77510	31660	2357	12.41	25.20	5.30	1.83	55		6,90
60010,3008	0-1000	.646	2770000	57050	25410	2361	12.38	25.42	5.30	1.60	39		6,91
60010,3008	150-200	.707	413600	10850	5570	1927	11.86	14.84	5.12	1.92	34		6,62
79035,128		.520	32770000	453200	52300	2683	12.68	29.65	5.30	1.95	92[f]		6,92
79035,128	150-200[a]	1.410	6556000	110600	16100	2407	12.36	23.53	5.18	2.28	104		6,34
79135,145		.351	30160000	442200	59870	2564	12.69	29.55	5.31	2.64	94[f]		6,94
79135,145	150-200[a]	.513	27100000	406000	52710	2735	12.63	28.69	5.29	2.67	120[f]		6,46
Plagioclases													
60002,89	100-150	.443	59300	1808	969	2277	7.80	3.14	3.56	5.09	69	103	6,55
60002,92	100-150	.504	57790	1942	1012	1998	7.55	3.12	3.54	4.78	77	110	6,60
60002,94	100-150	.574	54670	1688	1059	1813	7.35	3.00	3.61	5.82	72	108	6,73
60002,96	100-150	.630	68390	2504	1122	2139	8.53	3.95	3.77	4.78	68	99	6,75
60002,98	100-150	.807	67150	2062	1053	2184	8.44	3.82	3.88	5.43	58	84	6,77
60002,89+92	150-200	.522	35850	1007	581	1797	5.27	2.15	2.77	6.25	86	115	6,57
60002,94-98	150-200	.849	51550	1458	809	2249	7.20	2.87	3.43	5.21	67	96	6,53
60003,138	100-150	.230	84560	1825	1406	2143	10.50	6.08	4.52	2.36	24	54	6,78
60003,140	100-150	.261	92390	1939	1523	2089	10.19	5.56	4.47	2.84	29	63	6,79
60003,143	100-150	.260	120400	2358	1874	2129	10.80	7.02	4.64	1,84	25	61	6,80
60003,145	100-150	.224	88430	1877	1684	2142	10.50	6.14	4.58	2.48	24	60	6,84
60003,147	100-150	.291	113300	2600	1975	2201	10.83	7.23	4.57	1,97	27	72	6,85
60003,138-147	150-200	.570	64570	1406	969	2195	9.85	4.90	4.28	3.34	26	51	6,76
60009,3089	150-200	4.877	44960	720	399	2246	8.49	3.44	3.94	2.87	23	30	6,58
60010,3008	150-200	2.976	54440	947	706	2291	9.39	4.29	4.51	1,74	21	28	6,54
79035,128	150-200	.574	457200	7872	2876	2415	11.44	10.88	4.68	4.28	43	87	6,25
79135,145	100-150[a]	.334	209500	3167	761	1400	10.77	6.18	3.12	6.59	40	115	6,45
79135,145	150-200[a]	.320	208800	3146	571	2261	11.31	8.76	2.77	10.31	24	113	6,52
72501,61	150-200	.930	178500	3708	1930	2374	12.60	23.03	5.00	2,72	4	28	6,31
Pyroxenes													
60002,89-98	100-150	.333	301600	9237	1220	411	10.65	6.14	4.44	4.30	118	53	6,50
60002,89-98	150-200	.291	583400	8129	1353	833	10.29	5.15	4.40	6.30	133	62	6,63
60003,138-147	100-150	.257	585700	16150	1954	1090	11.96	12.42	5.08	1,72	69	22	6,72
60003,138-147	150-200	.150	187800	5225	955	496	10.72	6.09	4.88	1.74	67	19	6,71
79035,128	150-200	1.135	1205000	39460	2498	1291	12.15	16.30	4.72	1.91	101	71	6,30
79135,145	100-150[a]	.631	329500	6596	827	478	10.13	5.18	3.34	2.90	109	106	6,51
79135,145	150-200[a]	.926	333200	6691	849	549	10.24	5.40	3.51	4.32	104	94	6,59
72261,6	150-200	.270	1059000	25160	2216	1883	12.37	20.36	5.13	.44[g]	38		5,94
72501,61	150-200	.377	1438000	25150	1914	2048	12.54	19.20	5.20	.91	43	12	6,32
Ilmenites													
79035,128	100-150	.263	31670000	86500	3448	2617	12.83	28.40	5.09	2.02	28	38	6,70
79035,128	150-200	.978	21470000	70140	3336	2715	12.66	25.28	4.99	2.26	48	50	6,29
70271,5	150-200	1.140	9686000	40460	2088	2390	13.01	29.16	5.18	1.42	10	15	6,69
71501,38[b]	150-200	.816	8806000	35790	1843	2461	13.05	29.93	5.23	1.17	6	9	6,82

TABLE 1. (continued)

Sample	Grain size (μm)	Weight (mg)	^{4}He	^{20}Ne	^{36}Ar	$\frac{^{4}He}{^{3}He}$	$\frac{^{20}Ne}{^{22}Ne}$	$\frac{^{22}Ne}{^{21}Ne}$	$\frac{^{36}Ar}{^{38}Ar}$	$\frac{^{40}Ar}{^{36}Ar}$	Spallog. ^{21}Ne	Spallog. ^{38}Ar	Code
Agglutinates and Black Glass													
60002,94c)	150-200	.167	1913000	78440	37890	2233	12.08	20.94	5.23	2.79	111		6,47
60003,143c)	150-200	.192	1510000	43160	26970	2392	12.36	23.64	5.29	1.28	40		6,88
79035,128d)	150-200a)	.540	8481000	147400	26040	2573	12.34	25.90	5.23	2.21	91		6,43
79135,145e)	100-150	.183	2741000	44300	5363	2146	12.11	16.76	5.02	3.01	108		6,87
70271,5e)	150-200	.226	2686000	43680	5211	2304	12.34	21.16	5.12	1.20	59		6,86
72261,6c)	150-200	.989	2448000	64200	20780	2672	12.27	28.20	5.31	1.07	23		5,80
72501,61c)	150-200	.415	2699000	71880	22300	2727	12.54	28.32	5.31	1.10	24		6,35

Noble gas concentrations are in units of $10^{-8}cm^{3}STP/g$. Estimated errors are specified in the text. Concentrations of spallogenic ^{21}Ne and ^{38}Ar are calculated with the isotopic composition of solar and spallogenic gases used by Signer et al. (1977). Errors of these numbers are generally < 15%, exceptions are marked (see below). Numbers in the column "code" give our laboratory code for the respective samples. Remarks: a) sample crushed, nominal grain size probably not relevant. b) aliquot of the sample published by Signer et al. (1977). c) agglutinate. d) mainly microbreccias. e) black glass. f) error ∿30%. g) error ∿50%.

TABLE 2. Mean Central Track Densities of Plagioclase Separates

Sample	Number of Grains	%TRG	Mean Central Track Density (10^8tr./cm^2)
15002,50*)	59	95	7.1
60002,89-98	53	35	2.4
60003,138-147	50	68	4.3
60051*)	64	19	0.86
67601*)	40	93	3.9
67941*)	35	69	1.8
67960	43	78	3.6
71501*)	68	100	6.7

*) Data published by Wieler et al. (1981)

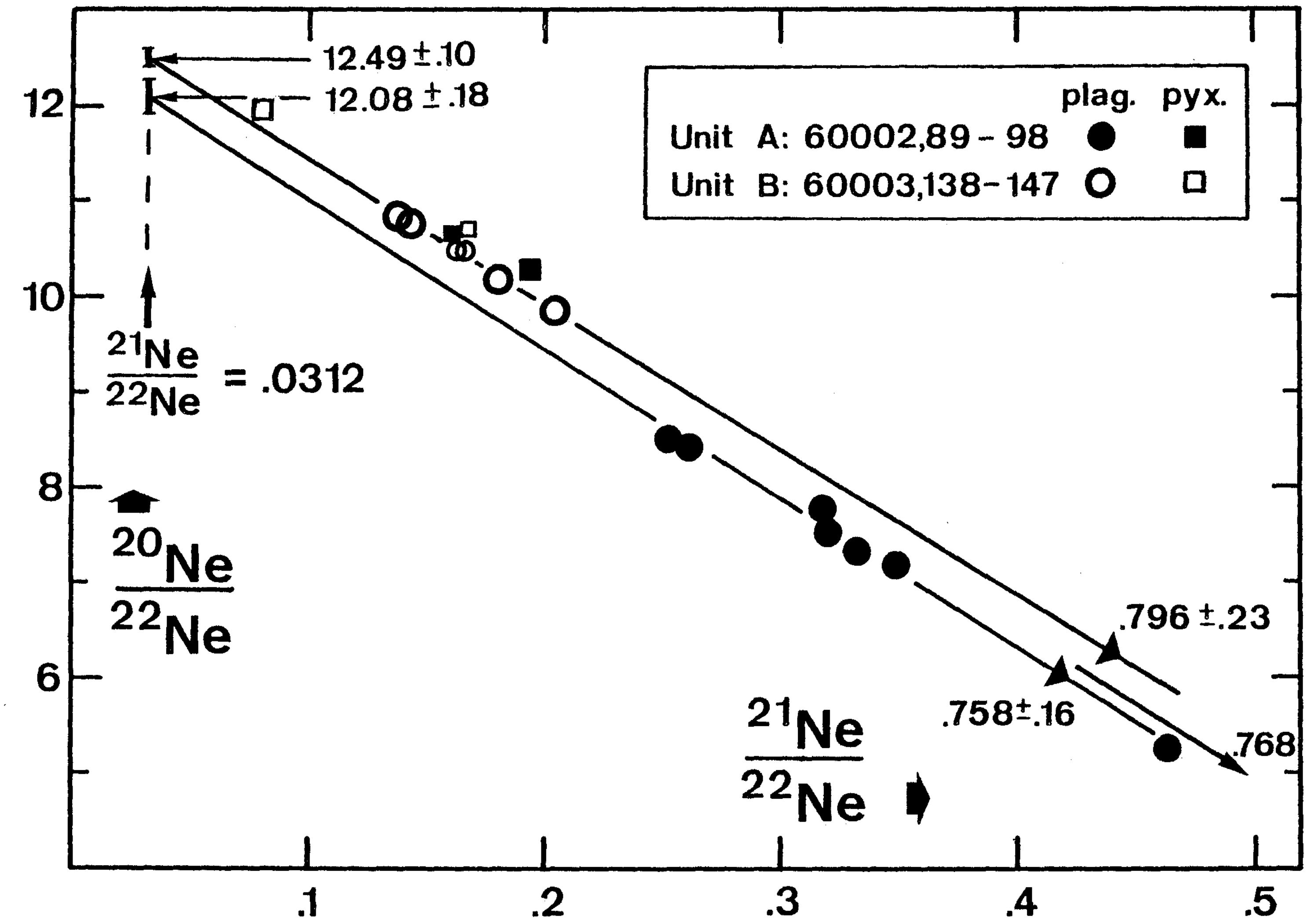
12.49 ± .10
12.08 ± .18
plag.
pyx.
Unit A: 60002,89 - 98 ● ■
Unit B: 60003,138 - 147 ○ □
12
10
8
6
$\frac{^{21}Ne}{^{22}Ne}$ = .0312
$\frac{^{20}Ne}{^{22}Ne}$
$\frac{^{21}Ne}{^{22}Ne}$
.796 ± .23
.758 ± .16
.768
.1
.2
.3
.4
.5

Figure 1

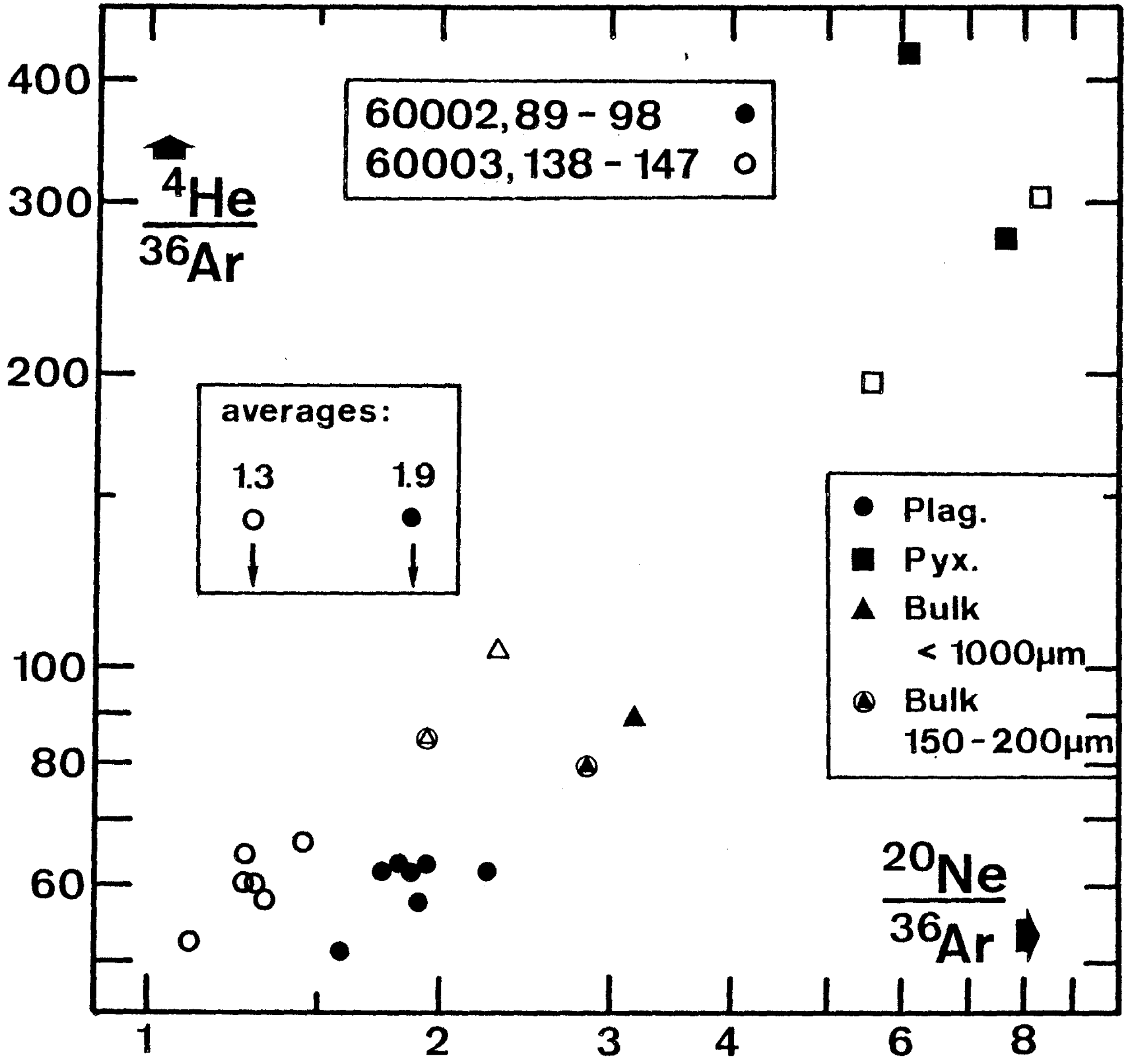
60002,89 - 98 ●
60003, 138 - 147 ○
averages:
1.3
1.9
Plag.
Pyx.
Bulk
< 1000µm
Bulk
150 - 200µm
^{4}He / ^{36}Ar
^{20}Ne / ^{36}Ar
400
300
200
100
80
60
1
2
3
4
6
8

Figure 2

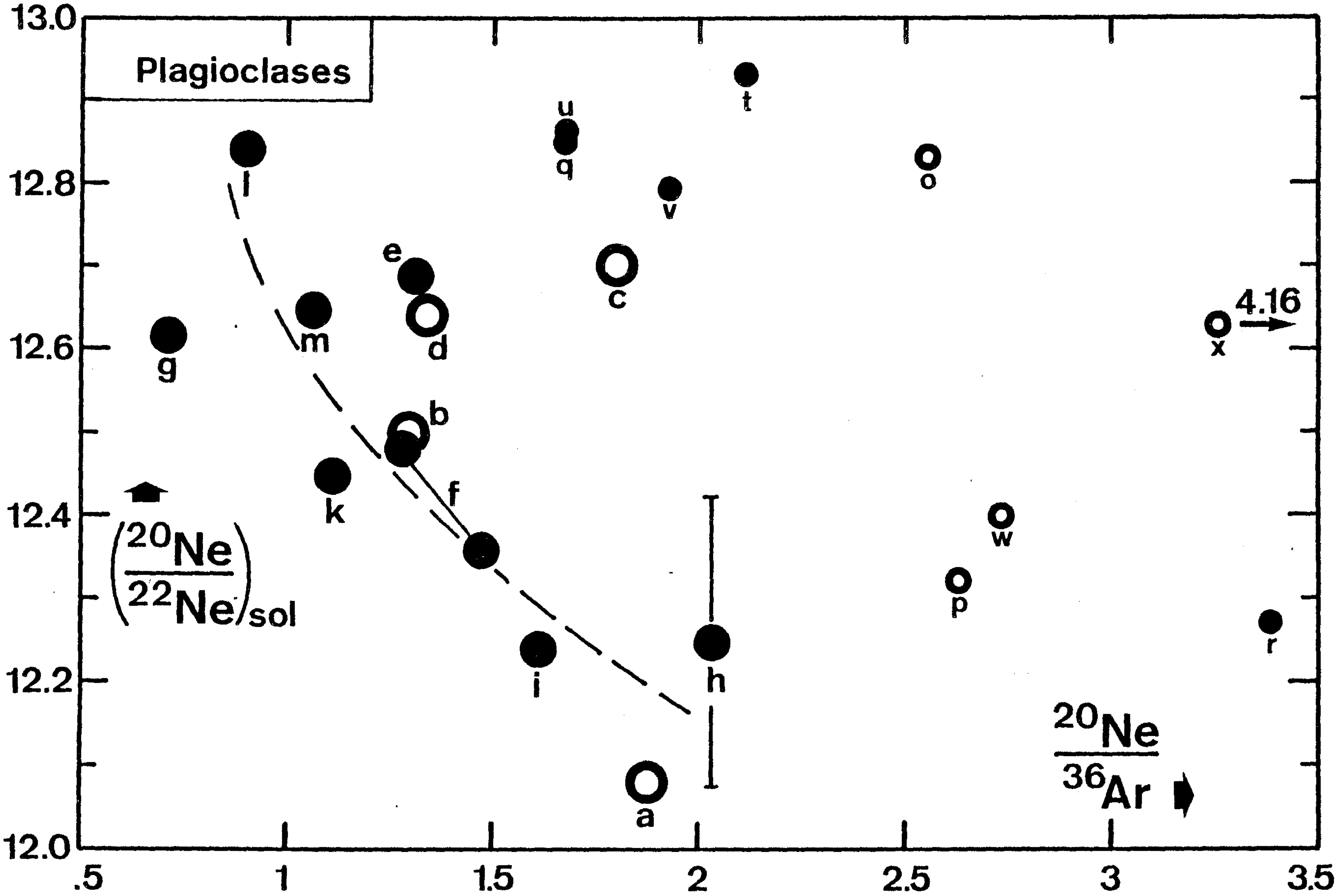
Plagioclases
13.0
12.8
12.6
12.4
12.2
12.0
.5
1
1.5
2
2.5
3
3.5
$\left(\frac{^{20}Ne}{^{22}Ne}\right)_{sol}$
$\frac{^{20}Ne}{^{36}Ar}$
4.16
a
b
c
d
e
f
g
h
i
k
l
m
o
p
q
r
t
u
v
w
x

Figure 3

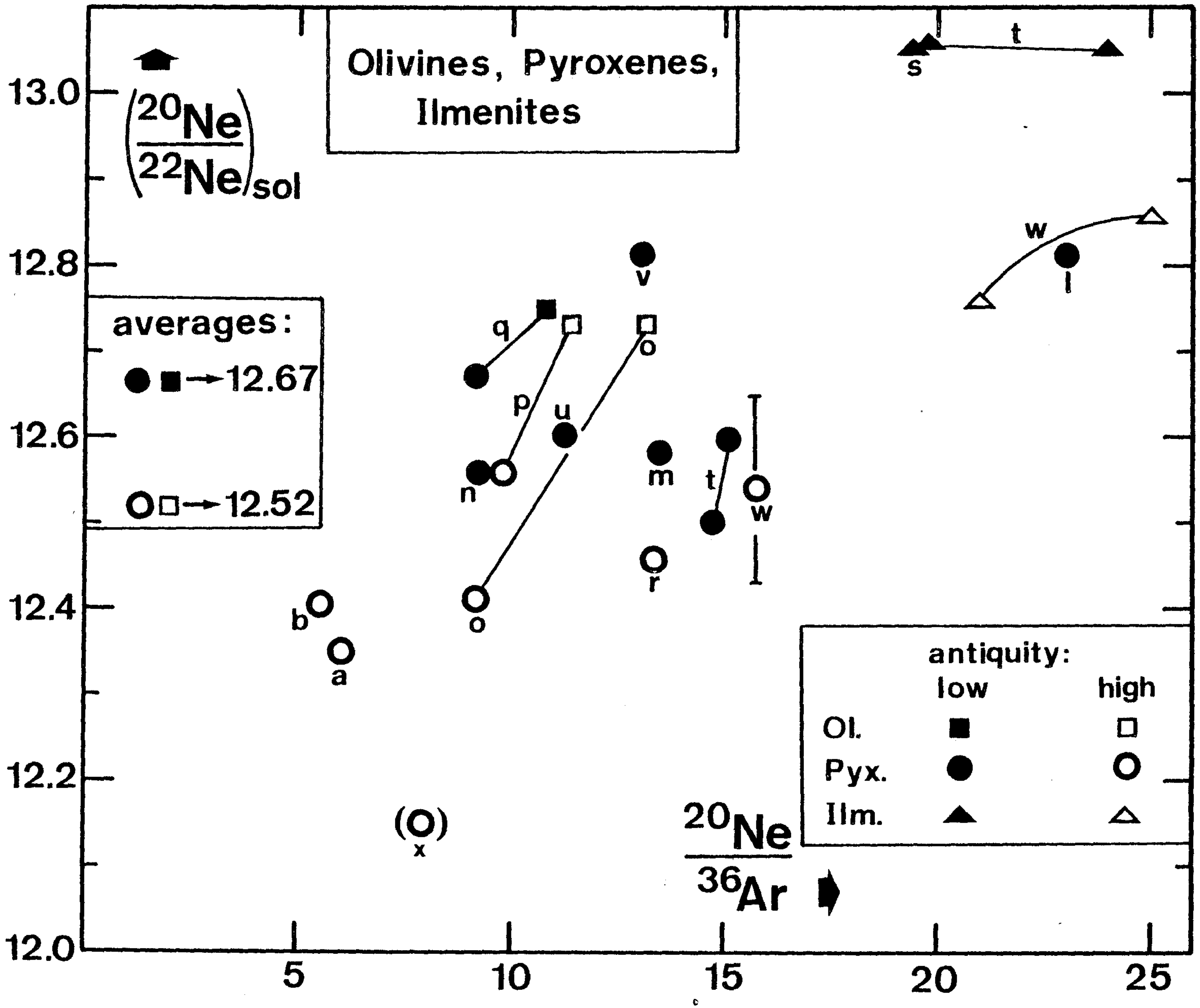
Olivines, Pyroxenes, Ilmenites
$\left(\frac{^{20}Ne}{^{22}Ne}\right)_{sol}$
13.0
12.8
12.6
12.4
12.2
12.0
5
10
15
20
25
$\frac{^{20}Ne}{^{36}Ar}$
averages:
● ■ → 12.67
○ □ → 12.52
antiquity:
low
high
Ol. ■ □
Pyx. ● ○
Ilm. ▲ △
s
t
w
l
v
o
q
p
u
n
m
t
w
r
b
a
o
(o)
x

Figure 4

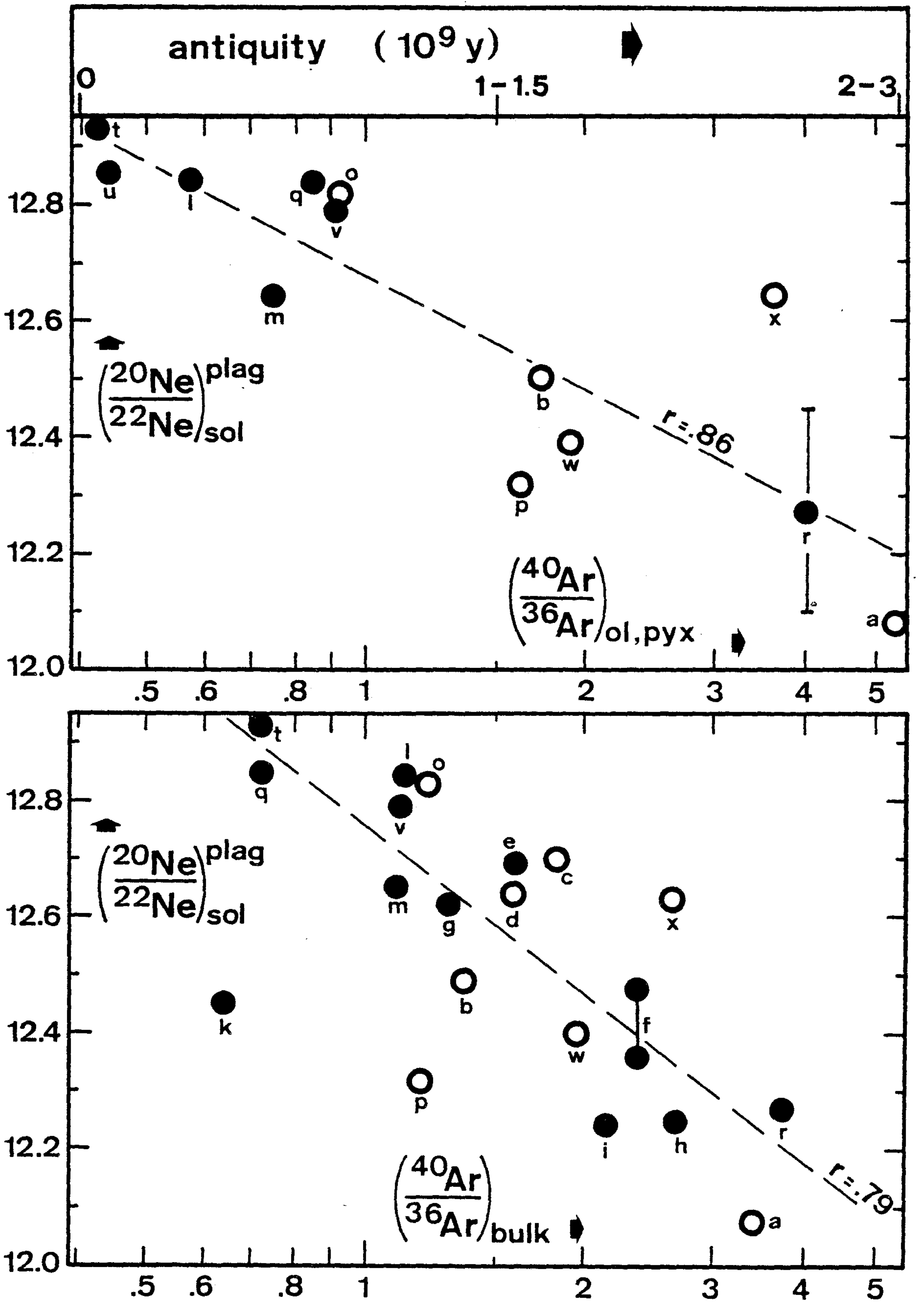
antiquity (10^9 y)
0
1–1.5
2–3
12.0
12.2
12.4
12.6
12.8
(20Ne/22Ne)plag sol
(40Ar/36Ar)ol,pyx
(40Ar/36Ar)bulk
r=.86
r=.79
.5
.6
.8
1
2
3
4
5
t
u
l
q
o
v
m
x
b
w
p
r
a
k
e
c
d
g
f
i
h

Figure 5

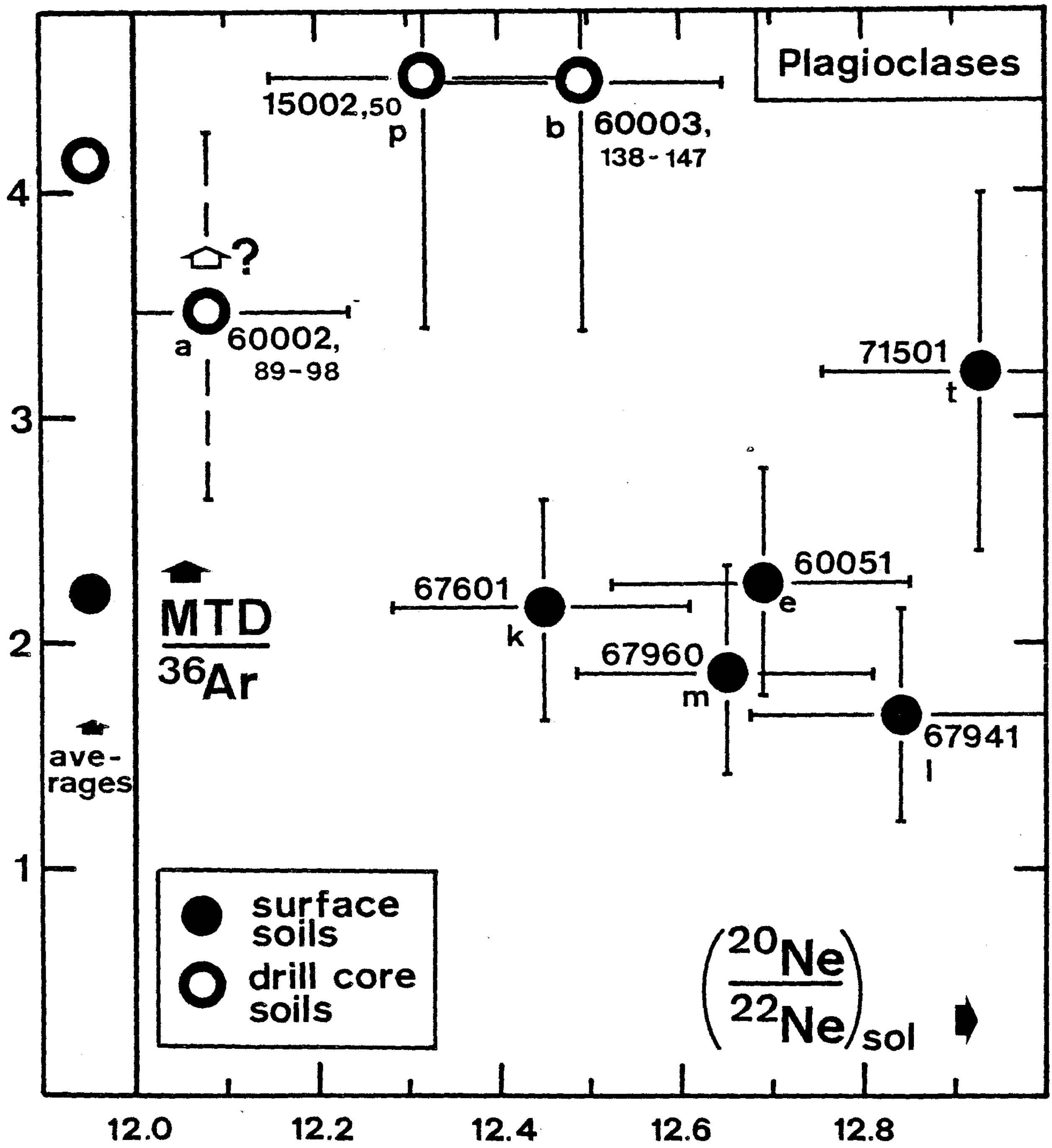
Plagioclases
15002,50
p
b
60003,
138-147
60002,
89-98
a
?
71501
t
60051
e
67601
k
67960
m
67941
l
MTD
^{36}Ar
ave-
rages
surface soils
drill core soils
$\left(\frac{^{20}Ne}{^{22}Ne}\right)_{sol}$
4
3
2
1
12.0
12.2
12.4
12.6
12.8

Figure 6