Abstract. The unique chirality of biomolecules is reviewed, and the prebiotic requirement for the absolute chiral homogeneity of such molecules prior to their capability of self-replication is emphasized. Biotic and abiotic theories embracing both chance and determinate mechanisms which have been proposed for the origin of terrestrial chiral molecules are briefly summarized and evaluated, as are abiotic mechanisms for the subsequent amplification of the small enantiomeric excesses (e.e.s) in the chiral molecules which might be formed by such processes. While amplification mechanisms are readily validated experimentally and are potentially viable on the primitive Earth, it is concluded that all terrestrial mechanisms proposed for the origin of chirality have one or more limitations which make them either intrinsically invalid or highly improbable in the chaotic and turbulent environment of the prebiotic Earth. To circumvent these difficulties we have proposed an extraterrestrial scenario for the production of terrestrial chirality in which circularly polarized synchrotron radiation from the neutron star remnant of a supernova interacts with the organic mantles on interstellar grains, producing chiral molecules by the partial asymmetric photolysis of racemic constituent in the mantles, after which the interstellar grains with their enantiomerically enriched mantles are transported to Earth either by direct accretion or through cometary impact. At this point one of the known terrestrial e.e. enrichment mechanisms could promote the small extraterrestrially produced e.e.s. into the state of chiral homogeneity required for self-replicating biomolecules.

Introduction

Over 130 years ago Pasteur (1860) emphasized the crucial importance of biomolecular chirality (handedness) in his contention that "... molecular asymmetry ... establishes ... the only well-marked line of demarcation ... between the chemistry of dead matter and the chemistry of living matter". Since that time chirality and optical activity have been deemed the principal hallmark for life, both on Earth (Gause, 1941) and elsewhere in the universe (Halpern et al., 1966; Draffen et al., 1969; Brack and Spach, 1987). It has also become evident that the principal terrestrial biopolymers each have a distinctive chirality associated with their monomeric subunits, i.e. L-amino acids for proteins, D-ribose and deoxyribose for DNA and RNA, and D-glucose for glycogen, starch and cellulose. More recently it has become further apparent that the crucial protein, DNA and RNA polymers are also composed of totally homochiral ('chirally pure') monomer units, and tolerate no vestige of 'unnatural' enantiomeric monomers in their makeup. While the question of chirality has been consistently ignored or devalued in most earlier theories and accounts of the origin of life, it is now becoming increasingly evident that unique molecular chiralities with accompanying chiral homogeneities are crucial (Avetisov et al., 1985, 1991; Keszthelyi, 1987; Goldanskii, 1988; Goldanskii and Kuzmin, 1988, 1991).
Why are chirality and chiral purity so important? In 1974 Miller and Orgel (1974) suggested that replicating double-stranded nucleic acids would be impossible with mixtures of D- and L-ribohides, and Goldanskii and Kuzmin (1988) showed with molecular models that when ‘unnatural’ L-sugars were present in double-stranded ribotide helices H-bonding between their bases was disrupted in such ways as to preclude complementarity, thus rationalizing the earlier observations of Joyce et al. (1984) involving the inhibition of template-directed nucleotide oligomerizations when chirally impure monomers were employed. Thus today it is increasingly accepted that the absolute homochirality of biopolymers is a prerequisite for self-replication, and hence for the origin of life. Since ordinary laboratory syntheses of chiral molecules result in racemic products, the questions of how molecules of a single handedness arose on Earth and how they achieved the state of chiral purity found in contemporary living matter have intrigued scientists since Pasteur’s time and have engendered numerous theoretical and experimental attempts to answer them. Our own interest in these questions has prompted us, along with conducting numerous experimental studies, to prepare three earlier comprehensive review articles on the origin and amplification of chirality, each attempting to provide an exhaustive coverage of the literature to the date of its publication. These are ‘Origins of Molecular Chirality’ (Bonner, 1972), ‘Origins of Chiral Homogeneity in Nature’ (Bonner, 1988) and ‘The Origin and Amplification of Biomolecular Chirality’ (Bonner, 1991). In the present article we attempt to recapitulate very briefly some of the topics covered in these previous reviews, conserving space by referring to pertinent pages of the earlier reviews (e.g. Bonner, 1991, pp. –) rather than to the original literature, and we expand some of our prior conclusions regarding certain of these topics.

Terrestrial Sources of Chirality

A number of biotic theories have been proposed historically to explain the unique chirality of biomolecules (Bonner, 1991, pp. 60–62; 1988, pp. 3–4; 1972, pp. 215–221). Such theories assume that primitive terrestrial life orginated by chemical evolution in a racemic environment, and that only as biomolecules evolved further did chirality enter the picture to allow for greater biochemical efficiency, thus culminating in the utilization of the L-amino acids and D-sugars characteristic to today’s biosphere. In short, chiral selection and ultimate homochirality were the inevitable consequences of the evolution of living matter itself. Other biotic scenarios have postulated that competing D- and L-organisms populated the primitive Earth, and that L-organisms finally prevailed by chance environmental or chemical events which eliminated the D-organisms, such as the fortuitous development of a D-peptidase ‘killer enzyme’ by the surviving L-organisms. Biotic theories might be expanded to included ‘panspermia’, the 84 years old notion that living organisms (presumably based on chiral molecules) from other solar systems were somehow transmitted to Earth, thereby ‘infecting’ it with life. A number of contemporary aspects of the panspermia hypothesis have been reviewed recently (Bonner, 1991,