



Cholesterol Autoxidation

Emily Lydia Schaefer



Cholesterol Autoxidation:

Cholesterol Autoxidation Leland L. Smith, 2013-11-22 Most components of the biosphere are continuously exposed to oxygen from the atmosphere Accordingly the inexorable deterioration of all organic compounds by the slow attack of oxygen must occur Despite this eventuality a definitive treatment of oxygen dependent decomposition of any single important natural product has not heretofore been made The instant monograph attempts to provide a complete description of the autoxidation of one such important natural product cholesterol as the matter is currently understood The autoxidation of cholesterol in Nature has been a matter of interest to others since the close of the nineteenth century and to me for the past three decades In this monograph I present aspects of what I have learned about cholesterol autoxidation during that interval Because of the diffuse and troublesome nature of the subject I have selected to cite references to the literature rather fully so that all items discussed may be properly evaluated by the interested reader Though such extensive citation of references makes for labored reading I hope the text will serve as a definitive treatment of the subject from which other studies may be engendered without extensive recourse to the older material An attempt has been made to include much related information so that a detailed awareness of the interrelationships between cholesterol autoxidation and other aspects of chemistry metabolism and toxicology may be had I risk teaching more on the subject than any reader care to learn Biological Effects of Cholesterol Oxides Shi-Kaung Peng, Robert J. Morin, 1991-11-22 In recent years researchers have accumulated a growing body of evidence linking cholesterol oxides with human diseases such as atherosclerosis and cancer This book presents what is presently known about the biological activities of cholesterol oxides and is intended to stimulate thinking in new areas of diet heart or diet cancer research Topics discussed include the oxidation of cholesterol methods of analysis of cholesterol oxides cholesterol oxide content in foods effects of cholesterol oxides on cell membranes and cytotoxicity of cholesterol oxides The book will be an essential reference source for researchers working directly with cholesterol and cholesterol oxides as well as researchers concerned with the wider aspects of chemistry biology and medicine

Cholesterol Autoxidation Revisited & Mechanistic Insights Into Hydrogen Atom Transfer Reactions to Peroxyl Radicals Zosia Anna Marie Zielinski, 2021 Oxygen causes the degradation of virtually all hydrocarbons over time through a process called autoxidation it is the reason engine oil breaks down plastics get brittle over time and butter turns rancid Autoxidation in many ways is what causes humans to degrade as well the process has been linked to aging cancer cardiovascular disease asthma macular degeneration and neurodegenerative conditions like Alzheimer's Parkinson's and multiple sclerosis Unsurprisingly the body has many natural antioxidant defences to combat oxidative stress and many so called superfoods are dense in antioxidants such as vitamins A C and E found in fruits and vegetables like leafy greens citrus and avocado the poly phenolic compounds found in berries green tea and red wine and the organosulfur compounds found in garlic and onions As such there has been significant interest in this area of research Our understanding of the kinetics and

mechanisms of autoxidation and the antioxidants that slow the process has advanced significantly over the past few decades however some questions remain unanswered and some widely accepted beliefs warrant revisitation One such widely accepted belief was that the autoxidation of cholesterol yields only a single regiosomeric hydroperoxide product the cholesterol 7 hydroperoxide We have shown as outlined in Chapters 2 and 3 that the mechanism of cholesterol autoxidation is far more complex than previously appreciated Indeed cholesterol 4 6 and 7 hydroperoxides are produced in the free radical process and an additional regiosomeric product cholesterol 5 hydroperoxide is produced when autoxidation occurs in the presence of a good H atom donor We have also demonstrated that the pathogenic secosterol compounds linked to a number of degenerative diseases can arise from the Hock fragmentation of cholesterol 5 or 6 hydroperoxide and heretofore uncharacterized secosterols may arise from a similar transformation in cholesterol 4 and 7 hydroperoxide This taken together with the well characterized pathogenic potential of such cholesterol derived electrophiles is excellent evidence for a link between cholesterol autoxidation and degenerative disease without invoking high energy oxidants like singlet oxygen or ozone Our mechanistic study of cholesterol autoxidation also led to the discovery of some surprising stereoelectronic and quantum mechanical effects In Chapter 3 we show that cholesteryl acetate a model for the esterified cholesterol particularly abundant in lipoproteins e g LDL and HDL autoxidizes at 4 times the rate of free cholesterol which is more abundant in lipid bilayers We also characterized the pathway of peroxy radical addition to cholesterol resulting in cholesterol epoxides This pathway could be favoured by deuterating the allylic positions in cholesterol to suppress H atom abstraction by the peroxy radical resulting in a deuterium kinetic isotope effect of 20 This evidence of quantum mechanical tunnelling in the H atom transfer pathway along with some recent reports of such an effect in other lipids led us to further explore the impact of tunnelling on H atom transfer reactions more generally Using a series of computational model systems as well as an experimental model for the simplest unsaturated lipid oleic acid we demonstrated that tunnelling in C H abstractions by peroxy radicals is likely significant across the board so long as the barriers to such reactions are sufficiently high We also suggest that oleate derived epoxides may have been overlooked as minor products of its autoxidation and the epoxides derived from plasmalogen lipids may be even more significant than the corresponding hydroperoxide products Throughout our work we have supplemented experimental evidence with computational investigations A common theme to the computations has been the influence of secondary orbital interactions in H atom transfers to peroxy radicals when the substrate has a secondary orbital donor lone pair or p system adjacent the position bearing the labile H atom which can interact with the internal peroxy oxygen atom Our work comparing such an effect in sulfenic and selenenic acids RSOH vs RSeOH as outlined in Chapter 5 is some of the best experimental evidence for the influence of secondary orbital interactions The hindered triptyceneselenenic acid reacted only 18 fold more slowly with peroxy radicals than the corresponding triptycenesulfenic acid despite its O H bond being 9 kcal mol stronger Furthermore we show that the reaction of peroxy

radicals with unhindered selenenic acids is predicted to be over an order of magnitude faster than their sulfenic acid counterparts owing to the influence of proper geometrical alignment on the secondary orbital interaction which is hindered with the bulky substituent. The influence of secondary orbital interactions is further explored in Chapter 6 where we evaluate the feasibility of quantifying or predicting the effects thereof on H atom transfer reactivity across more diverse classes of compounds. While the many confounding stereoelectronic factors present in these transition states makes such an endeavour difficult, this work has underscored the significance of computations in helping to explain seemingly anomalous experimental evidence.

On the Formation of Cholesterol Autoxidation Products in Lipid Bilayers and Electrophilic Secosterols Derived Therefrom Emily Lydia Schaefer, 2019. Lipid peroxidation is believed to play a key role in the onset and progression of degenerative disease. Interestingly, although cholesterol is the most abundant lipid in the human body, our understanding of its autoxidation and subsequent decomposition is relatively limited. In fact, until recently, cholesterol 7-hydroperoxide was accepted as the only primary product of cholesterol autoxidation in organic solution; however, our group exhibited that the 4, 5, and 6-hydroperoxides are also formed. Although this work facilitated thorough investigation of the complexities of both H atom abstraction and addition in cholesterol autoxidation in organic solution, it did not account for the dynamic environment of a cell membrane. Herein, we report on the product distribution of these primary autoxidation products in lipid bilayers and how antioxidant supplementation, H-bonding interactions, and concentration of polyunsaturated fatty acid (PUFA) substrate influence both the product distribution and efficiency of autoxidation. Indeed, not only does H-bonding of the 3-OH of cholesterol appear to shut down C4-H atom abstraction, the absence of kinetic chol-5-OOH product is likely due to the poor potency of tocopherol (TOH) also as a result of H-bonding with phosphate head group of lipid membrane phospholipids. Therefore, within a lipid membrane, the 7-hydroperoxide products predominate, consistent with literature precedent; however, the factors involved are more complex than previously understood. Moreover, with the authentic cholesterol hydroperoxides in hand, we sought to determine if the different regioisomers exhibit different cytotoxicity. Glutathione peroxidases (GPXs) are cytoprotective enzymes that reduce harmful hydroperoxides to benign alcohols in vivo. Using RSL3, a small molecule inhibitor for GPX4, we were able to sensitize mammalian cells to ferroptotic cell death via administration of our exogenously prepared chol-OOHs. Surprisingly, we found that the toxicities of each of 7-OOH, 6-OOH, and 5-OOH were only marginally augmented by RSL3 treatment, suggesting that they do not substantially sensitize cells to ferroptosis, perhaps because their decomposition to lipid peroxidation chain-initiating species, i.e., alkoxy radicals, is not particularly efficient. Instead, their cytotoxicities may derive from other mechanisms, such as the induction of apoptosis. This inspired our investigation of the fate of lipid hydroperoxides in vivo, namely the secondary products of the predominant 7-hydroperoxide species. Acid-catalyzed Hock fragmentation, known for the industrial synthesis of phenol and acetone from cumene, or its implication in the generation of 4-hydroxynonenal (4-HNE) from 5- and 6-OOH, has been shown by our group to

produce highly electrophilic secosterol species we sought to investigate the same decomposition mechanism for 7 OOH in light of our investigations in the lipid membrane Interestingly we found that Hock fragmentation of 7 OOH does not exhibit products resulting from the anticipated O vinyl oxocarbenium intermediate rather the mechanism appears to funnel through an epoxy carbenium to produce unprecedented A ring cleavage and epoxide products Herein we describe our thorough analysis of this chol 7 OOH Hock fragmentation and attempts to investigate the presence of these products in biological samples similar to previous analyses of similar products in atherosclerotic plaque extracts The products isolated and characterized through this work have provided new mechanistic insight with regards to the primary and secondary oxidation products of cholesterol in vivo through further development of these findings we hope to provide a better understanding of the implications of cholesterol oxidation in the pathogenesis of atherosclerosis

Impact of Processing on Food Safety

Lauren S. Jackson, Mark G. Knize, Jeffrey N. Morgan, 2012-12-06 The contents of this book are the proceedings of the ACS symposium Impact of Processing on Food Safety which was held April 16 17 1997 at the American Chemical Society National Meeting in San Francisco CA This symposium brought together researchers from diverse backgrounds in academia government and industry Twenty speakers discussed topics ranging from the regulatory aspects of food processing to the microbiological and chemical changes in food during processing The main goal of food processing is to improve the microbial safety of food by destroying pathogenic and spoilage organisms Food processing can also improve food safety by destroying or eliminating naturally occurring toxins chemical contaminants and antinutritive factors Unfortunately processing can also cause chemical changes that result in the formation of toxic or antinutritive factors The purpose of this book is to summarize our knowledge of both the beneficial and deleterious effects of processing Chapter I considers the consumer's perceptions about food contaminants and food processing Chapter 2 summarizes the effects of traditional and nontraditional processing methods on microorganisms in food Chapters 3 6 review the effects of processing on lipids fatty acids and cholesterol in food Changes in the nutritive value of vitamins and minerals as a result of processing are discussed in chapter 7 Chapter 8 concentrates on how processing reduces the allergenicity of some foods

Nutritional Considerations in a Changing World

G. H. Bourne, 1984-03-19 Lipid Peroxides in Biology and Medicine Kunio Yagi, 2012-12-02 Lipid Peroxides in Biology and Medicine emphasizes the importance of the control of lipid peroxides in the body for the prevention and treatment of degenerative diseases This book discusses the production of free radicals in vivo from the action of xenobiotics and comparative aspects of several model lipid peroxidation systems The lipid peroxidation and membrane alterations in erythrocyte survival and lipid peroxidations of cholesterol are also deliberated This text likewise covers the mechanism of protection against membrane peroxidation lipid peroxides as a cause of vascular diseases and peroxide mediated metabolic activation of carcinogens Other topics include lipid peroxide in aging process and production of ethane and pentane during lipid peroxidation This publication is valuable to biologists medical practitioners and clinicians researching on lipid peroxides

Bioactive Natural Products (Part E) Atta-ur Rahman, 2000-12-22 Natural products play an integral and ongoing role in promoting numerous aspects of scientific advancement and many aspects of basic research programs are intimately related to natural products. The significance therefore of the Studies in Natural Product Chemistry series edited by Professor Atta ur Rahman cannot be overestimated. This volume in accordance with previous volumes presents us with cutting edge contributions of great importance.

Oxygen Radicals in Biological Systems Lester Packer, 1994. Since biological tissues are unstable in an oxygen atmosphere a great deal of effort is expended by organisms to metabolically limit or repair oxidative tissue damage. This volume of *Methods in Enzymology* and its companion Volume 234 present methods developed to investigate the roles of oxygen radicals and antioxidants in disease. Key Features: Generation, detection and characterization of oxygen radicals; chemistry, biochemistry and intermediate states of reduction; Isolation, characterization and assay of enzymes or substrates involved in formation or removal of oxygen radical; Methods for assessing molecular cell and tissue damage; assays and repair of oxidative damage.

Cholesterol Oxidation of Superoxide Anion in Aproctic and Proctic Solutions Joo-Hee Lee, 1987. Quantification of Oxidized Cholesterol Derivatives in Foods by Liquid and Gas Chromatography Seok Won Park, 1985. Photooxidation of Cholesterol in Butter Julie Marie Luby, 1982. **Oxidation of Cholesterol in Heated Tallow** Thomas Clavin Ryan, 1982. Artifact Generation and Monitoring in the Analysis of Cholesterol Oxide Products Teresa Jean Priesbe, 2006. Lipoprotein and Cholesterol Metabolism in Steroidogenic Tissues, 1985. **Influence of Spray Drying Method on the Formation of Cholesterol Oxidation Products in Egg Powder** Jessalin Anise Winterhoff Faulkner, 1990. *Nutritional Problems of the Elderly* Flaminio Fidanza, 1983. The Haemolytic Anaemias: The hereditary haemolytic anaemias Sir John Vivian Dacie, 1985. **Journal of Food Protection**, 1989. **The Biochemical Journal**, 1928. Vols 36 include Proceedings of the Biochemical Society.

The Enigmatic Realm of **Cholesterol Autoxidation**: Unleashing the Language is Inner Magic

In a fast-paced digital era where connections and knowledge intertwine, the enigmatic realm of language reveals its inherent magic. Its capacity to stir emotions, ignite contemplation, and catalyze profound transformations is nothing in short supply of extraordinary. Within the captivating pages of **Cholesterol Autoxidation** a literary masterpiece penned by a renowned author, readers set about a transformative journey, unlocking the secrets and untapped potential embedded within each word. In this evaluation, we shall explore the book's core themes, assess its distinct writing style, and delve into its lasting effect on the hearts and minds of those that partake in its reading experience.

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