Peirce's extensive and continual research had a profound impact on the general direction of his thought.

Characteristically, Peirce did not hesitate to write several drafts of his definitions, especially when they bore on significant terms. Part of the difficulty was of course to classify the many shades of meaning for any given term, and to subcategorize them accordingly so as to allow the reader to distinguish them perceptually. As he put it, "the task of classifying all the words of language, or what's the same thing, all the ideas that seek expression, is the most stupendous of logical tasks. Anybody but the most accomplished logician must break down in it utterly; and even for the strongest man it is the severest possible tax on the logical equipment and faculty" (CSP to B.E. Smith, L80: 39–40, summer 1897).

For Peirce a good definition must do two things: it has to state the signification of the definiendum (what is essential to its conception), and it has to give an explanation of how a given kind is distinguished from all other kinds. In September 1908 Peirce jotted down the following remark in his Logic Notebook: "A dictionary definition will be (or at any rate contains) a definition proper in the case of a scientific or other exact conception; but an ordinary word needs an explanation, not a definition which almost itself needs to be expounded. . . . [A] definition proper offers as a substitute for a word whose difficulty consists in its prepossessive abstractness, a composite of words more abstract still, while an "explanation" familiarizes the mind with the use of the word by bringing together in the briefest terms possible the subclasses of occasions in which it is used and giving an interpretation of it in each of them" (MS 339: 574, 576). Peirce's C.D. definition of the verb "to explain" similarly insists that the aim is to make something evident to the minds of others by analysis, description, interpretation, elucidation, and exemplification. Hence the great importance given to the selection of many quotations that show precisely the different classes of use of a definiendum—a "much superior method" (MS 339: 576) for which Peirce especially praised the Oxford Dictionary. Hunting for and selecting the most relevant quotations was thus one of the many activities Peirce devoted himself to (the Century editors also sent him hundreds of quotations compiled by their employees and pasted on paper slips).

Critiquing definitions and improving them was a major component of Peirce's methodological arsenal. Having spent years of his life composing definitions for the C.D., he spent years studying the dictionary almost daily. He was occasionally called upon to review other dictionaries (e.g., the Funk & Wagnalls Standard Dictionary in the Nation of 8 March 1894) or works about dictionaries (e.g., the review of R.O. Williams's Our Dictionaries, in the Nation, 30 October 1890). On such occasions Peirce would invariably subject the dictionaries to statistical comparisons, notably because "one of the first questions to be asked concerning a dictionary is whether it is well proportioned in the sense of doing equal justice to different parts of the alphabet" (Contributions to the Nation, part 2, p. 40, 8 March 1894). He thought for instance that the space occupied by the A in the Century was disproportionately large. He had great respect for Murray's Dictionary (the future OED), but no single work gained his complete approval. He was not impressed with the increasing number of words dictionaries claimed to define: "the strenuous effort of the good lexicographer is to keep down his vocabulary. In an ordinary dictionary of reference, 25,000 words comprise all that anybody ever looks out. The rest is obtrusive rubbish. Completeness is not to be thought of in any dictionary" (ibid.).

The correspondence between Peirce and the Century editors must have been vast, but unfortunately very little remains (the financial and editorial records of the Century Company, which ceased to exist in 1933 when it merged with Appleton, have apparently not been preserved), and the letters we do have mostly date from after 1891, after the first publication of the C.D. We may suppose, however, that the bulk of the correspondence consisted of notes reminding Peirce of deadlines for certain sets of definitions or for sending proofsheets back, notes asking for definition clarification, special requests (sometimes from other contributors), criticisms from Peirce regarding how entries (his or others') were being edited, etc. Peirce seems to have had occasional difficulties in getting entries as he intended them into the final publication. One particular instance involved his definition of the word "university," phrased as follows: "An association of men for the purpose of study, which confers degrees which are acknowledged as valid throughout Christendom, is endowed, and is privileged by the state in order that the people may receive intellectual guidance, and that the theoretical problems may suppose, however, in the development of civilization may be resolved." Century editors suggested a revision stressing that a university had been and continued to be an institution for instruction. Peirce replied that this view was badly mistaken, and that until Americans understood that a university had nothing...