A method for computing charge distributions in saturated molecules and sigma systems is proposed. The method is based on a simple LCAO bond orbital scheme, the influence of neighboring bonds being taken into account by assuming a linear dependence of each atomic parameter on the atomic parameters of the neighboring atoms. [The *SCI*® indicates that this paper has been cited in over 315 publications since 1961.]

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"In 1952, I started PhD work with Giovanni Speroni in Florence. As a physicist, I was assigned to electric dipole moment measurements of intriguing molecules such as benzonitriloxide. At the time, additivity of bond moments was extensively applied to molecular structures. It soon appeared to me that a stumbling block was that in the case of sigma bonds no account was taken of the specific environment of each given bond. In Oxford and Varenna, Italy, lectures by C.A. Coulson and H.C. Longuet-Higgins familiarized me with pi-system calculations, and I found that we had there a technique for adjusting pi dipole moments to the environment. Unfortunately, being localized, sigma bonds could not be subjected to the same kind of treatment. But then I noticed that in the Hückel method heteroatoms were assumed to bring about an 'inductive effect' by modifying the alphas of the neighboring atoms. Why not apply the same idea to sigma systems?

"Thus the idea took shape of applying a two-center Hückel scheme to each bond, taking adjacent bonds into account by adjusting the alphas according to the surrounding atoms. As the given bond in turn affected the adjacent ones, an iterative procedure resulted, which turned out to be equivalent to a linear system for the alphas. If suitable numerical values for the parameters appearing in it could be found, there lay a solution to my problem. Eventually, I came out with correct predictions of the dipole moments for many more saturated molecules than the adjustable quantities used. The method worked.

"The next step was publication. I was still a newcomer in theory, and so it was that on one pearly evening on Lake Como I presented my case to Longuet-Higgins. To him I owe not only encouragement and suggestions, but the reading of the final manuscript, and (I suspect) actual publication of the paper. (I realized years later, on rereading the editor's letter, that publication of the paper had not been decided very easily.)

"That was in 1958. The next part of the story took place four years later, when I came back to Europe after a stay in America. For a while I was a guest of Bernard Pullman in Paris. It was he who dug out my paper and suggested to Tejiro Yonezawa and me that we apply my method to amino acids.[1,2] Only then did the 'Del Re method' start its career. A curious one indeed, as I could see when reviewing the method in 1980.[3] In spite of its simplicity, and in spite of being based on a strictly localized picture of sigma bonds, it gives reasonable charges even compared with *ab initio*.[4] The atomic parameters appear to be measures of *in situ* electronegativities.[3] With large molecules like proteins, it is still the only practicable method for electronic distributions."

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4. Flisár S, Kean G & Macaulay R. Charge distributions and chemical effects. IV. On a criterion for selecting a theoretical method for the study of molecular properties involving charges. *J. Amer. Chem. Soc.* 96:4353-7, 1974.