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Zn(II) coordination influences the secondary structure, but not antimicrobial activity of the N-terminal histatin 3 hydrolysis product

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Streszczenie

The relationship between the coordination chemistry and antimicrobial activity of Zn(II) and Cu(II)-bound histatins, salivary antimicrobial peptides, remains enigmatic. We focus on metal complexes of histatin 3 and its two products of hydrolysis: histatin 4 and its N-terminal fragment (histatin 3-4). The thermodynamic stability of these complexes is quite expected - the binding of Cu(II) via the ATCUN motif results in the formation of very stable complexes. In histatin-Zn(II) complexes, the {2Nim} type of coordination dominates, with polymorphic binding sites observed for histatin 3-4 and 5-8, result in their low thermodynamic stability compared to the complexes of histatin 3, 4, 5 and 8 with Zn(II), in which we observe a {2Nim, O- } type of coordination. Histatin 3, 3-4 and 4 have greater activity against Gram-positive bacteria than against Gram-negative ones, and Cu(II) or Zn(II) binding can, in some cases, moderately increase the antimicrobial activity of the native histatin 3 and 4, but not the remaining 3-4 fragment. The most probable reason for the metal-enhanced antimicrobial activity is, in this case, a local change of charge, while the chemically fascinating metal binding induced structural changes do not result in a change of biological activity. Neither histatin 3-4, the N-terminal fragment of histatin 3, which remains in solution after cleavage, nor its metal complexes have any antimicrobial activity, but histatin 3-4 presents intriguing Zn(II)-induced structural behavior, changing its secondary structure, with a tendency to form an α-helix.

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