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Bioinorganic chemistry of calcitermin – the picklock of its antimicrobial activity.

Autorzy

Denise Bellotti

Mattia Toniolo

Dorota Dudek

Aleksandra Mikołajczyk

Remo Guerrini

Agnieszka Matera-Witkiewicz

Maurizio Remelli

Magdalena Rowińska-Żyrek

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Streszczenie

Calcitermin, an antimicrobial peptide from the fluid of the human airways, is a well-conserved, 15 amino acid C-terminal cleavage fragment of calgranulin C (VAIALKAAHYHTHKE), which is active under acidic pH conditions (pH 5.4). In an attempt to understand the impact of the coordination of Zn(II) and Cu(II) on the biological activity of calcitermin, we mutated each of the histidines with an alanine and studied the thermodynamics, binding mode and antimicrobial activity of wild type calcitermin and its H9A, H11A and H13A mutants and their Zn(II) and Cu(II) complexes. Both metals strongly enhance the antimicrobial activity of calcitermin-like peptides, although the link between the minimal inhibitory concentration (MIC) values and the stability, charge or structure of the complexes is not so obvious. As expected, the increase in the number of histidines makes the coordination of both metals more effective. There is no preferred Cu(II) binding site in calcitermin: the stabilities of the Cu(II)–H9A and Cu(II)–H13A complexes are almost identical, while the Cu(II)–H11A complex (in which two histidines are separated by three amino acids and only one His residue is involved in binding) is less stable. On the other hand, the higher stability of the Zn(II)–H13A complex with respect to those formed by H9A and H11A suggests a pivotal role of His9 and His11 in Zn(II) complexation. Impressive MIC breakpoints were obtained, similar and lower than those for commonly used antimicrobial agents that treat *Candida albicans* (Zn(II) and Cu(II) complexes of WT calcitermin and H9A, as well as H9A alone), Enterococcus faecalis (H11A, H13A and their metal complexes) and *Staphylococcus aureus* (H13A and its complexes).

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