

Anthracyclines: Selected New Developments

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Abstract: Anthracycline antibiotics play an important role in cancer chemotherapy. The need for an improvement of their therapeutic index has stimulated an ongoing search for anthracycline analogues with improved properties. Analogue development was originally limited by a lack of information on the cellular drug target, nevertheless almost 20 years ago the mechanism of action of doxorubicin and daunorubicin was revealed and DNA topoisomerase II was recognised to be their main cellular target.

Several anthracyclines interfere with topoisomerase II functions by stabilizing a reaction intermediate in which DNA strands are cut and covalently linked to tyrosine residues of the enzyme. Investigations on the sequence specificity of doxorubicin *in vitro* and in nuclear chromatin of living cell have led to a molecular model of drug receptor on the topoisomerase II-DNA complex. Anthracyclines are likely placed at the interface between the DNA cleavage site and the active site of the enzyme, forming a DNA-drug-enzyme ternary complex. Moreover, a quite detailed structure-function relationship has been established for anthracyclines. First, drug intercalation is necessary but not sufficient for topoisomerase II poisoning; second, the removal of the 4-methoxy and 3'-amino substituents greatly increases the drug activity and third, the 3' substituent of the sugar moiety markedly influences the sequence selectivity of anthracycline-stimulated DNA cleavage. These relationships have been exploited during the last decade by several groups, including ours, in the search for new anthracycline drugs with lower side effects and higher activity against resistant cancer cells.

This review will focus on areas of the anthracycline field including synthesis of new analogues, new strategies of synthesis and recent developments in the area of drug delivery.



INTRODUCTION

Anthracyclines are among the most utilised antitumor drugs ever developed [1-4]. The first identified anthracyclines, daunorubicin and doxorubicin, were isolated from pigment-producing *Streptomyces* spp. in the early 1960s and still remain in widespread clinical use today [5]. Anthracyclines are essential components of several curative drug combinations for the therapy of a number of tumors. It is a drug of primary use in the treatment of patients with lymphoma, breast cancer and sarcoma.

Although active against a variety of solid and haematological malignancies, their clinical use is hindered by tumor resistance and toxicity to healthy tissues.

Thus, the clinical success of doxorubicin, as well as its limitations in the clinical use has been the basis to find a "better doxorubicin" [6].

The research has moved in different directions: 1) the understanding of the mechanism of actions of anthracyclines; 2) the synthesis of analogues to improve the ability to form stable complexes with DNA-topoisomerase II, possibly leading to an increase in antitumor efficacy. Since doxorubicin

was discovered, more than two thousands of analogues have been reported. 3) Modify the pharmacokinetic properties and the delivery of the drugs to cancer cells in order to increase antitumor efficacy *in vivo* and/or reduce side effects.

From the chemical point of view all anthracyclines consist of an aglycone ring coupled with an aminosugar and their major molecular target that determines the clinical activity is the poisoning of DNA topoisomerase II [7]. A wealth of data shows that antitumor anthracyclines stabilize a topoisomeraseII-DNA covalent complex and hinder a specific step of the enzyme catalytic cycle, the resealing of DNA breaks [8-11]. This is followed by enhancement of DNA cleavage levels in living cells as well as in DNA fragments in the presence of purified topoisomerase II [10]. Thus, anthracyclines transform a harmless protein, topoisomerase II, into potent nuclear toxins that lethally damage the double helix [10].

The peculiarity of anthracyclines and, in general, of topoisomerase II poisons, is that their action is DNA sequence specific. Each topoisomerase II poison can stimulate DNA cleavage at certain sites, but not at all the sites recognized by the enzyme, suggesting that drug interactions depend on the local nucleotide base sequence.

Investigations on the sequence specificity of drug stimulation of topoisomeraseII-mediated cleavage have shed light on the drug mechanism of action [8,12] and have

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