



Universidade Federal do Rio de Janeiro



# "Química Medicinal e a inovação farmacêutica: experiência do LASSBio & INCT-INO FAR"

**SIMQUIMED**

II Simpósio de Química Medicinal da UNESP  
São José do Rio Preto - SP / 08 a 12 de agosto de 2016



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Professor Titular -UFRJ




Laboratório de Avaliação e Síntese de Substâncias Bioativas

### Laboratório de Avaliação e Síntese de Substâncias Bioativas

<http://www.lassbio.icb.ufrj.br/>



# Índice

- Definição & *berço* da Química Medicinal;
- A inovação tecnológica & farmacêutica; 
- O processo de *drug discovery* e a interdisciplinaridade;
- Os fármacos e o prêmio Nobel;
- Exemplos de importantes inovações farmacêuticas:  
AAS, penicilina, atorvastatina & imatinibe;
- Os fármacos do século 21;
- Exemplo “*de casa*” LASSBio/ICB-UFRJ & INCT-INO FAR:  
LASSBio-1819;
- Considerações finais.



# IUPAC - Subcommittee Medicinal Chemistry & Drug Development

Definição: **Química Medicinal** é a

*disciplina* que estuda os aspectos relacionados

à descoberta ou invenção dos fármacos, OS

aspectos moleculares envolvidos em seu

mecanismo de ação e aqueles que governam a

*absorção, distribuição, metabolismo, eliminação*

*toxicidade* (ADMET), incluindo a compreensão

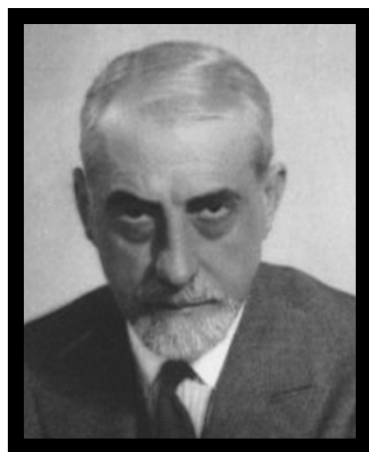
da relação entre a estrutura química e a

atividade terapêutica (REA ou SAR).



**Química**  
m e d  
**Medicinal**  
c h e m

# O berço da Química Medicinal

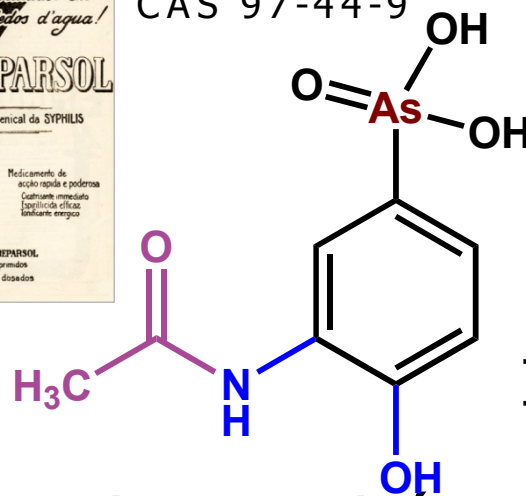


Ernest Fourneau  
1872-1949



Stovarsol

CAS 97-44-9



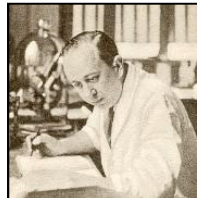
Institut Pasteur (1887)

## 1911- Laboratoire de Chimie Thérapeutique



Diretor: Emile Roux

1911-1944 – Jacques Tréfouël (1897-1977)  
Thérese Tréfouël (1892-1978)  
Germaine Benoit (1901-1983)  
Federico Nitti (1903-1947)



Daniel Bovet  
1907-1992 \*

\*Farmacêutico suíço  
Doutor *h.c.* UFRJ

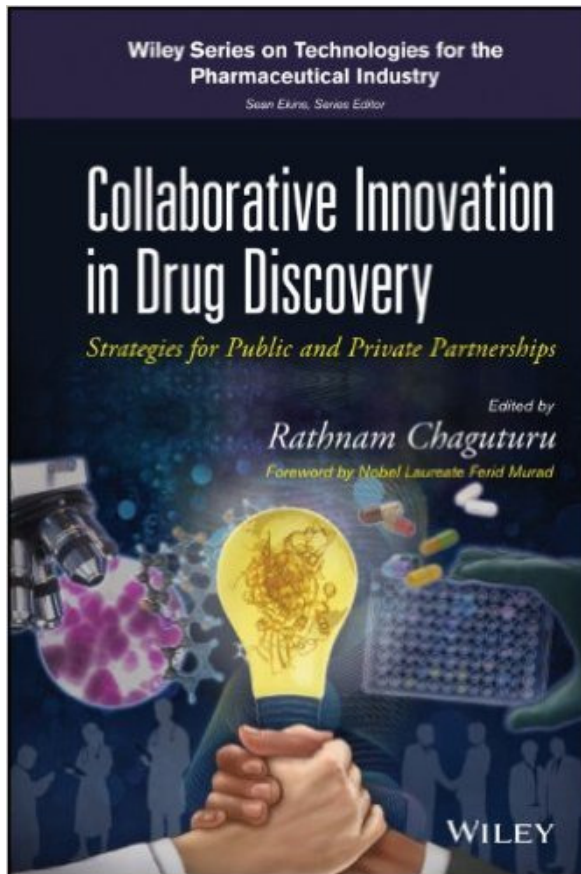
Prêmio Nobel de  
Fisiologia/Medicina  
1957

*Sulfonamidas,*  
anti-histamínicos.

Curare: SAR



# A inovação tecnológica



CEO of iDDPartners, US

[XXII Escola de Verão em Química Farmacêutica Medicinal](#)  
(LASSBio/ICB-UFRJ, 2016)

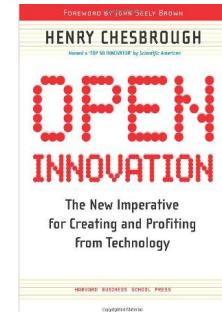
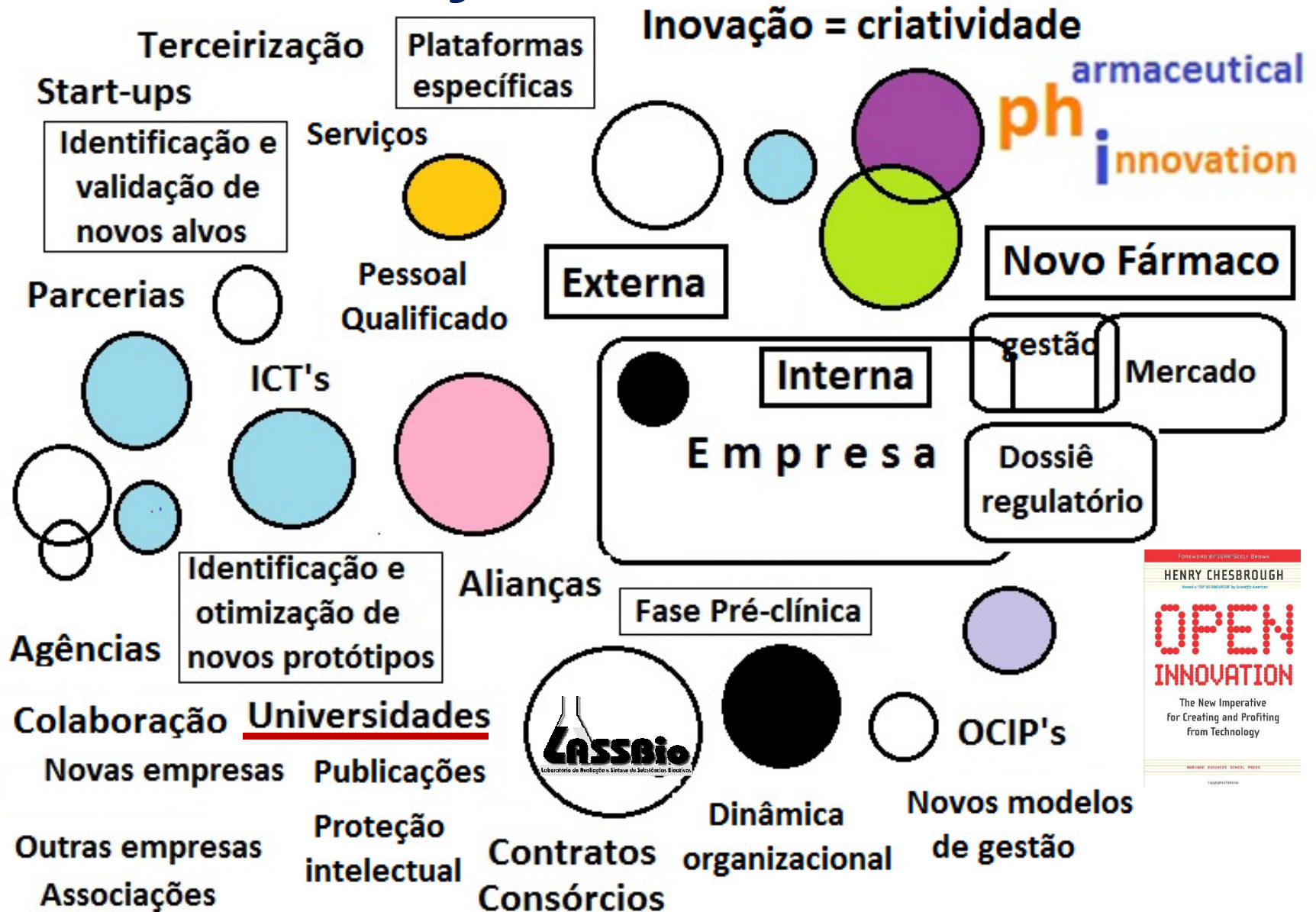
é o processo **mais** dinâmico da atividade industrial. Este **dinamismo** é **acentuado na** inovação farmacêutica que, **mais do que** qualquer outra, depende da **efetiva interação entre** **Ciência & Tecnologia.**

Impact **Innovation** Initiative  
Ideas Inspiration



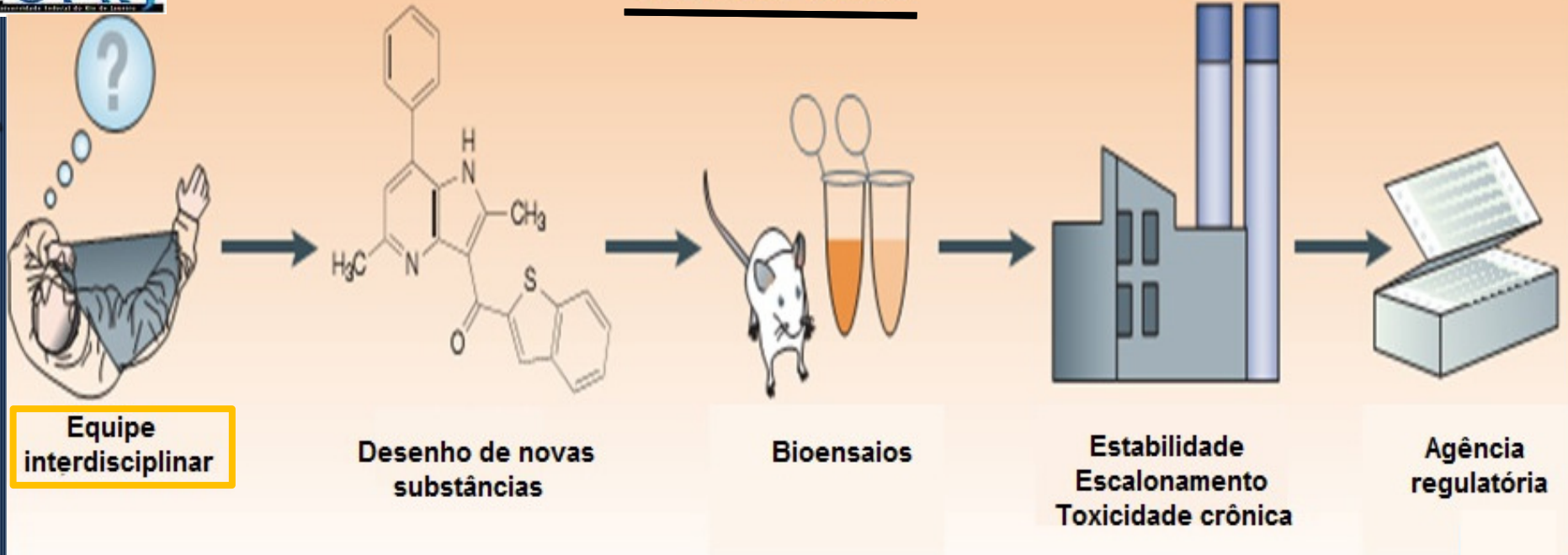
A inovação não espera...

# A gestão da inovação na indústria farmacêutica atual





## Fase pré-clínica



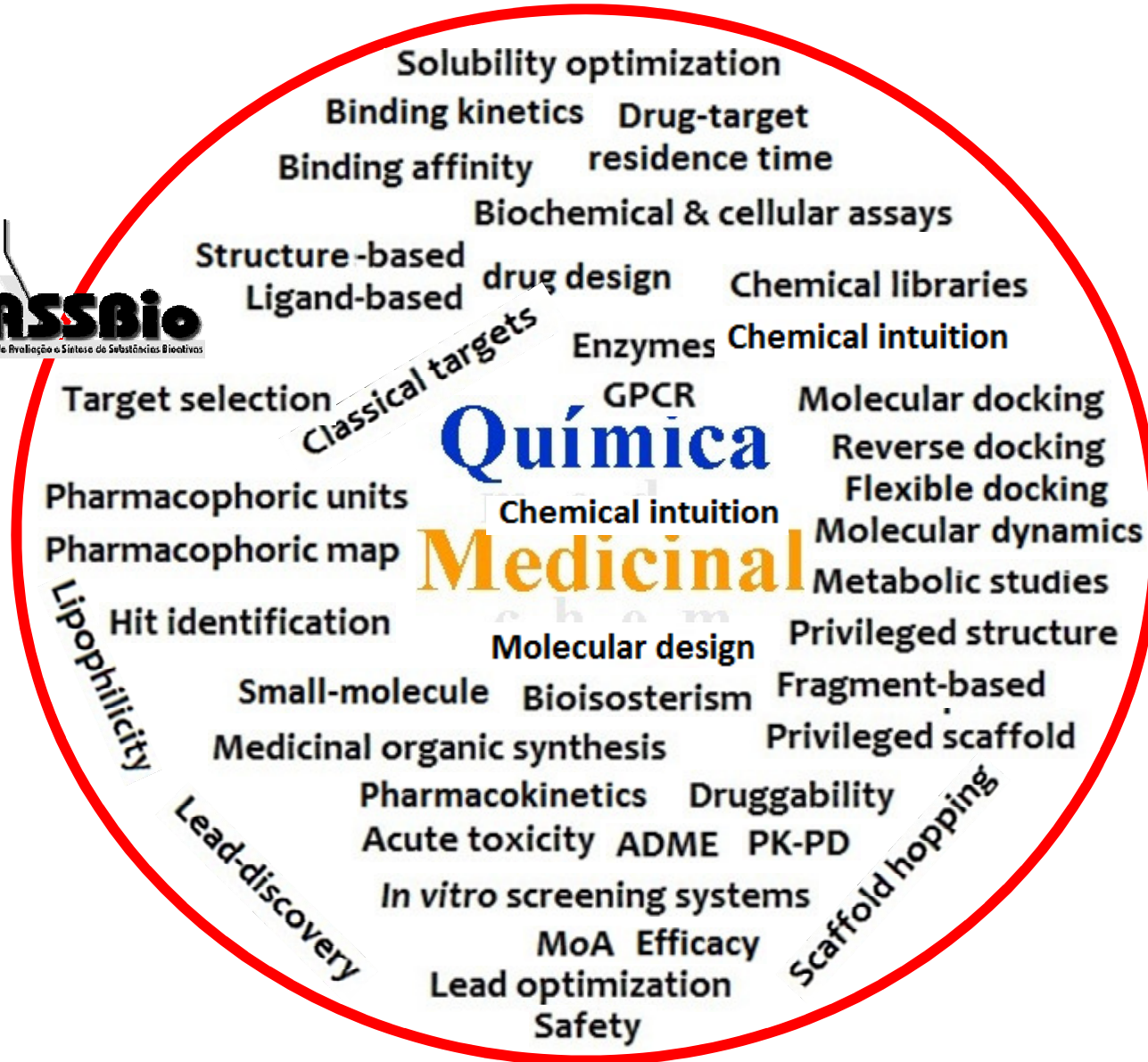
## Fase clínica

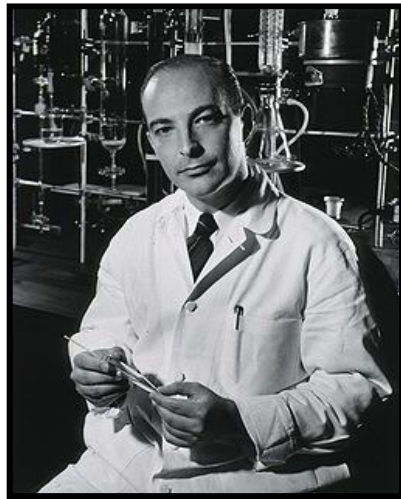
O processo *D2* é complexo & interdisciplinar...



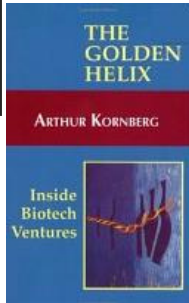
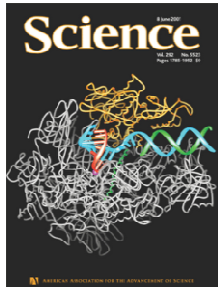


# O processo de *drug discovery*



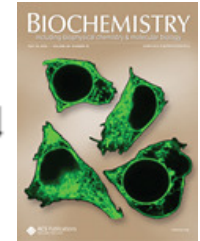


Arthur Kornberg (41)  
1918-2007



FORN

# Prêmio Nobel, 1959



## The Two Cultures: Chemistry and Biology<sup>1</sup>

Arthur Kornberg

Department of Biochemistry, Stanford University, Stanford, California 94305

Received July 14, 1987

*“Much of life can be understood in rational terms if expressed in the language of chemistry... the historical roots of chemistry and biology are intertwined in many places...*

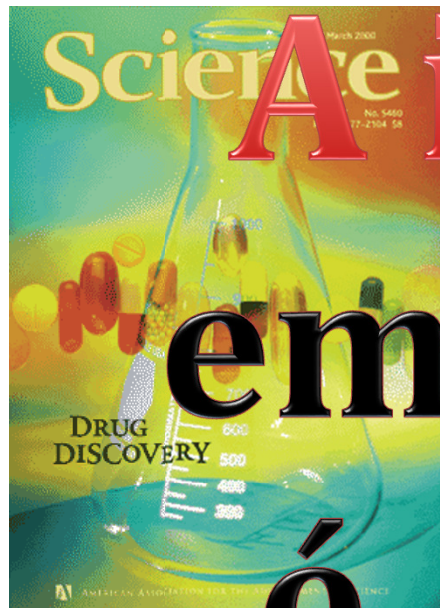
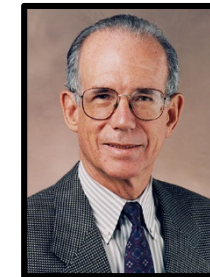
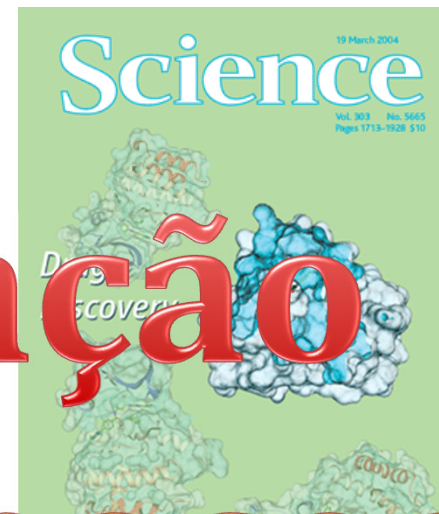
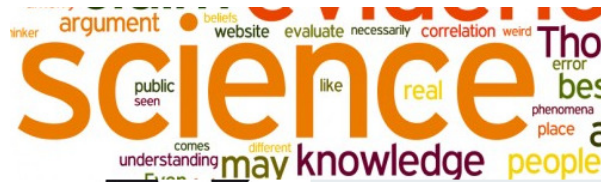


*Pharmaceutical chemistry was until recently the bastion of organic chemistry... in the search for alternative or superior drugs for the treatment of various diseases...”*

# Interdisciplinaridade

*J Biol Chem* 1987, 26, 6888-6891





# A inovação

# em fármacos

[OnLine](#)

• *Science* **2004**, 303, 1713

(Donald Kennedy)

# é baseada

• *Science* **2000**, 287, 1951

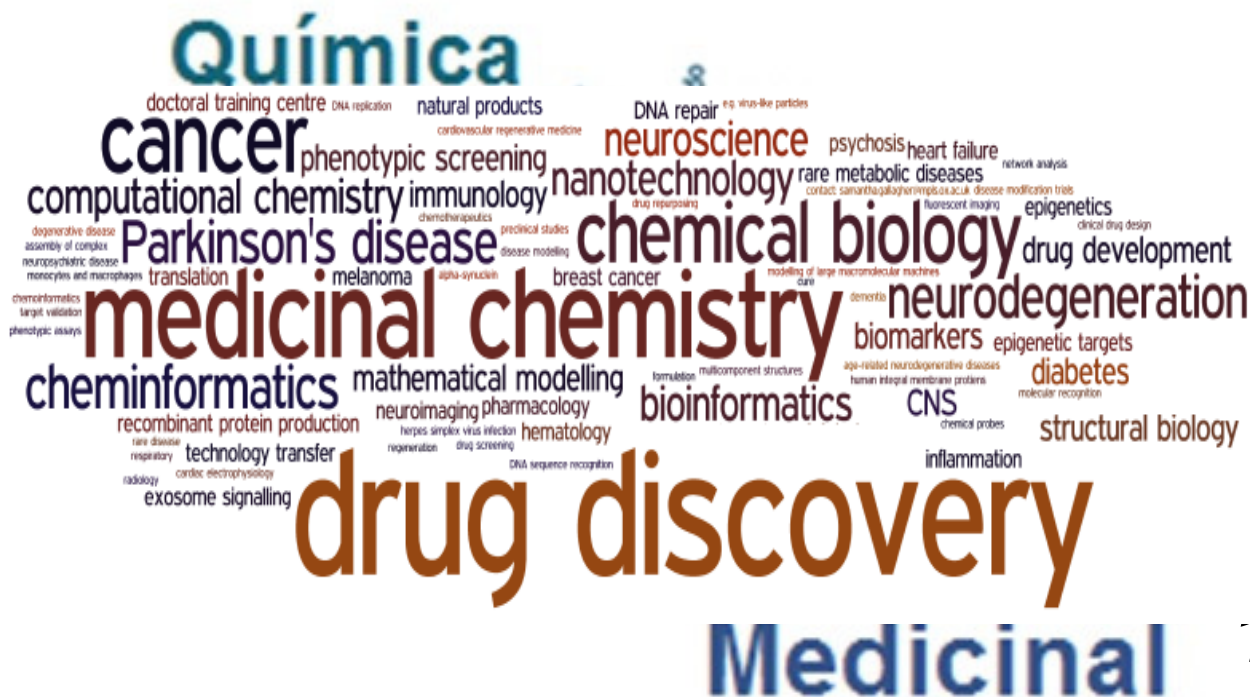
(Julia Uppenbrink, J. Mervis)

# em Ciência!

• *Science* **2005**, 309, 728

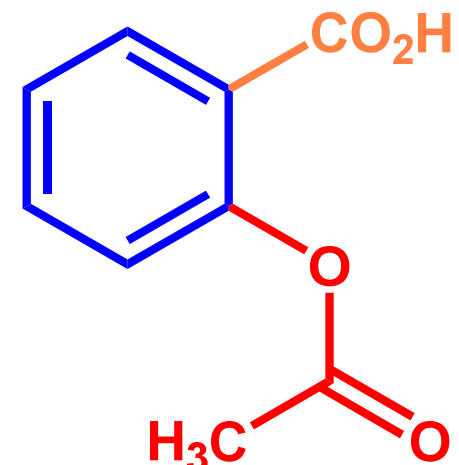
(Jeffrey Mervis)





CAS contém 116 milhões de substâncias químicas

<https://www.cas.org/>

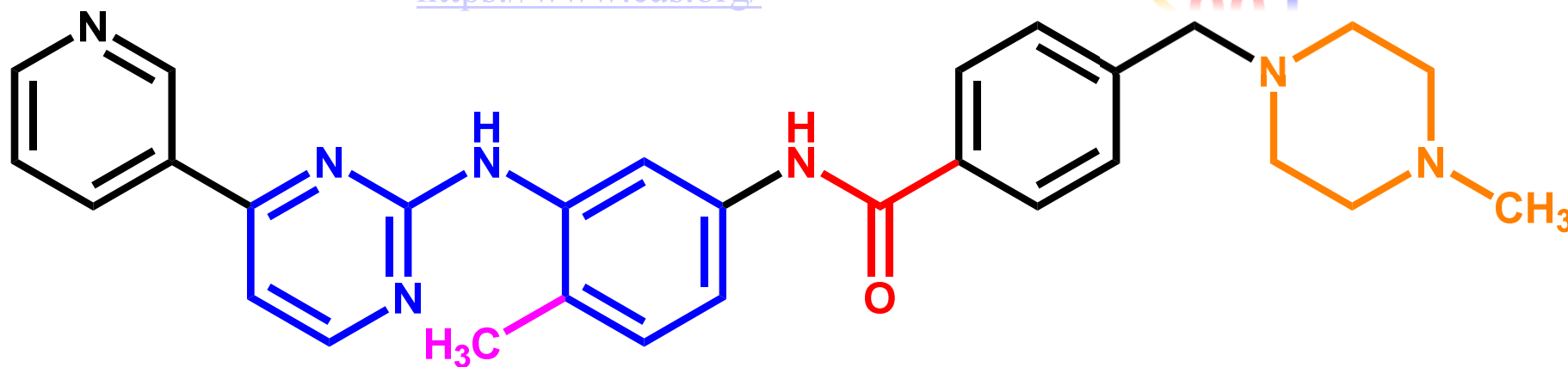


1889

112y



2001







1901

■ 210 pesquisadores ganharam o Prêmio Nobel de Medicina de 1901 a 2015

● 171 pesquisadores ganharam o Prêmio Nobel de Química de 1901 a 2015



Alexander Fleming  
Robert J. Lefkowitz



Emil Fischer  
Sune K Bergström  
George Hitchings  
Ernest B Chain  
Edwin G Krebs  
Howard W. Florey



John R Vane



Penicilina



AAS

Dorothy C Hodgkin  
Robert Robinson

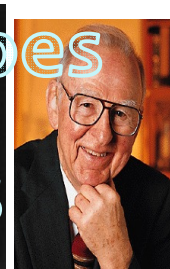


Estatinas

Arieh Warshel  
Brian K Kobilka  
Gerhard Domagk



Tinibes



Martin Karplus  
Gertrude B Elion  
James W Black  
Bengt I Samuelsson  
Edmond H Fischer  
Michael Levitt



Aciclovir

2015

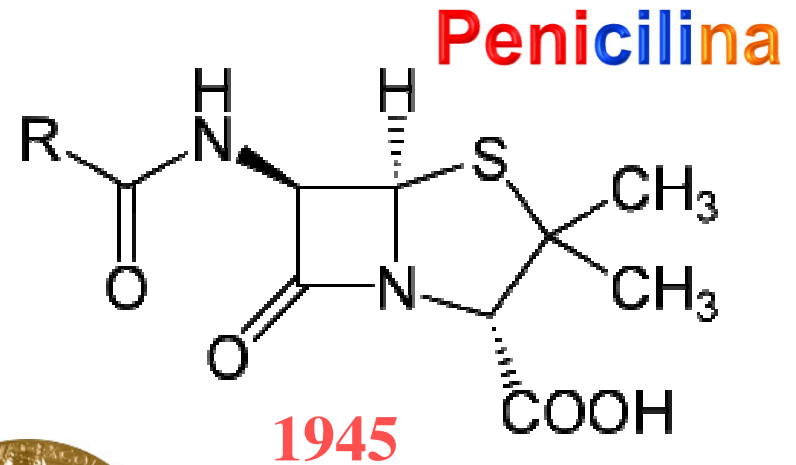
# Os fármacos e o Nobel !



**Sune K. Bergström**  
(1916-2004)



**John R. Vane**  
(1927-2004)



**Howard W. Florey**  
1898-1968



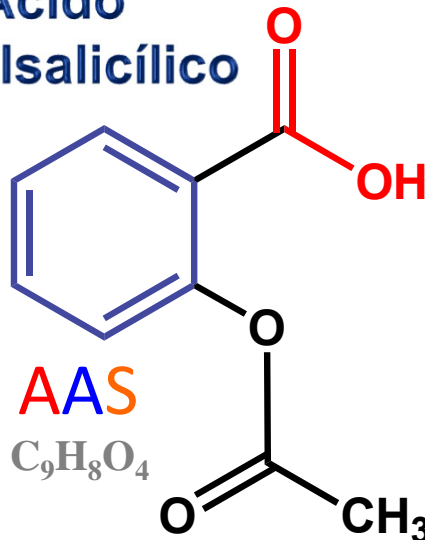
**E. Boris Chain**  
1906-1979



**Bengt I. Samuelsson**  
(1934- )

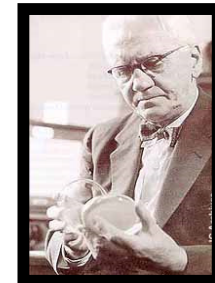
1982

**Ácido acetilsalicílico**



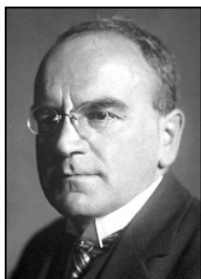
**Dorothy C. Hodgkin**  
1910-1994

1964



**Alexander Fleming**  
1881-1955

# Uma inovação bilionária: as estatinas



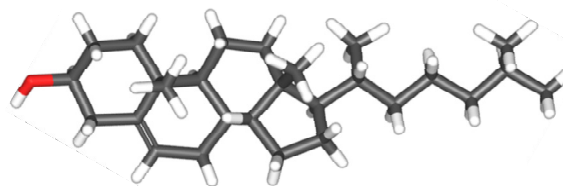
Heinrich Wieland  
1877-1957

1927

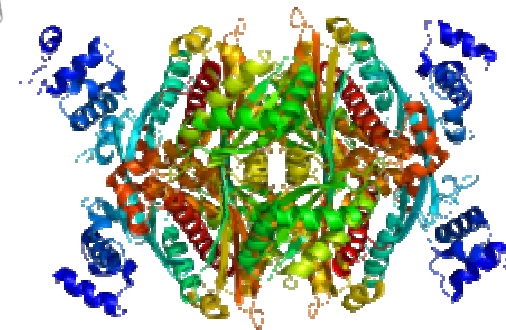


Adolf Windaus  
1876-1959

1928



colesterol



HMGCoAR



1964



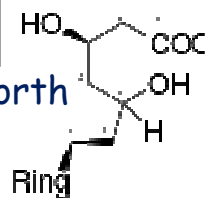
Konrad Bloch  
1912-2000



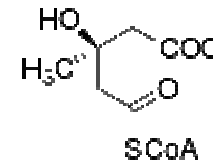
Feodor Lynen  
1911-1979



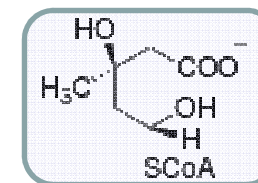
John Cornforth  
1975



HMG CoA Reductase inhibitor



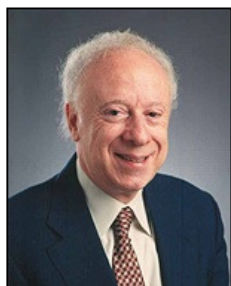
HMG CoA



Mevaldyl CoA transition state intermediate

1985

LDL

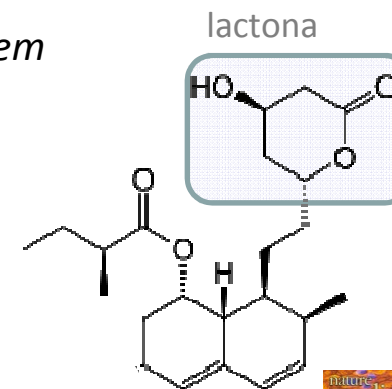


Joseph L Goldstein Michael S Brown  
University of Texas, Dallas



Akira Endo  
Albert Lasker Award  
for Clinical  
Medical Research, 2008\*

A.Endo, *J Med Chem*  
1985, 28, 1



mevilonina

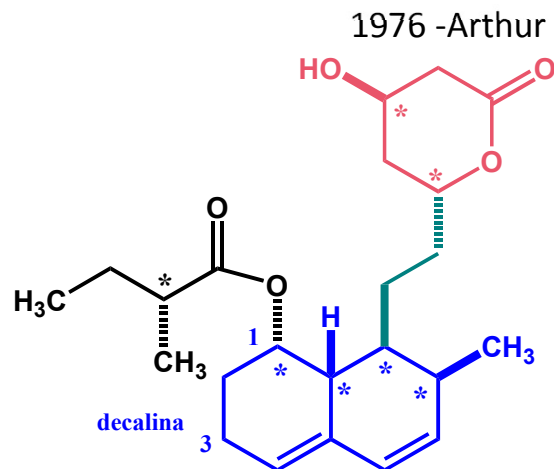
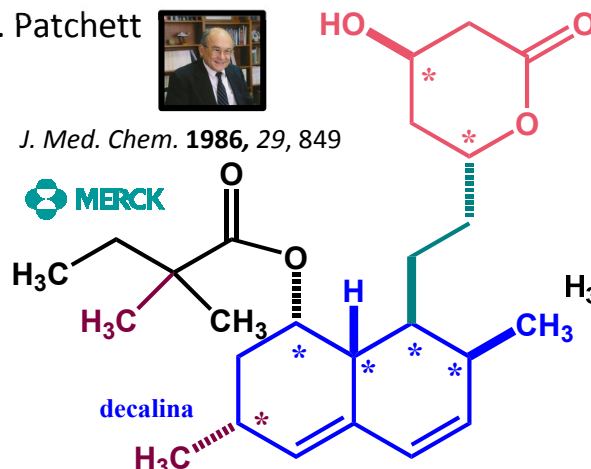
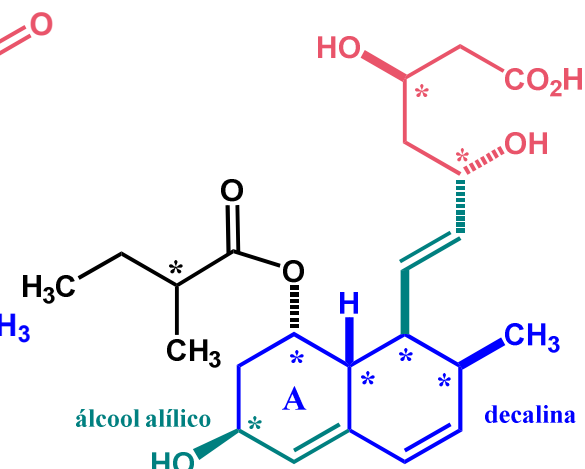
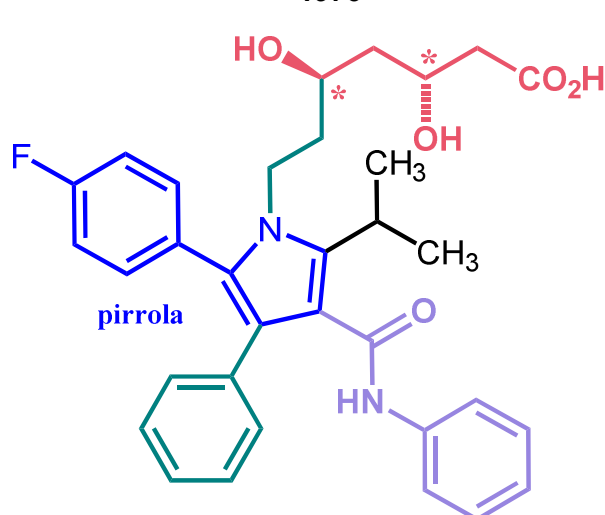


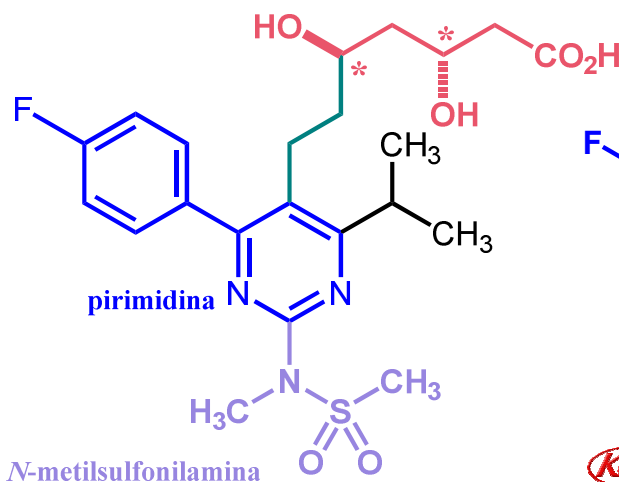
\* A Endo, A gift from nature: the birth of the statins, *Nature Medicine* 2008, 14, 1050

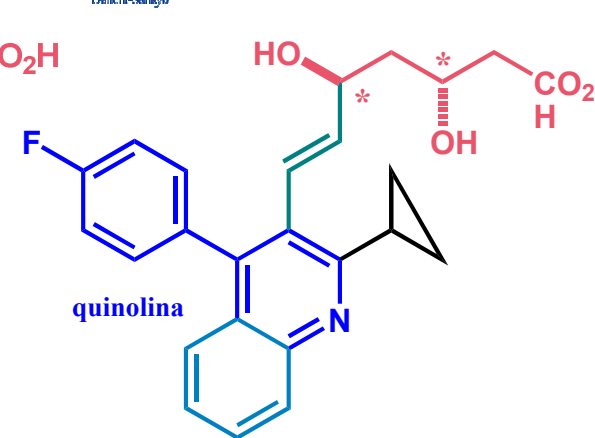


# A descoberta das estatinas

1976 - Arthur A. Patchett


*J. Med. Chem.* **1986**, 29, 849

**compactina**  
1976

**simvastatina**  
1986

**pravastatina**  
1988


**atorvastatina**  
1991


**rosuvastatina**  
2004

**pitavastatina**  
2009

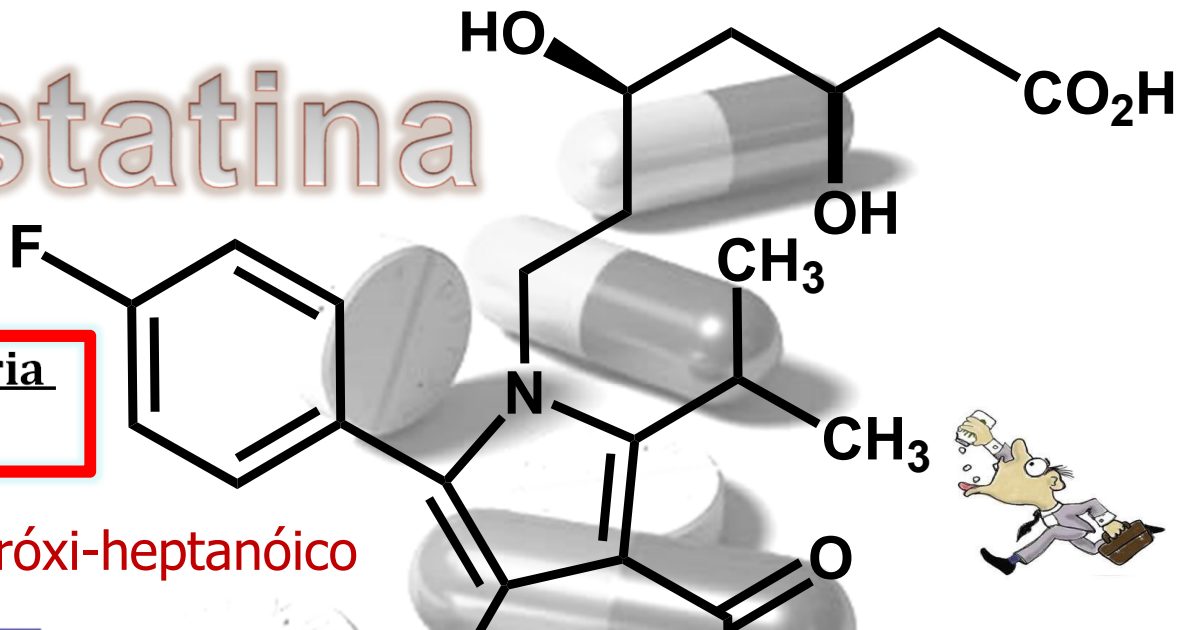
**"...In 2009, statins were used to treat ca. 30 million people."**



# Atorvastatina

**Maior *bestseller* da história dos fármacos**

ácido (*N*-pirrol)-3,5-di-hidróxi-heptanóico



1991

Organic & Biomolecular Chemistry

Qu  
Me

Warn  
Park I

PAPER



View Article Online  
View Journal | View Issue



Bruce D. Roth  
2013 SCI Perkin Me

B. D. Roth, *Progr. Med.*  
B. D. Roth, et al., *J. Med*



Cite this: *Org. Biomol. Chem.*, 2016, 14, 2291

Received 12th December 2015,  
Accepted 29th December 2015

DOI: 10.1039/c5ob02546j

www.rsc.org/obc

## The total synthesis of calcium atorvastatin†

Luiz C. Dias,<sup>\*a</sup> Adriano S. Vieira<sup>a</sup> and Eliezer J. Barreiro<sup>b</sup>

A practical and convergent asymmetric route to calcium atorvastatin (**1**) is reported. The synthesis of calcium atorvastatin (**1**) was performed using the remote 1,5-*anti* asymmetric induction in the boron-mediated aldol reaction of  $\beta$ -alkoxy methylketone (**4**) with pyrrolic aldehyde (**3**) as a key step. Calcium atorvastatin was obtained from aldehyde (**3**) after 6 steps, with a 41% overall yield.

Total de Vendas = ca. US\$ 150 bilhões (1991–2011)

# Tinibes: inibidores de TK's

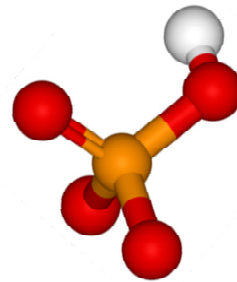
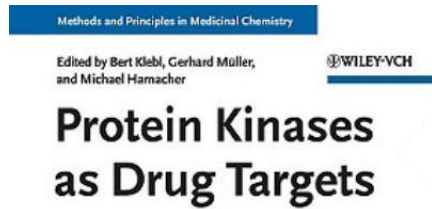
Edwin G Krebs  
(1918 –2009)



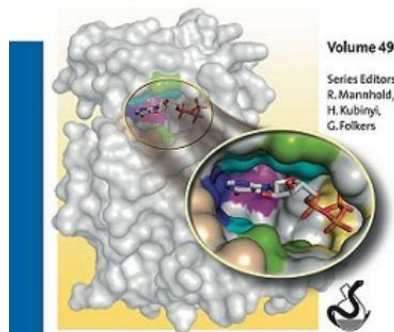
1992



Edmond H Fischer  
(1920)



kinoma

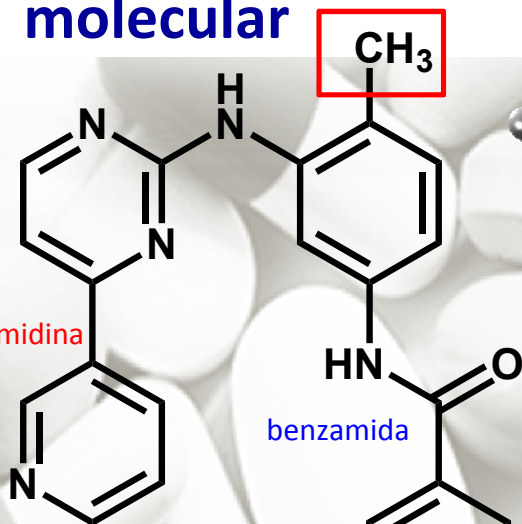


K Nawaz, RM Webster, The non-small-cell lung cancer drug market *Nature Rev. Drug Discov.* **2016**, *15*, 229;  
S. Aggarwal, Targeted cancer therapies, *Nature Rev. Drug Discov.* **2010**, *9*, 427; P. Cohen, Timeline: Protein kinases — the major drug targets of the twenty-first century? *Nature Rev. Drug Discov.* **2002**, *1*, 309.

## Novo padrão molecular

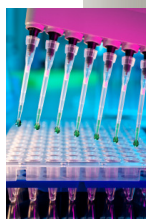
2001

2-fenilaminopirimidina



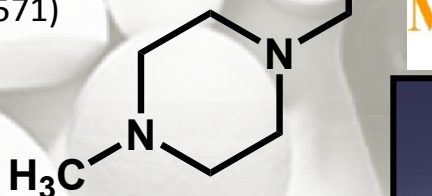
benzamida

High throughput screening



imatinibe  
(STI571)

therapeutic  
innovation

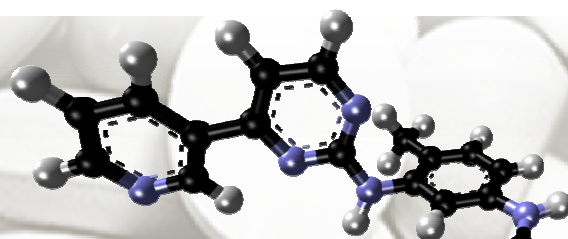


Ciba-Geigy

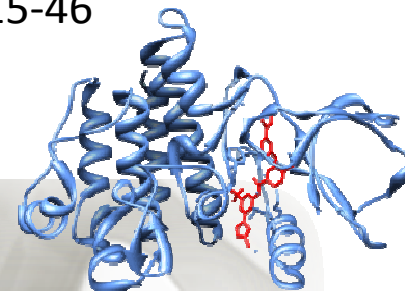
NOVARTIS



Leucemia mieloide  
crônica  
(CML)

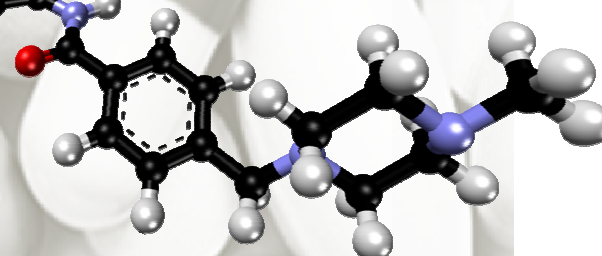


Estrutura cristalina  
da Abl cinase



Mult-target drug

Abril 1992  
tyrosine-kinase  
inhibitor



Química  
med  
Medicinal  
chem

1988 – Nicholas Lydon, Brian J. Druker & Charles L Sawyers &

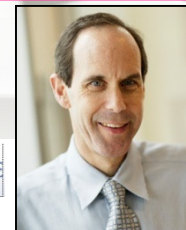
1995 - Compound STI571 ++

2001 – Imatinib (Gleevec<sup>R</sup>, [Novartis](#))[[link](#)]

WW: US\$ ca. 5.1 bi (2014)



Nicholas B. Lydon  
Blueprint Medicines Inc\*



Brian J. Druker\*  
Blueprint Medicines Inc



Charles L. Sawyers\*\*  
Blueprint Medicines Inc

& 2009 - Lasker Foundation Clinical Award (*J. Clin. Invest.* **2009**, *119*, 2863)

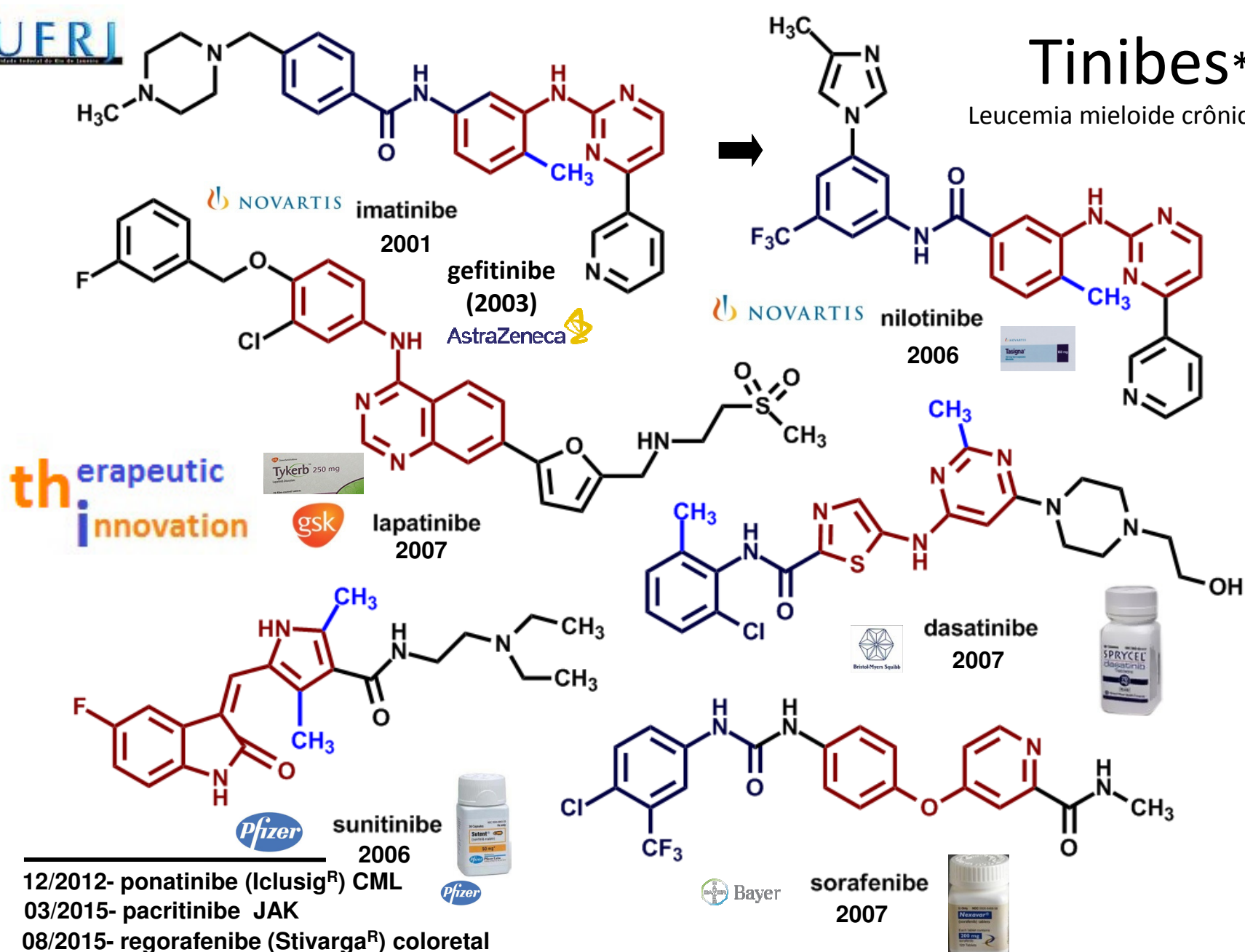
\* B. J. Druker has been awarded with the 2012 Japan Prize in Healthcare and Medical Technology;

\*\* C. L. Sawyers was named in 2011, Thomson Reuters Citation Laureate in Medicine;



# Tinibes\*

Leucemia mieloide crônica (CML)



12/2012- ponatinibe (Iclusig<sup>®</sup>) CML  
 03/2015- pacritinibe JAK  
 08/2015- regorafenibe (Stivarga<sup>®</sup>) coloretal  
 02/2016 – imatinibe genérico (US2/pill)

• Mercado mundial em 2014: US\$ 20,2 bi\*

\* C. W. Lindsley, *ACS Med. Chem. Lett.* **2014**, 5, 1066

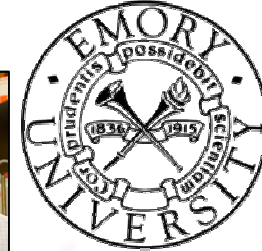
# Recente Inovação em Fármacos

- 1998 – Pharmasset Inc., New Jersey, EUA



medicinal chemistry

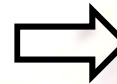
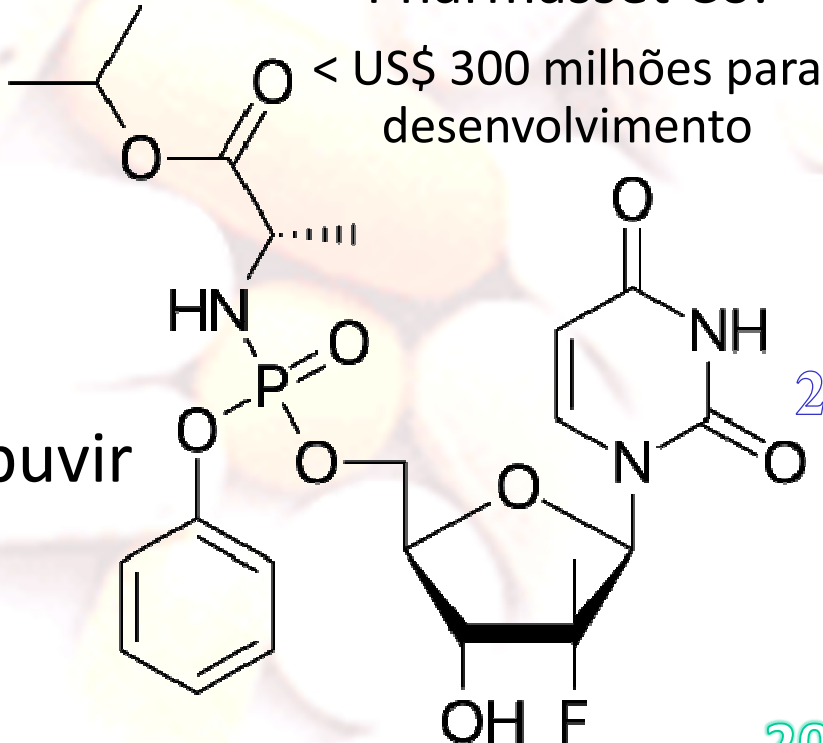
Professor Raymond F. Schinazi  
& Professor Dennis C. Liotta



School of Medicine

Department of Chemistry

Pharmasset Co.



Fundada em 1987, Foster City,  
Califórnia, EUA

Compra a Pharmasset Co.  
em novembro de 2011  
por US\$ **11,2 bilhões**

2013

Sofosbuvir



ANVISA: 30 de mar de 2015

2013-5 BLOCKBUSTER DRUG  
US\$ 20,7 billion

# Recente Inovação em Fármacos, contribuição da academia

## TOP 10 PRODUCTS

DRUG NAME	TYPE	MARKETER	INDICATION	ESTIMATED 2015 SALE (\$ BILLIONS)	% CHANGE FROM 2014
Humira	Antibody	AbbVie, Eisai	Inflammation	14.2	13
Ledipasvir/sofosbuvir	Small molecule	Gilead Sciences	Hepatitis C	13.9	554
Enbrel	Protein	Amgen, Pfizer, Takeda	Inflammation	8.7	-2
Remicade	Antibody	Janssen, Merck & Co.	Inflammation	8.3	-10
Rituxan	Antibody	Roche	Cancer	7.0	3
Lantus	Peptide	Sanofi	Diabetes	6.9	2
Avastin	Antibody	Roche, Chugai	Cancer	6.6	4
Herceptin	Antibody	Roche	Cancer	6.5	5
Sitagliptin	Small molecule	Merck & Co.	Diabetes	6.2	3
fluticasone/salmeterol	Small molecule	GlaxoSmithKline	Asthma	5.7	-11

**NOTE:** Foreign currencies converted at current exchange rates.

**SOURCES:** Company data, stock analysts, and C&EN estimates

C&EN 2015, Dec 07, 14

**O mercado farmacêutico mundial em 2015: US\$ 921 bi**



Universidade Federal do Rio de Janeiro

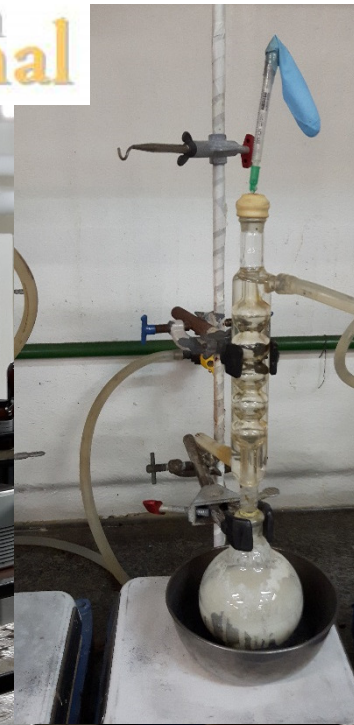


# CASSBIO

Laboratório de Avaliação e Síntese de Substâncias Bioativas

Cidade Universitária, ilha do Fundão,  
Rio de Janeiro, RJ

Química Medicinal



Criado em 19/04/1994 Laboratório de Avaliação e Síntese de Substâncias Bioativas



Bioensaios  
**Bioensaios**

*Rev. Virtual Quim.* 2013, 5, 266.



A quimioteca com  
2012 moléculas  
bioativas.



**Molecular  
Modelagem**



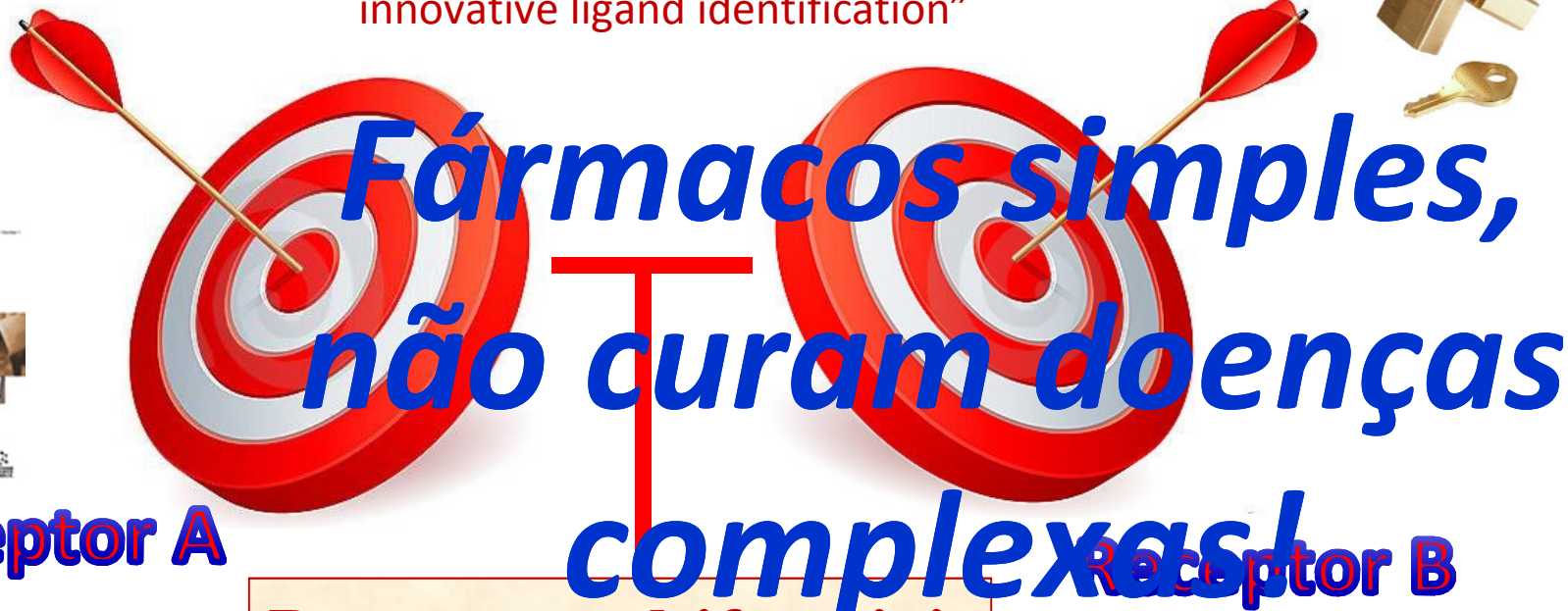
# FÁRMACOS DO SÉCULO 21



# Novo paradigma do século 21

“Multi-target paradigm for innovative ligand identification”

Química  
med  
Medicina  
chem



Receptor A

Receptor B

Doenças multifatoriais



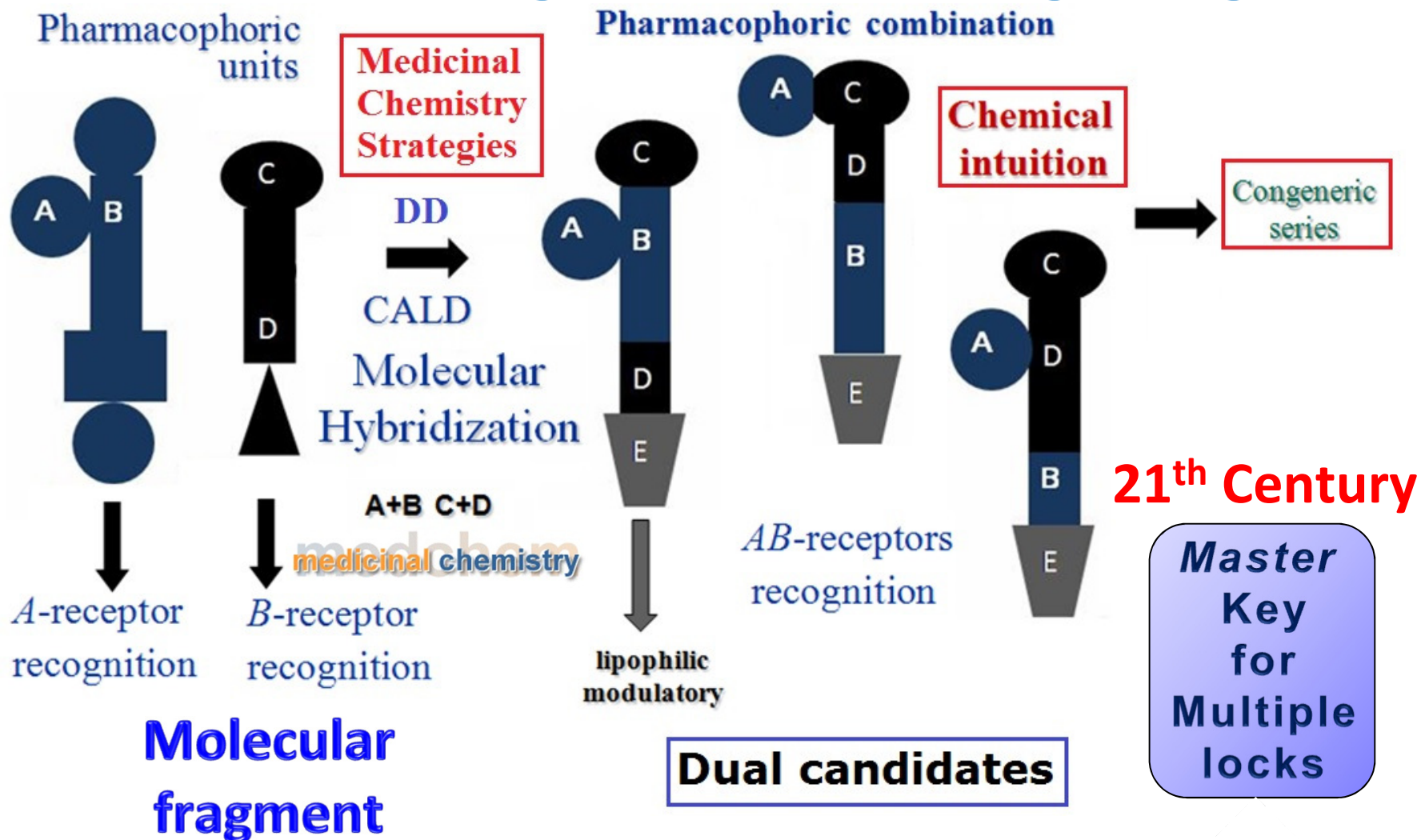
O desenho racional de fármacos *multi-alvos* depende da capacidade de se combinarem fragmentos moleculares farmacofóricos, capazes de assegurarem o reconhecimento molecular pelos receptores envolvidos.

M L Bolognesi, A Cavalli, **Multitarget Drug Discovery** and Polypharmacology, *ChemMedChem* **2016**, *11*, 1190; A Anighoro et al., **Polypharmacology: challenges and opportunities in drug discovery**, *J. Med. Chem.* **2014**, *57*, 7874; JL Medina-Franco et al. Shifting from the single to the **multitarget paradigm** in drug discovery, *Drug Discov. Today* **2013**, *18*, 495; C Hiller, J Kühhorn, P Gmeiner, Class A G-Protein-Coupled Receptor (GPCR) Dimers and Bivalent Ligands, *J. Med. Chem.* **2013**, *56*, 6542; G Phillips, M Salmon, **Bifunctional compounds** for the treatment of COPD, *Annu. Rev. Med. Chem.* **2012**, *47*, 209; JR Morphy, CJ Harris, Eds., *Designing multi-target drugs*, RSC Publishing, 2012; E J Barreiro, C A M Fraga, **New insights for multifactorial diseases therapy...**, *Curr Drug Therapy* **2008**, *3*, 1; K Strebhardt, A Ullrich, Paul Ehrlich's magic bullet concept: 100 years of progress. *Nat Rev Cancer* **2008**, *8*, 473.





# Rational design of multi target ligand

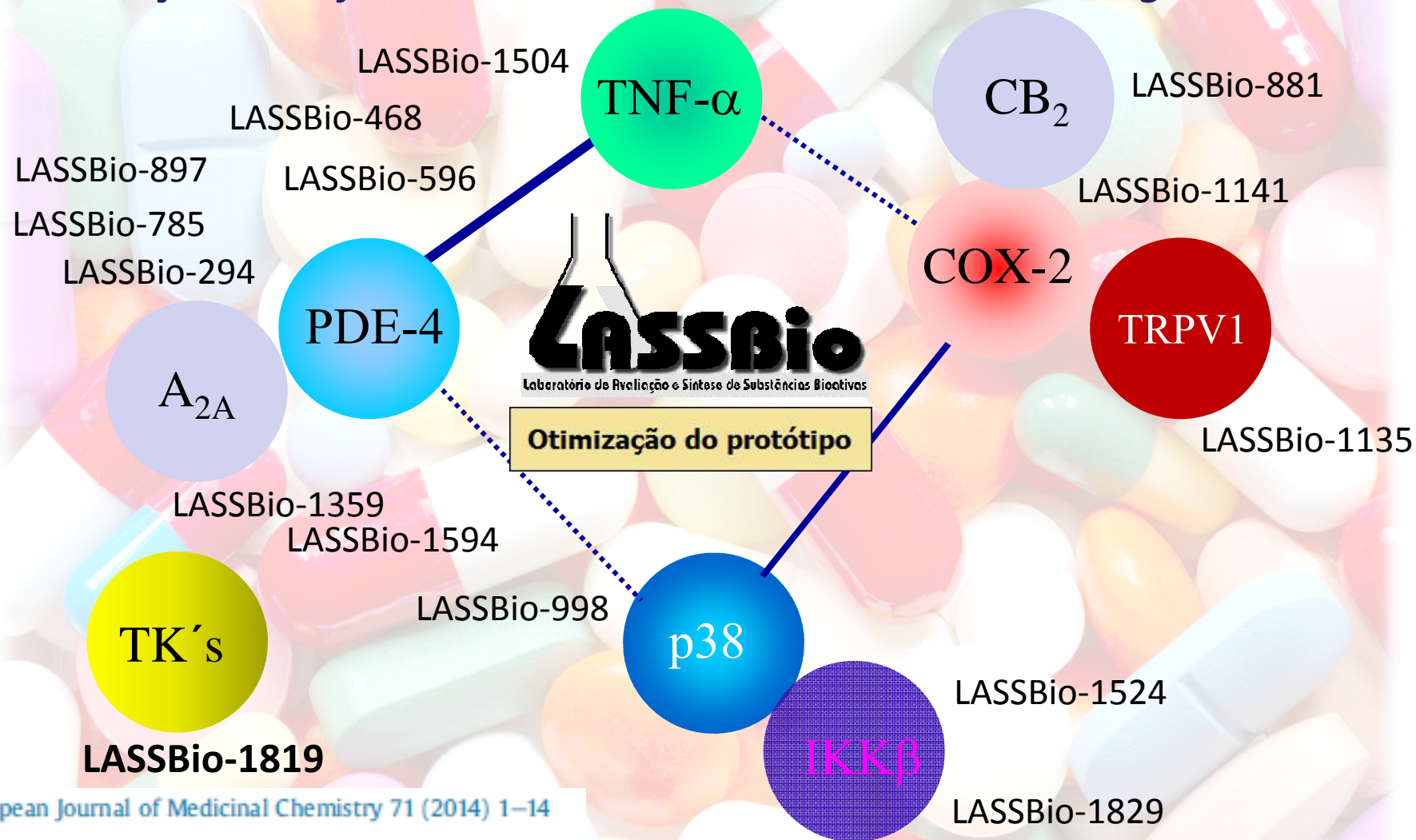


C Viegas-Jr, A Danuello, VS Bolzani, E J Barreiro, CAM Fraga, *Molecular Hybridization: A useful tool in the design of new drug prototypes*, *Curr. Med. Chem.* **2007**, *14*, 1829



# Novos protótipos de fármacos multialvos

*Doenças multifatoriais não-transmissíveis crônico-degenerativas*



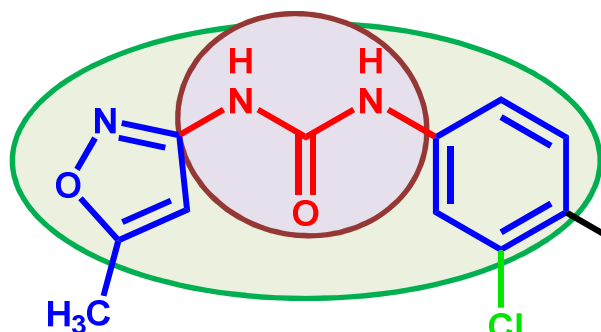
European Journal of Medicinal Chemistry 71 (2014) 1–14

**Chimeric receptor model**

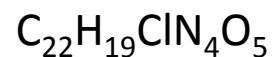
medchem  
**Química Medicinal**

# Dual Ligand Design

Ligand for target-1: **VEGFR-2**



Tivozanibe



VEGFR-1 = 30 nM

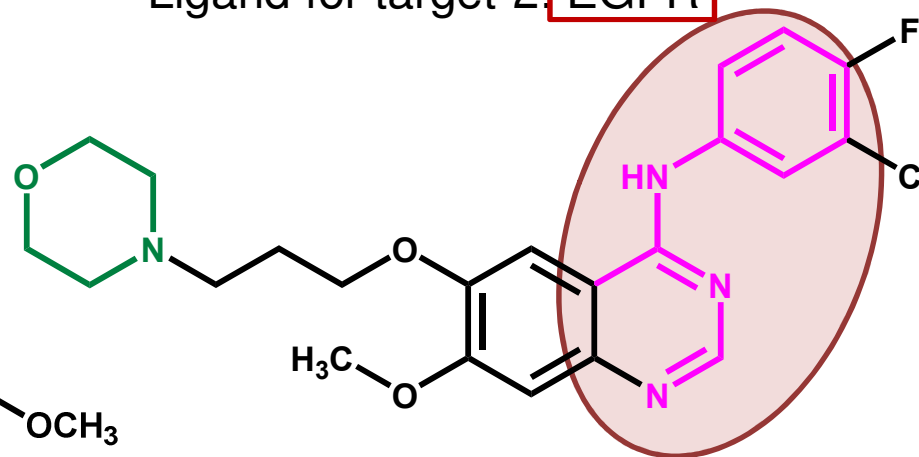
VEGFR-2 = 6,5 nM

VEGFR-3 = 15 nM

VEGFR tyrosine kinase inhibitor

2016

Ligand for target-2: **EGFR**



Gefitinibe



EGFR = 33 nM

2003



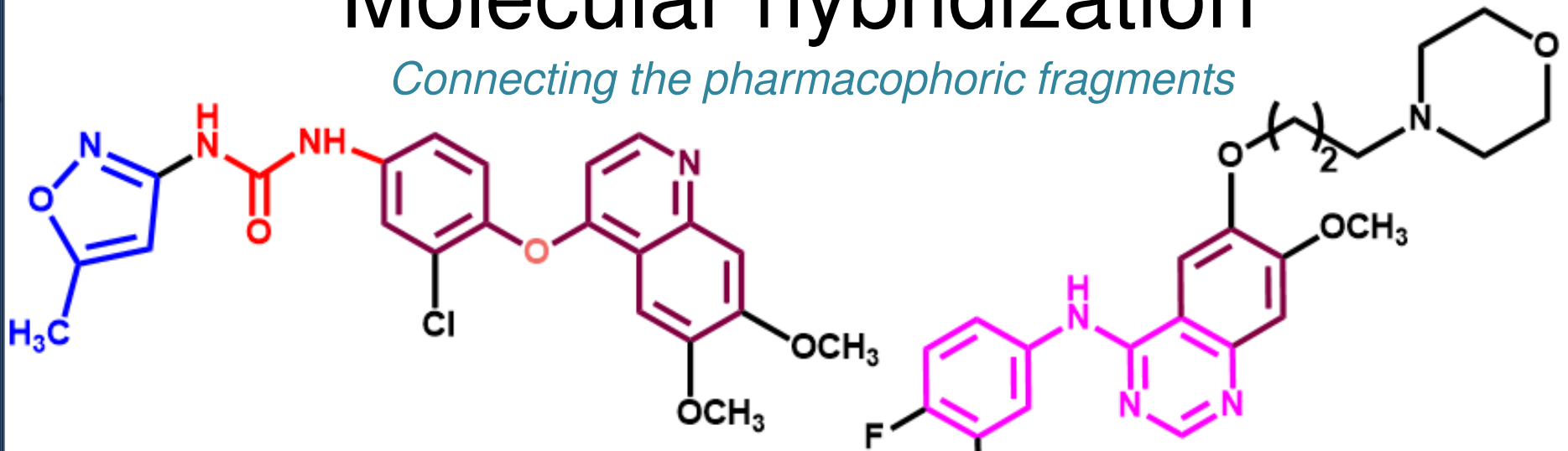
Originalidade

**Dissecação Molecular:** identificação de fragmentos farmacofóricos



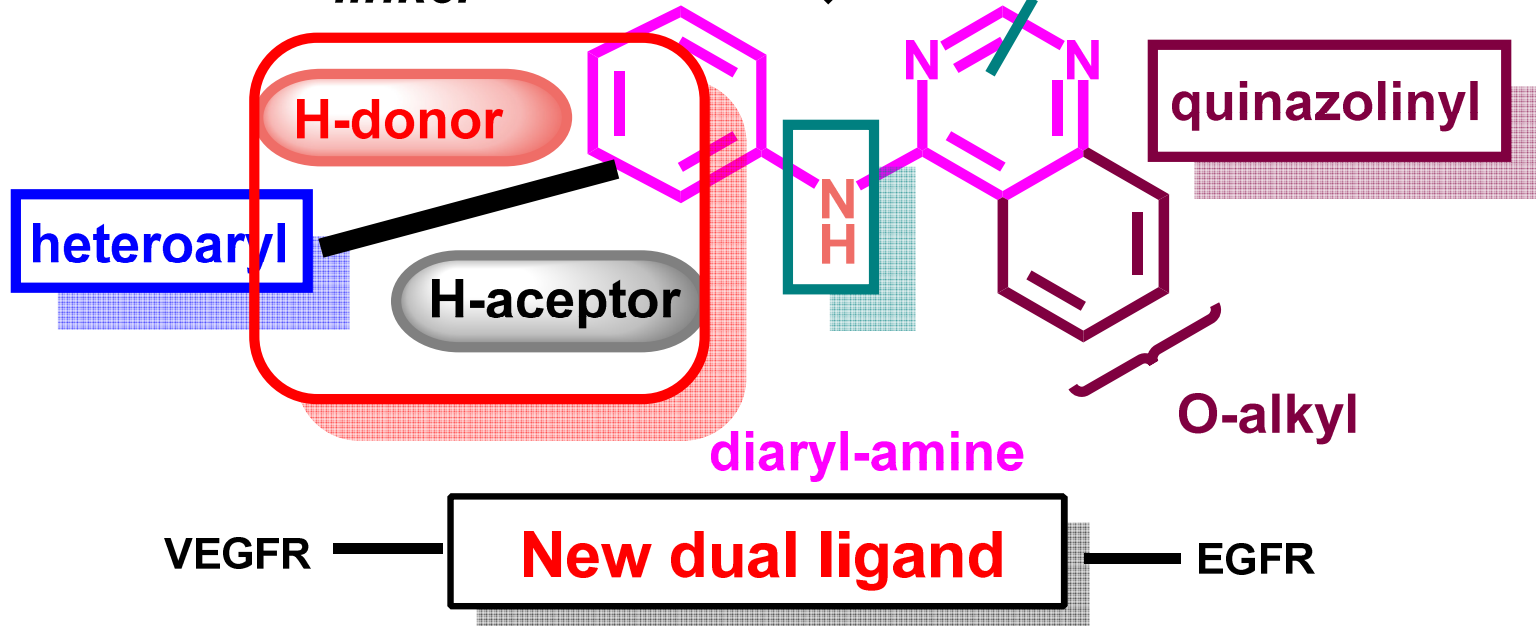
# Molecular hybridization

*Connecting the pharmacophoric fragments*



tivozanib  
VEGFR  
*linker*

gefitinib  
EGFR



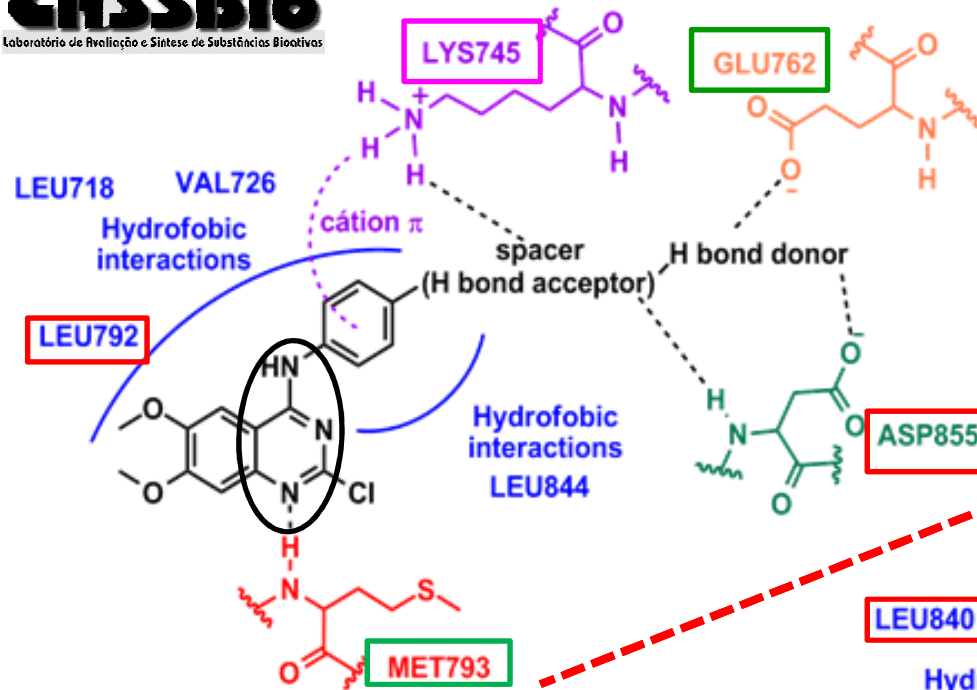
# Molecular design of new dual kinase inhibitor

*From molecular docking studies*

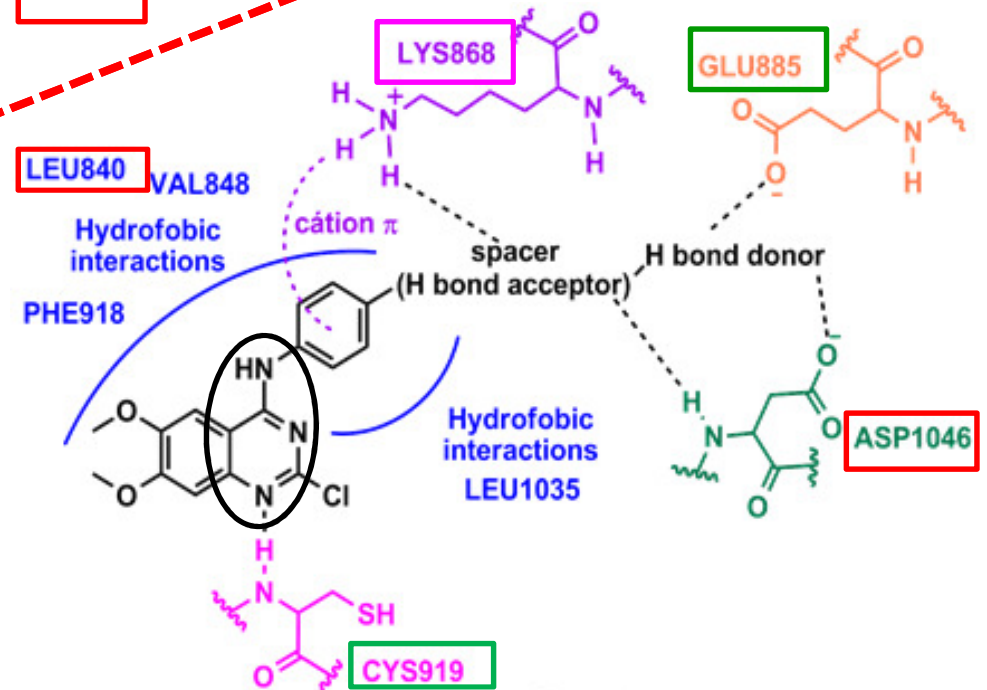
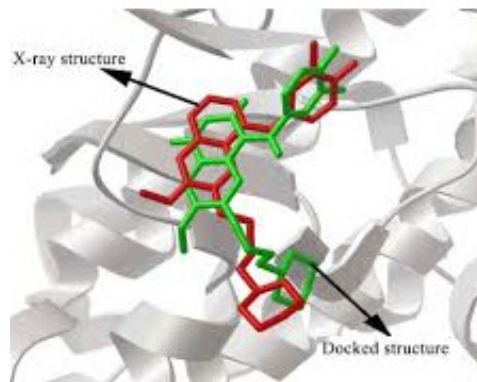


medicinal chemistry

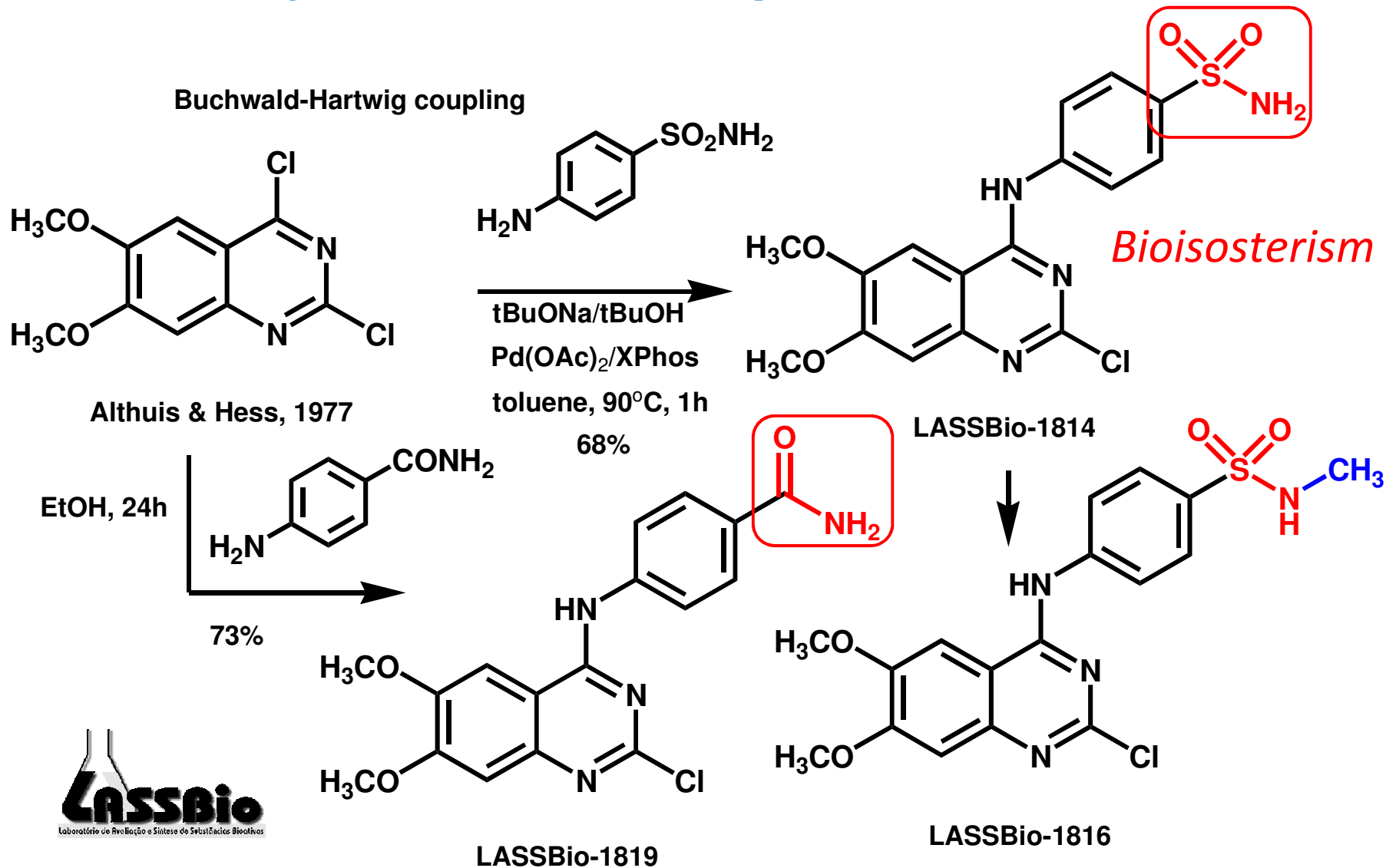
**Pharmacophoric map**



EGFR



VEGFR-2



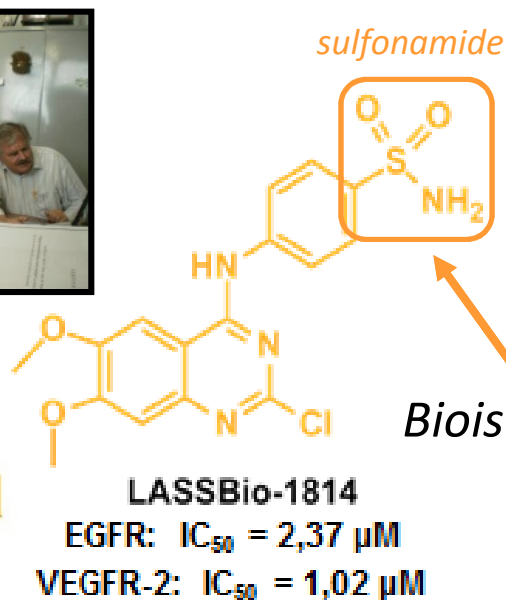
High overall yield; accessible starting material; scaleable-up



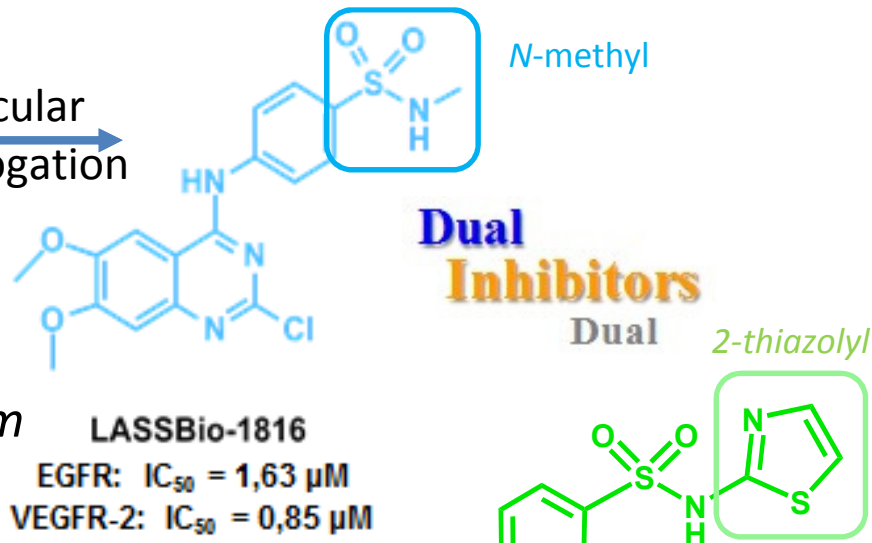
# Binding assay of the congener series



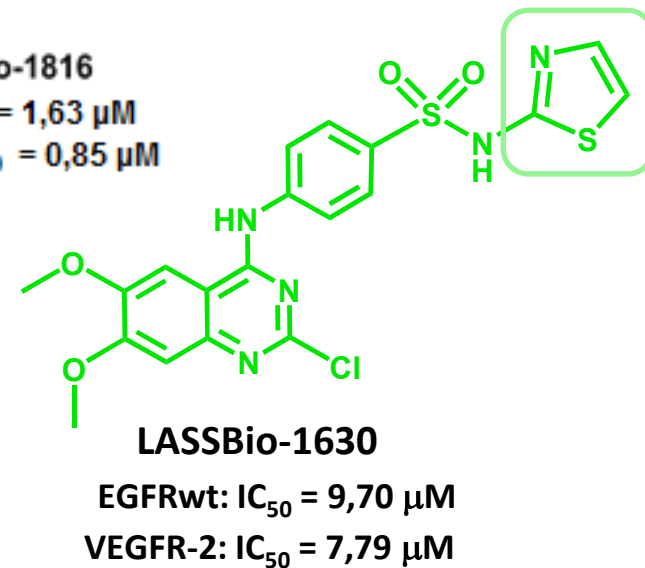
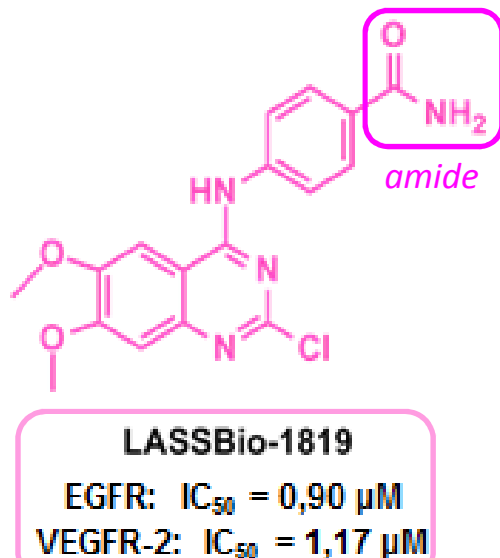
**Química**  
med  
**Medicinal**  
chem



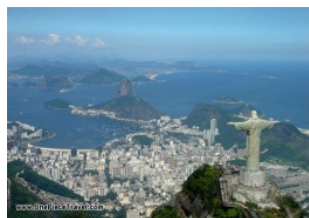
Molecular  
homologation

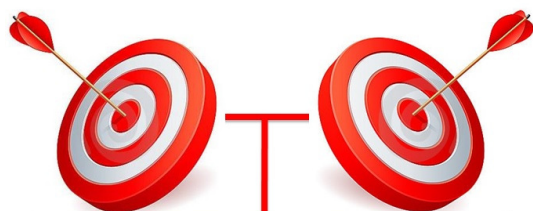


*Bioisosterism*



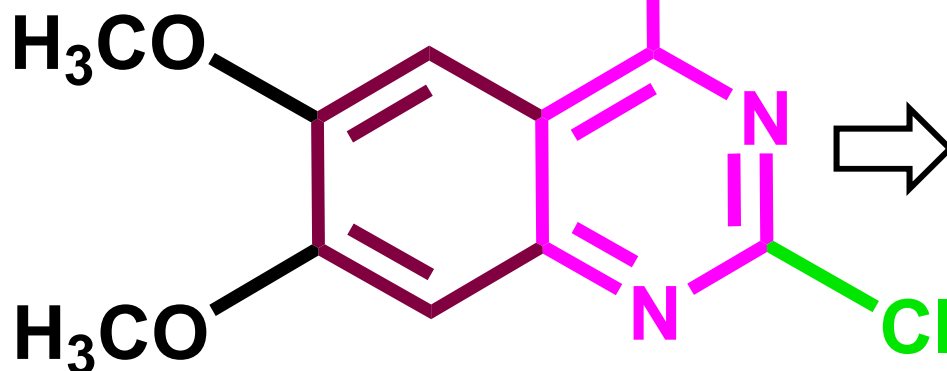
ca. **16** new compounds  
in the congener series





**Dual Inhibitor**  
Dual

medicinal chemistry



**Dual**

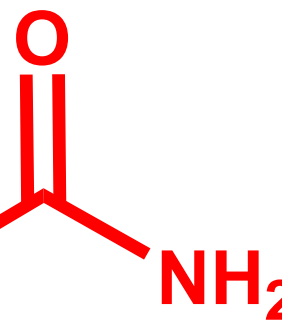
**Ligand**  
Dual

$C_{17}H_{15}ClN_4O_3$

**LASSBio-1819**

Isosteric replacement

carboxamide

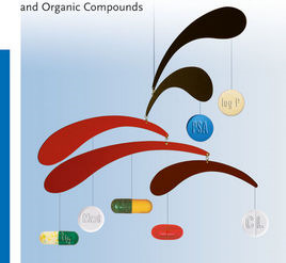


Florencio Zaragoza Dörwald

WILEY-VCH

Lead Optimization for Medicinal Chemists

Pharmacokinetic Properties of Functional Groups and Organic Compounds



**Dual kinase activity**

EGFRwt  $IC_{50} = 0,90 \mu M$

VEGFR-2  $IC_{50} = 1,17 \mu M$



**Novel molecular pattern**

**Lead Optimization**

M. L. C. Barbosa, *Novos derivados quinazolinicos funcionalizados inibidores duais das tirosina cinases receptoras EGFR & VEGFR-2*, Tese Dr, Instituto de Química, UFRJ, 2013.

**PEDIDO INTERNACIONAL PUBLICADO SOB O TRATADO DE COOPERAÇÃO EM MATÉRIA DE PATENTES  
(PCT)**

**(19) Organização Mundial da  
Propriedade Intelectual  
Secretaria Internacional**



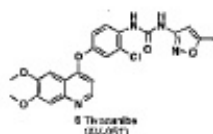
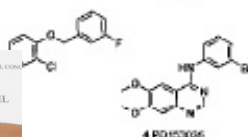
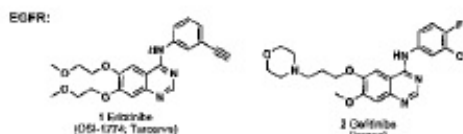
**(10) Número de Publicação Internacional  
WO 2014/113859 A1**

**(43) Data de Publicação Internacional  
31 de Julho de 2014 (31.07.2014) WIPO | PCT**

**(54) Title :** 2-CHLORO-4-ANILINO-QUINAZOLINE COMPOUNDS INHIBITING PROTEIN TYROSINE KINASES, PHARMACEUTICAL COMPOSITIONS COMPRISING THE SAME, METHOD FOR PRODUCING THE SAME AND TYROSINE KINASE INHIBITION METHOD

**(54) Título :** COMPOSTOS 2-CLORO-4-ANILINO-QUINAZOLINICOS INIBIDORES DE PROTEÍNAS TIROSINA CINASES, COMPOSIÇÕES FARMACÊUTICAS COMPREENDENDO OS MESMOS, PROCESSO PARA SUA PRODUÇÃO E MÉTODO PARA INIBIÇÃO DE TIROSINA CINASES

**(57) Abstract :** The present invention relates to 2-chloro-4-anilino-quinazoline derivatives with EGFR and/or VEGFR-2 protein tyrosine kinase inhibiting activity, to anti-tumour pharmaceutical compositions that comprise said compounds, and to methods for producing the same. The present invention further provides a method for treating solid tumours by inhibition of tyrosine kinases.




**(72) Inventores :** BARREIRO, Eliezer Jesus;  
DE CASTRO BARBOSA, Maria Letícia;  
MOREIRA LIMA, Lidia;  
LAUFER, Stefan, Andreas;  
RABELLO SANT'ANNA, Carlos Mauricio;  
TESCH, Roberta;

Patent







Sample Issue

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Lead Optimization



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Original article Volume 71, 7 January 2014, Pages 1–14

## Novel 2-chloro-4-anilino-quinazoline derivatives as EGFR and VEGFR-2 dual inhibitors

Maria Letícia de Castro Barbosa<sup>a,b</sup>, Lídia Moreira Lima<sup>a,b</sup>, Roberta Tesch<sup>a</sup>, Carlos Mauricio R. Sant'Anna<sup>c</sup>, Frank Totzke<sup>d</sup>, Michael H.G. Kubbutat<sup>d</sup>, Christoph Schächtele<sup>d</sup>, Stefan A. Laufer<sup>e</sup>, Eliezer J. Barreiro<sup>a,b</sup>

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Maria Letícia de Castro Barbosa | Lídia Moreira Lima | ...
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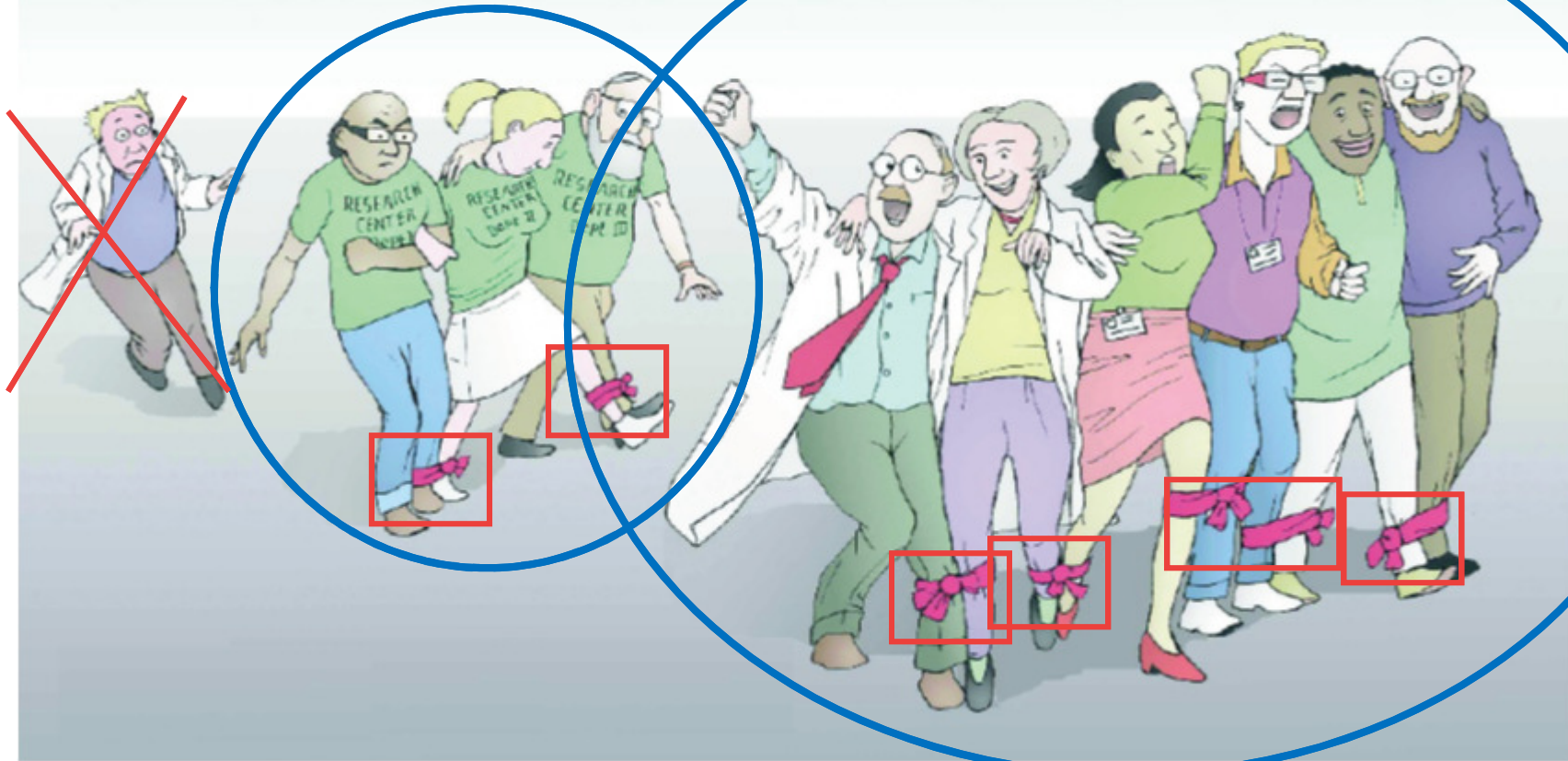
\* Top-1 Poster Prize in **Frontiers in Medicinal Chemistry Congress, 2014, Tübingen, GE**

J. Whitfield, *Nature* **2008**, 455, 720

NEWS FEATURE

NATURE [Vol 455] 9 October 2008

## What makes a successful research team?



W Masona, D J Watts, Collaborative learning in networks, *PNAS* **2012**, 109, 764; M Williams, Productivity Shortfalls in Drug Discovery: Contributions from the Preclinical Sciences?, *JPET* **2011**, 336, 3; R Guimera, B Uzzi, J Spiro, L A N Amaral, Team Assembly Mechanisms Determine Collaboration Network Structure and Team Performance, *Science* **2005**, 308, 697.





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Sábado, 23 de Julho de 2016



Projeto CNPq nº 573.564/2008-6 « » FAPERJ nº E-26/170.020/2008

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# BRAZMEDCHEM 2016

## NOVEMBER 27-30

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RIO DE JANEIRO



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THE 8<sup>TH</sup> IS COMING UP WITH NEW TRENDS  
IN MEDICINAL CHEMISTRY

BrazMedChem 2016

ESPACO TRANSPARÊNCIA

Confira o trabalho realizado  
pela equipe





# Parceiros INCT-*IMO*FAR



Química Medicinal

**LASSBio**

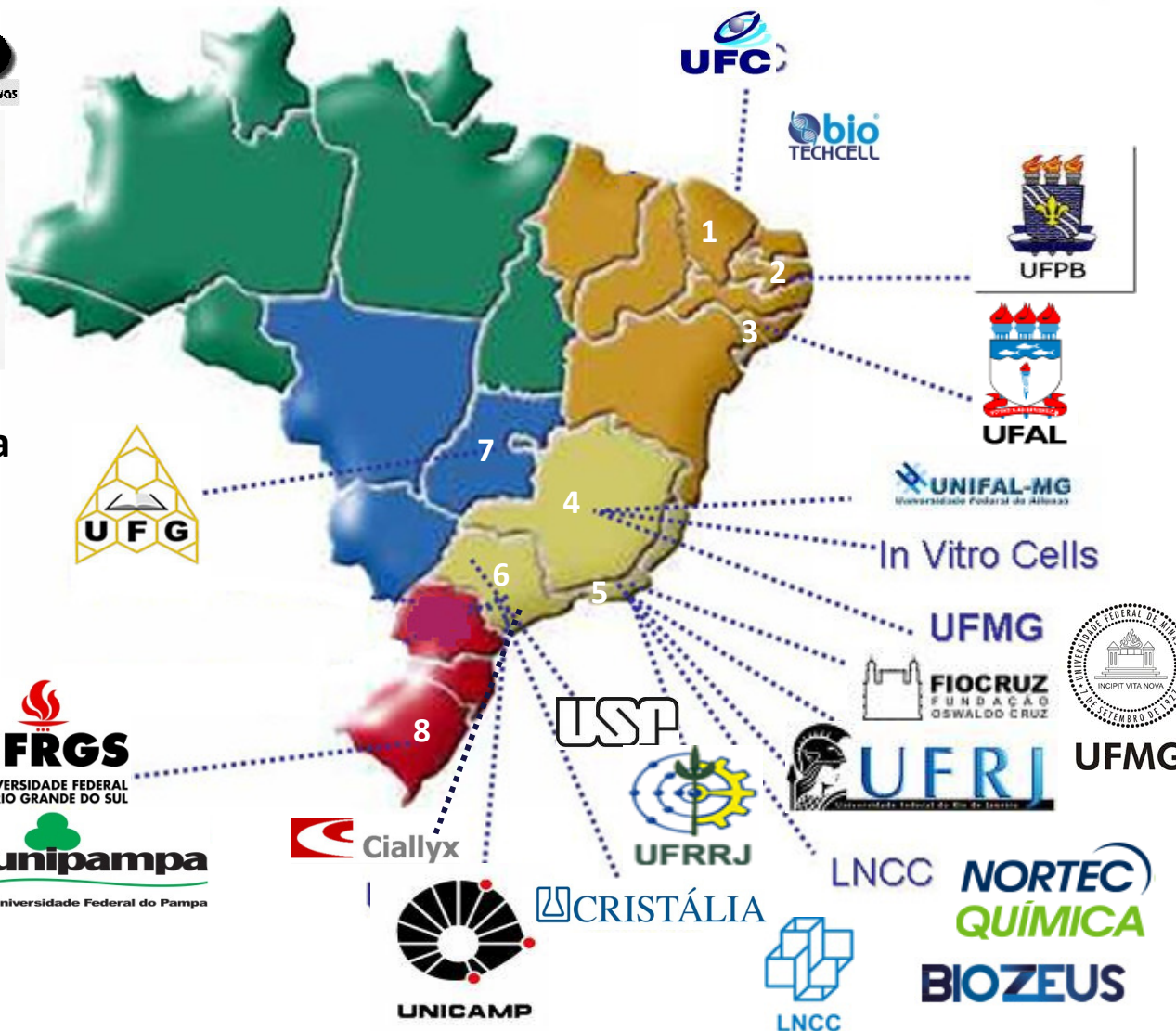
Laboratório de Avaliação e Síntese de Substâncias Bioativas



16 ICT's

30 grupos de pesquisa

5 empresas



# Radical *Innovation*

medchem

Studies of anti-inflammatory & analgesic effect of LASSBio-591, a new candidate of AIA drug.

LASSBio-UFRJ/FM-USP,RP

Studies of new oncolytic agent, dual inhibitor of kinases

LASSBio-UFRJ

WO2014113859

WO2013142935

Studies on design & discovery of new antidiabetes drug candidates.

LASSBio-UFRJ

BR102013012646-2

Discovery of novel anticancer drug candidates

LASSBio-UFRJ / FM-UFC

PCT/BR2013/000095

WO2013142935 A1

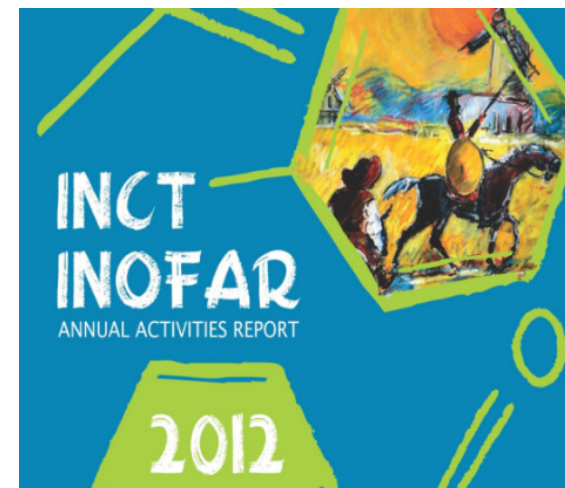
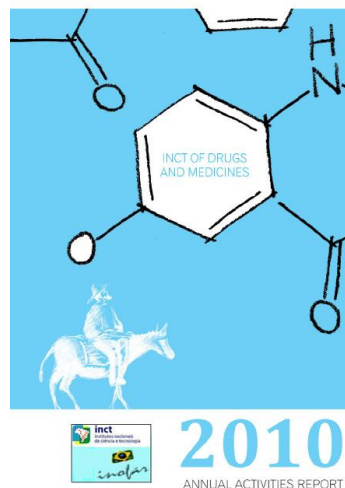
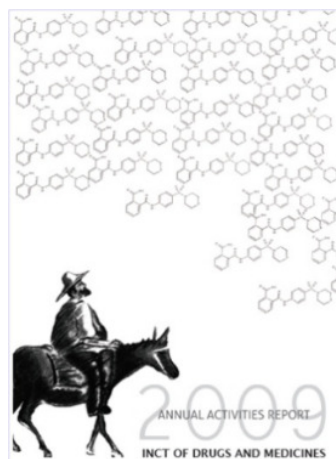


Studies on new drug candidate useful for neuropatic pain

LASSBio-UFRJ / FM-USP, RP

WO2012054996

# Annual Activities Report





Considerações finais

**A** *Química*  
*Medicinal*  
**é simplesmente**  
*fascinante!*



**Considerações finais**

**Os medicamentos**  
**foram uma das**  
*maiores* **invenções**  
**do século 20 !**

**São moléculas que mudam o mundo!**

Convite

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CURSOS



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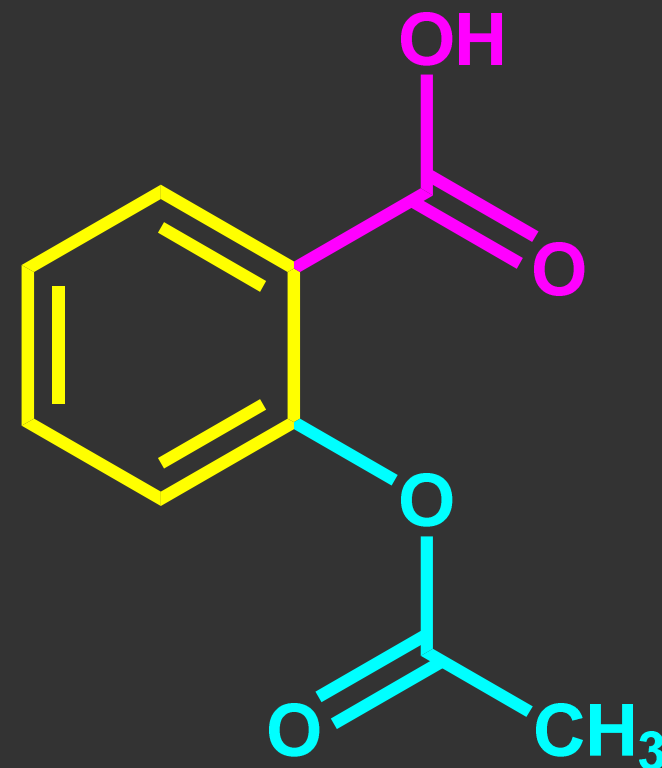
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Pretende-se tratar de temas, opiniões, comentários sobre a Ciência dos Fármacos, seu uso seguro e benefícios. Aspectos da formação qualificada de universitários e pós-graduandos nas Ciências dos Fármacos também são de interesse.



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