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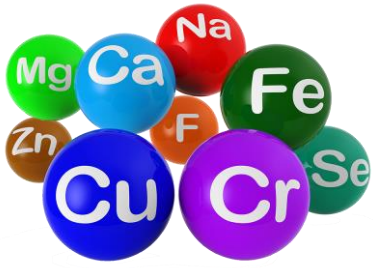

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Aafaq Ur Rehman,
Graduate Research Assistant,
School of Civil and Environmental Engineering

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Upcoming ACS Webinars®

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Thursday, November 19, 2015

"Prodrugs in Drug Discovery"

John Higgins, Senior Principal Scientist and Network Technology Lead, Merck

Nicholas Meanwell, Executive Director, Discovery Chemistry, Bristol-Myers Squibb

Thursday, December 3, 2015

"Chemistry & the Economy: Global Outlook 2016"

Paul Hodges, Chairman of International eChem (IeC)

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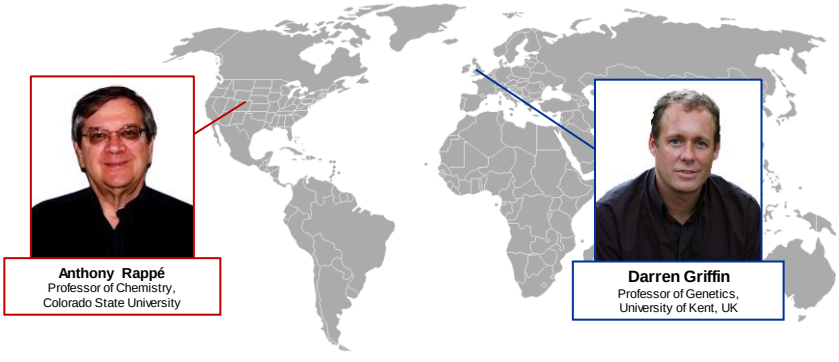
"The Chemistry of Addiction"



Anthony Rappé
Professor of Chemistry,
Colorado State University

Darren Griffin
Professor of Genetics,
University of Kent, UK

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Chemistry of Addiction

VALENTIN OTTONE VIA FLICKR, CREATIVE COMMONS

Photo Credit: Leah Noel (Creative Commons)

"Despite the importance of numerous psychosocial factors, at its core, drug addiction involves a biological process: the ability of repeated exposure to a drug of abuse to induce changes in a vulnerable brain that drive the compulsive seeking and taking of drugs, and loss of control over drug use, that define a state of addiction."

E. J. Nestler "Cellular basis of memory for addiction", *Dialogues Clin Neurosci.* **2013**, *15*, 431–443.

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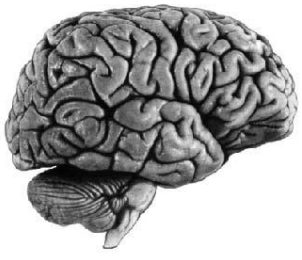
Chemistry of Addiction

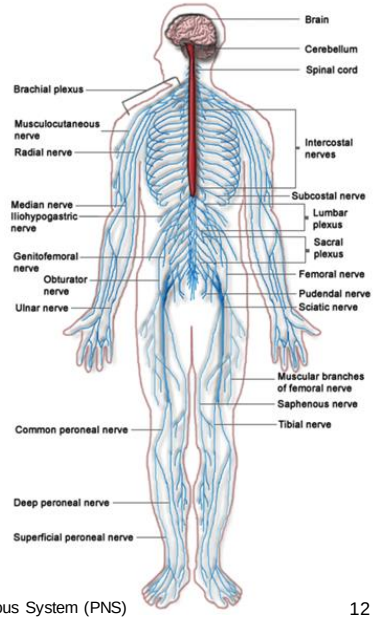
Neuronal synaptic transmission involves:

- neurotransmitter presynaptic release
- receptor binding, binding site release,
- and neurotransmitter degradation/reuptake

Drug molecules "look like" natural substrates bind to receptor, transporter, or enzyme active sites



Central Nervous System (CNS)



Peripherial Nervous System (PNS)

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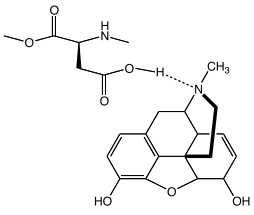
Chemistry of Addiction

hydrogen bonding

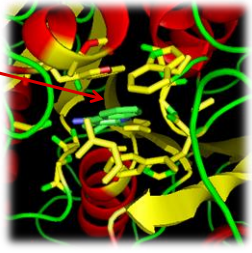


Impact of a drug depends upon:

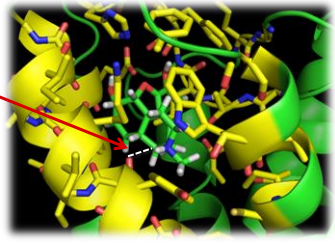
Strength of binding, which depends upon:

- shape & positioning of functional groups
- hydrogen bonding
- salt bridges
- π-stacking
- π-cation interactions
- hydrophobic contacts
- conformational rigidity
- Ability to pass through hydrophobic blood-brain barrier

π-stacking



Salt bridge



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Addiction / Substance Abuse

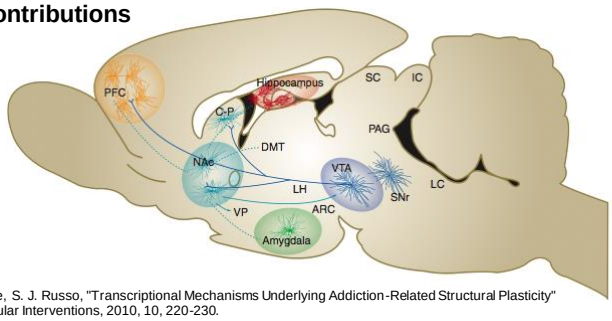
- **Tolerance** = decrease in potency with repeated administration of same dose.

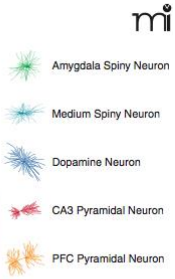
  Cross-tolerance = tolerance to one drug in a class confers tolerance to others in that class.
- **Dependence** = withdrawal symptoms when drug use is terminated.
- **Addiction** = persistent use, even in the face of physical, psychological or social harm.

Psychological factors

Physiological changes

Chemical contributions





I. Maze, S. J. Russo, "Transcriptional Mechanisms Underlying Addiction-Related Structural Plasticity" Molecular Interventions, 2010, 10, 220-230.

14

Drug Tolerance

Progressively decreasing responsiveness to a drug

Three Basic Mechanisms:

1. **Metabolic Tolerance**
   enzyme (e.g. cyt p450) production increased
   leading to greater metabolism, leading to more
   drug needing to be administered for same effect

2. **Cellular-Adaptive** (pharmacodynamic)
   neurons adapt to continued presence of the drug
   either by reducing the number of receptors
   or by decreasing the sensitivity of the receptors to the drug

3. **Behavioral Conditioning**
   tolerance can be induced when a drug is administered
   in the presence of usual predrug cues

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Drugs of Abuse

What makes for the difference between someone who can drink or dabble in illicit drugs without developing dependence (or many negative consequences) **versus someone who becomes an addict?**

- Stress level
- genetic background
- other biological factors
- environment
- social context in which drug use is occurring

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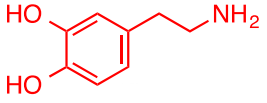
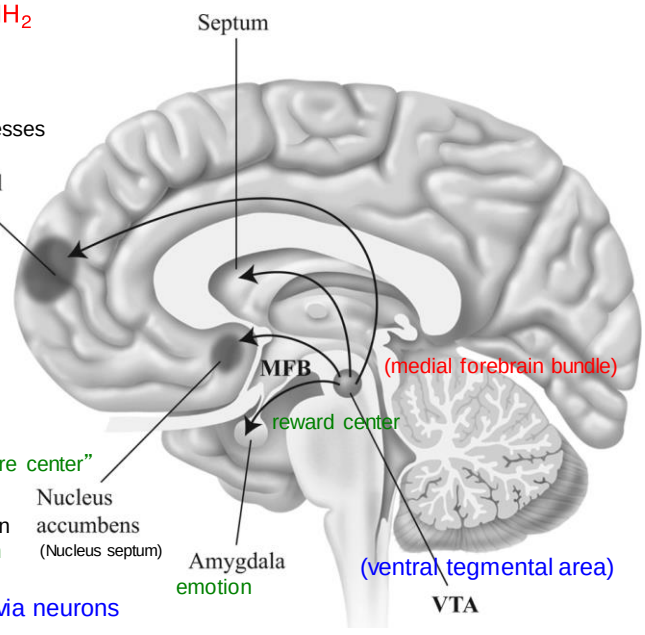
Reward Circuit



Involved in addictive processes
Dopamine rich regions



Involved in planning complex cognitive behaviors

Hypothalamus is not shown
Is also involved in emotion

Signals project outward via neurons

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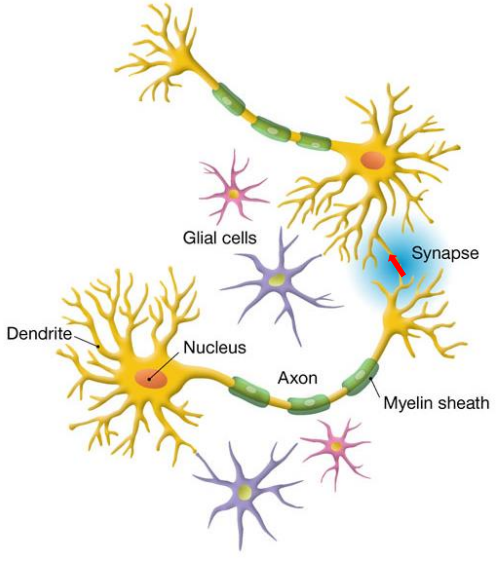
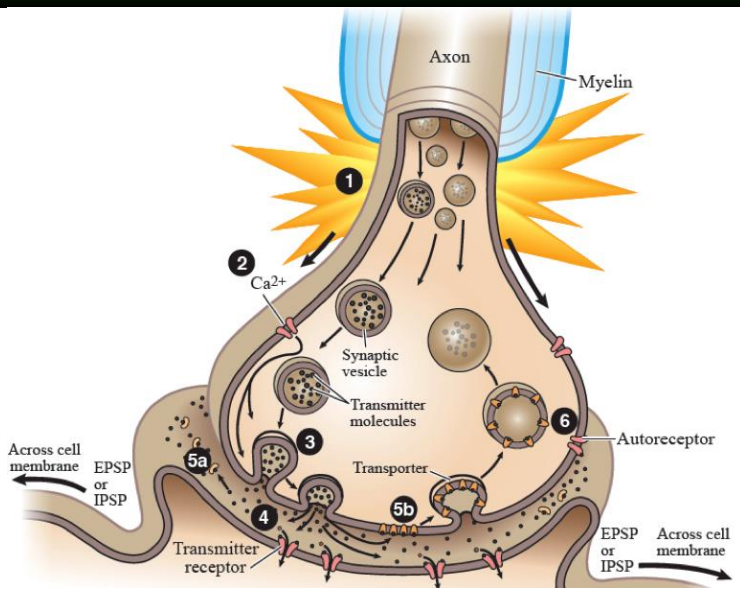
Overview of Synaptic Transmission



Synaptic Transmission



Julien's Primer of Drug Action | Thirteen Edition | Advokat•Comaty•Julien Worth Publishers

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Audience Survey Question

ANSWER THE QUESTION ON BLUE SCREEN IN ONE MOMENT

The glutamate receptor is thought to be responsible for which of the following:

- generic nerve transmission
- inhibitory action
- excitatory transmission (learning)
- reward response
- primary stress nerve transmission

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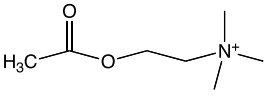
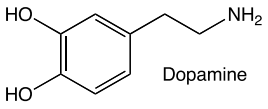
Neurotransmitters

Common



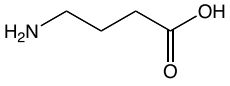
acetylcholine

generic nerve transmission



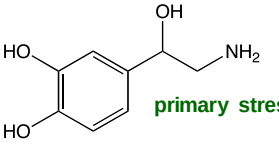
Dopamine

reward response



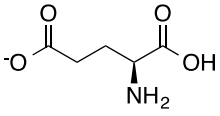
Gamma aminobutyric acid (GABA)

Inhibitory action



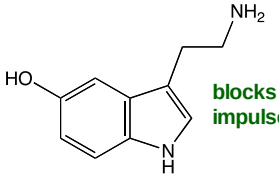
primary stress nerve transmission

Norepinephrine



glutamate

excitatory transmission
(learning)



blocks unneeded nerve impulses

Serotonin

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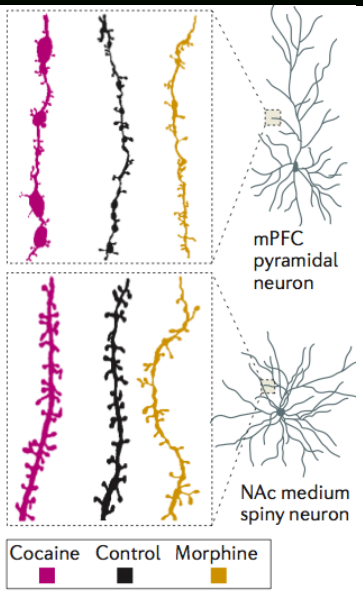
Neuron Nodules

Addictive substances produce structural changes in neurons

(the changes are substance-dependent)

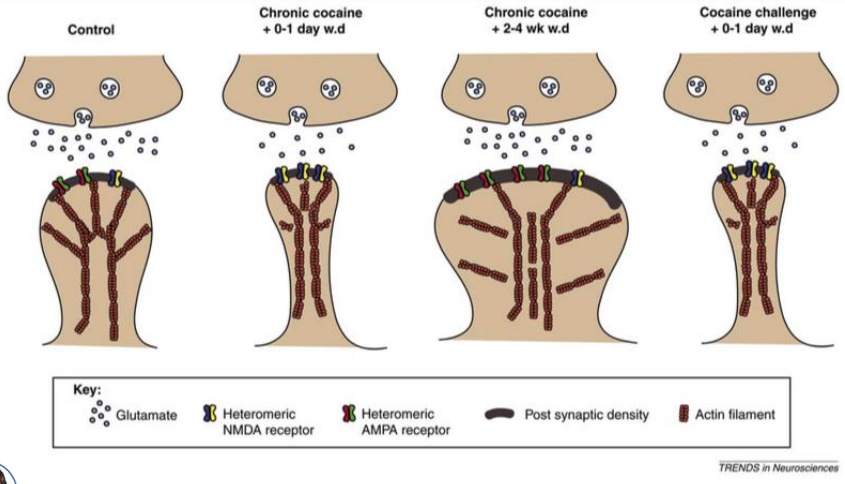


Aldo Badiani, David Belin, David Epstein, Donna Calu and Yavin Shaham "Opiate versus psychostimulant addiction: the differences do matter" Nature Reviews, Neuroscience, 2011, 12, 685-700.

22

Observable Plasticity

Chronic exposure leads to time-dependent reorganization and structure of the of a-amino-3- hydroxyl-5-methyl-4-isoxazole-propionate (AMPA) and N-methyl-D-aspartate (NMDA) glutamate receptors at nucleus accumbens (NAc) medium spiny neuron (MSN) synapses



S. J. Russo, D. M. Dietz, D. Dumitriu, J. H. Morrison,R. C. Malenka, E. J. Nestler "The addicted synapse: mechanisms of synaptic and structural plasticity in nucleus accumbens" *Trends in Neurosciences* **2010**, *33*, 267–276.

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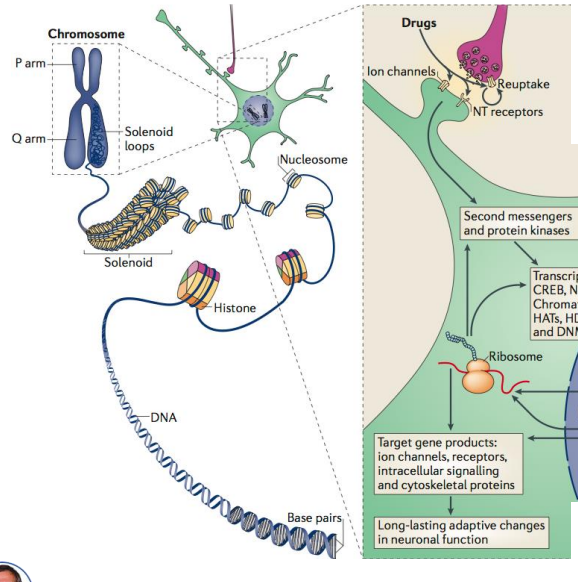
Mechanisms of Transcriptional and Epigenetic Regulation by Drugs of Abuse



Drugs of abuse act through synaptic targets such as reuptake mechanisms, ion channels and neurotransmitter (NT) receptors to alter intracellular signaling cascades.

This leads to activation or inhibition of transcription factors (TFs) and other nuclear targets.

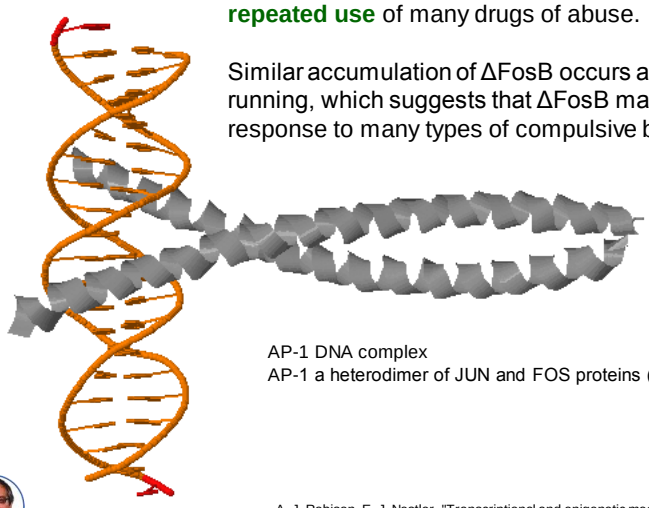
A. J. Robison, E. J. Nestler, "Transcriptional and epigenetic mechanisms of addiction" Nature Reviews, Neuroscience, 2011, 12, 623-635

24

Δ-FosB Transcription Factor

ΔFosB accumulates in nucleus accumbens and dorsal striatum (brain regions important for addiction) after **repeated use** of many drugs of abuse.

Similar accumulation of ΔFosB occurs after compulsive running, which suggests that ΔFosB may accumulate in response to many types of compulsive behaviors.



AP-1 DNA complex
AP-1 a heterodimer of JUN and FOS proteins (ΔFosB a truncated variant)

A. J. Robison, E. J. Nestler, "Transcriptional and epigenetic mechanisms of addiction" Nature Reviews, Neuroscience, 2011, 12, 623-635

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Why Chronic Use?

In general, speed of reward delivery contributes to effectiveness of reward. Rats learn first and run fastest in the portions of a maze that are closest to the reward.

In choosing between an immediate and a delayed reward; the immediate reward is preferred to the delayed reward even when the delayed reward is better.

Photo Credit: George Thomas (Creative Commons)

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Audience Survey Question

ANSWER THE QUESTION ON BLUE SCREEN IN ONE MOMENT

If speed of delivery matters, which mode of delivery will be slowest:

- Chewing
- Intranasal
- Smoking
- Intravenous injection

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Why Chronic Use? (Cont'd)

Reward *rate* plays a role in Addiction:
Heroin — which is highly addictive — enters the brain more rapidly but activates the same receptor as the less addictive morphine.

Administration Method Matters:
Nicotine reaches the brain faster (and is more addictive) when tobacco is smoked than when the leaf is chewed or when nicotine itself is given by the transdermal nicotine patch.

Smoked or intravenous cocaine reaches the brain faster than intranasal or oral routes, this contributes to their greater addictive potency.

R. A. Wise, E. A. Kiyatkin, Nature Rev. Neurosci., 2011, 12, 479-484.

"Smoking Crack" by Oaktown Crack Comics. - http://oaktowncrack.com/Smoking_Crack/index.html

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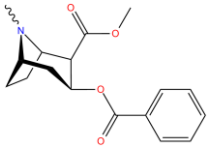
Cocaine Timeline

"Colcoca02". Licensed under CC BY-SA 3.0 via Commons - https://commons.wikimedia.org/wiki/File:Colcoca02.jpg#/media/File:Colcoca02.jpg



"Cocaine structure" by Nuklear at en.wikipedia. Licensed under CC BY-SA 3.0 via Commons - https://commons.wikimedia.org/wiki/File:Cocaine_structure.png#/media/File:Cocaine_structure.png

5000 BCE	Evidence of Coca chewing in South America
15th Century	Coca plantations are operated by Incas in Peru
1859	German graduate student Albert Niemann isolates cocaine from coca leaves
1862	Merck produces 1/4 pound of Cocaine
1883	Merck produces 3/4 pound of Cocaine
1884	Freud publishes On Coca in which he recommends the use of cocaine to treat a variety of conditions including morphine addiction
1884	Merck produces 3,179 pounds of Cocaine
1886	Merck produces 158,352 pounds of Cocaine
1886	Coca-Cola is first introduced by John Pemberton, containing cocaine laced syrup and caffeine.
1914	The United States Congress passes Harrison Narcotics Act, outlawing the sale of narcotics and stimulants, such as cocaine, without a prescription

Timeline based on www.erowid.org (search x timeline)

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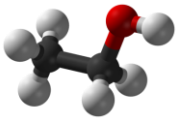
Alcohol Timeline

6000 BCE	Neolithic tribes make wine from fermented berries
2200 BCE	Cuneiform tablet recommends beer as a tonic for lactating women
1800 BCE	Beer is produced in quantity in northern Syria
625	Mohammed orders followers to abstain from alcohol
800	Arabs discover distillation of alcohol process
1100	Alcohol distillation is documented by the medical school at Salerno, Italy
Middle Ages	Distillation of grain alcohol in Europe follows the earlier distillation of wine
1525-1550	Excessive use of distilled spirits first becomes apparent in England
1600 – 1625	During the reign of James I, numerous writers describe widespread drunkenness from beer and wine among all classes. Alcohol use is tied to every endeavor and phase of life, a condition that continues well into the eighteenth century
1643	Britain imposes an excise tax on distilled spirits. Along with a tax of alcohol came the development of the moonshine trade.
1920-1933	Prohibition (of alcohol) begins in the United States

Timeline based on www.erowid.org (search x timeline)



Ball-and-stick model of ethanol

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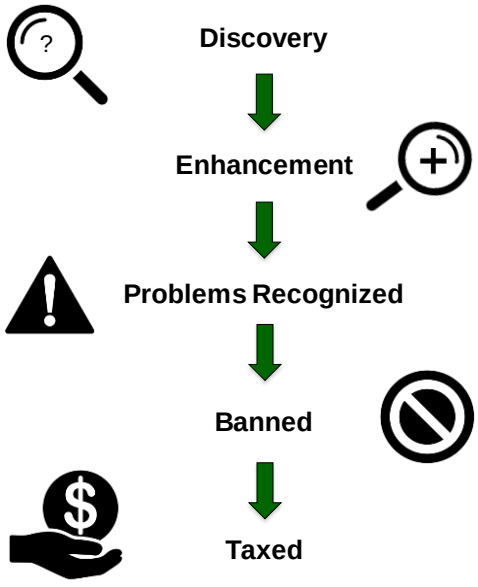
Timeline



Concentrate
- selective breeding
- isolation
- distillation

Increase potency
- alter structure

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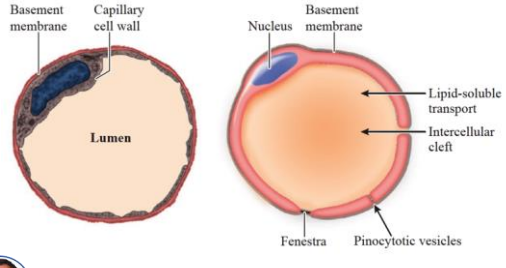
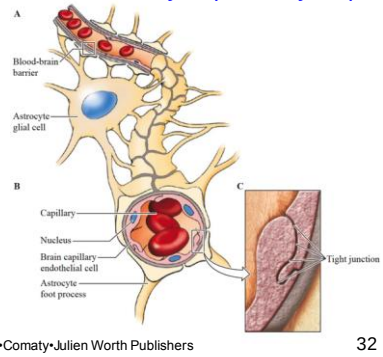
Pharmacokinetics

Capillaries
- Tiny, cylindrical blood vessels
- Have small pores (between 90 and 150 angstroms), which are larger than most drugs
- Allow transport of drugs regardless of lipid-solubility
- Blood & protein are too big for pores; drugs that bind to plasma proteins cannot pass through

Blood-Brain Barrier
- The brain must protect neurons from toxins
- But the brain has a great need for nutrients and oxygen (it has a high blood flow), which increases the risk of toxic danger
- Solution = the blood-brain barrier (BBB)
- Capillaries in brain do not allow drugs to pass as easily as capillaries in rest of body

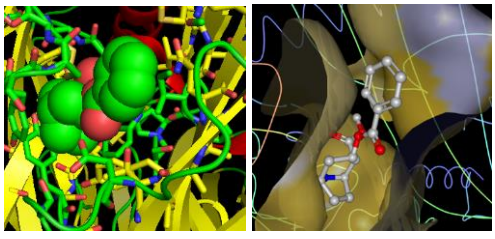
Hydrophobicity helps





Pharmacodynamics (Receptors for Drug Action)

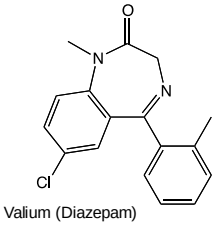
- **Receptor** = large biomolecule; site(s) where naturally occurring compounds (*transmitters* or *modulators)* produce biological effect
- Hundreds of receptor types known (http://gpcr.scripps.edu/index.html)
- Neurotransmitters can be specific to certain receptors, but a drug may be more specific than the endogenous neurotransmitter
- Drugs form reversible interactions with specific receptors:
  - salt bridges
  - π-stacking
  - hydrogen bonds
  - π-cation interactions
- Receptor protein changes conformation (shape) & response occurs



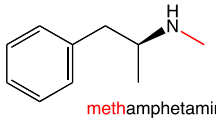
Binding Results in 1 of 3 Actions:

1. Binding to site of normal endogenous neurotransmitter initiates similar cellular response (*agonistic action*).
2. Binding to nearby site to facilitate transmitter binding (*allosteric action*).
3. Binding to receptor site, blocks access of transmitter to binding site (*antagonistic action*).

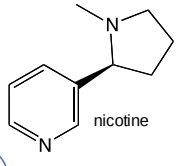
Addictive Processes



Valium (Diazepam)



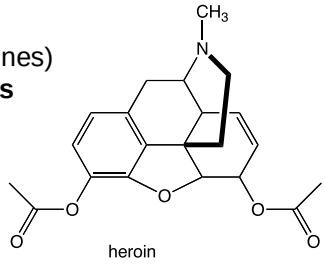
methamphetamine



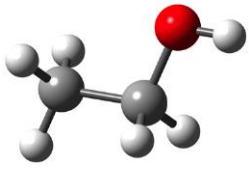
nicotine

Substances of Abuse

A. **Sedatives** (benzodiazepines)
B. **Opiate-based painkillers**
C. **Stimulants**
   - Caffeine
   - Nicotine
   - Cocaine
   - Amphetamines
D. **Alcohol**



heroin

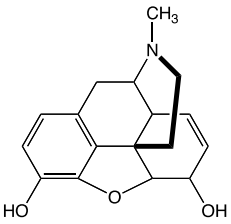


ethanol

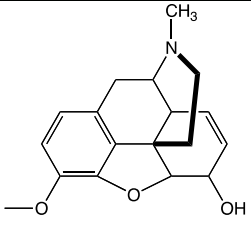
Non-chemical Addictions

- Gambling
- Tanning beds
- Food (Sugar/Fats)

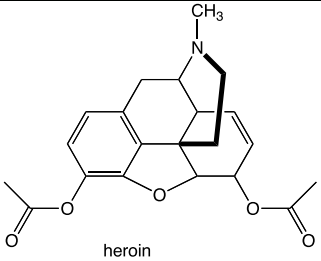
Opioids



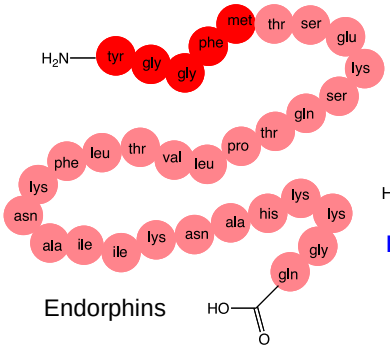
morphine



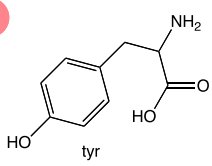
codeine



heroin

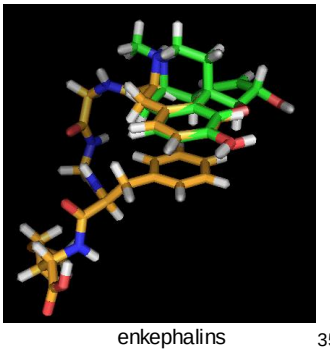


Endorphins



tyr

Natural agonists



enkephalins

Audience Survey Question

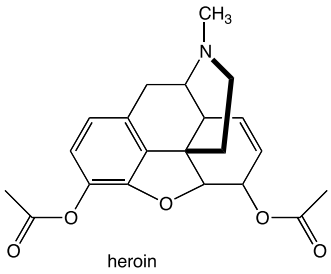
ANSWER THE QUESTION ON BLUE SCREEN IN ONE MOMENT

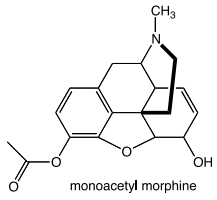
Which of the following is NOT an opioid-based drug that is commonly abused:

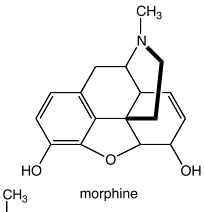
- Vicodin
- OxyContin
- Oxytocin
- Percocet

Heroin (Diacetylmorphine)

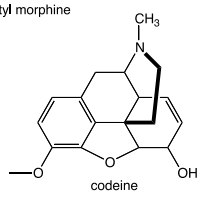
- Rapidly crosses blood-brain barrier; smoked or injected.
- Metabolized to monoacetylmorphine, morphine, and codeine.



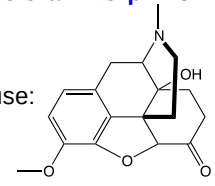




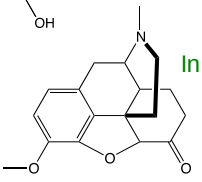
more hydrophobic than morphine



Other opiods of abuse:



In OxyContin



In Vicodin

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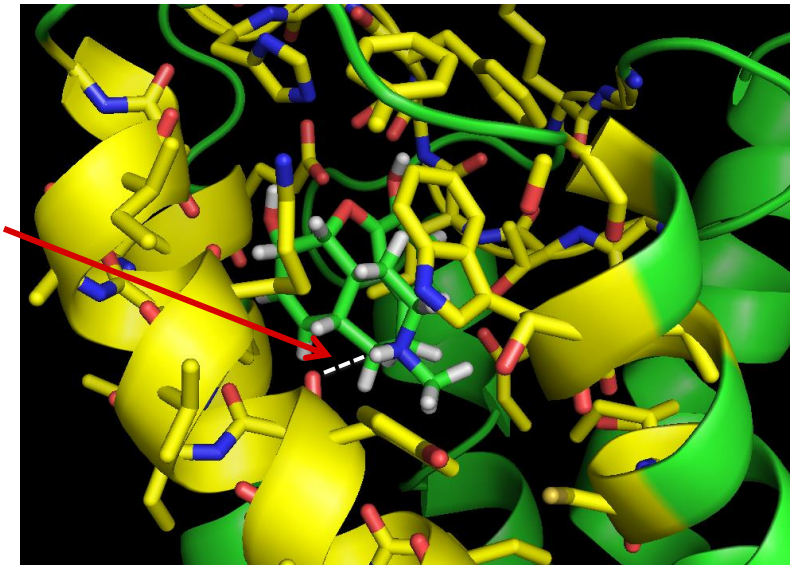
Opioid Receptor-Morphine Salt Bridge

Salt bridge



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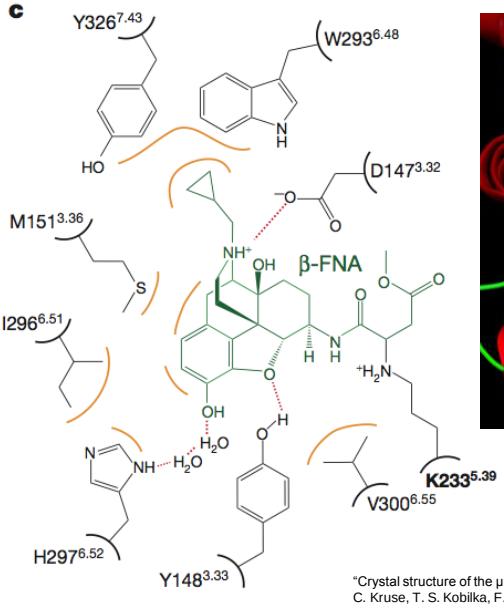
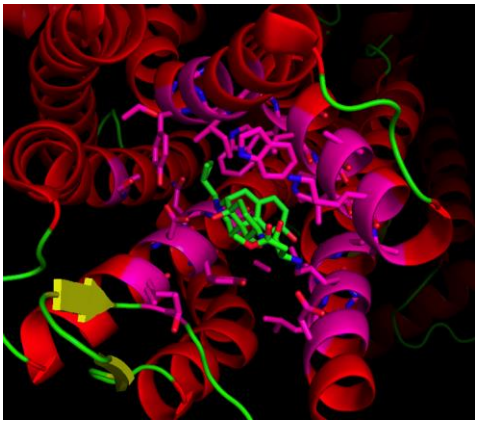
μ-opioid receptor





"Crystal structure of the μ-opioid receptor bound to a morphinan antagonist", A. Manglik, A. C. Kruse, T. S. Kobilka, F. S. Thian, J. M. Mathiesen, R. K. Sunahara, L. Pardo, W. I. Weis, B. K. Kobilka & S. Granier, Nature, 485 321 (2012)

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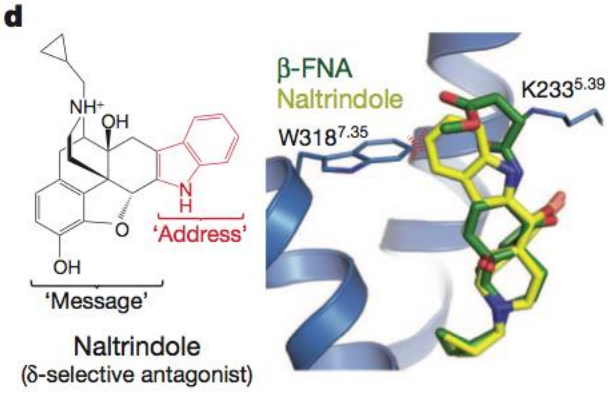
μ-opioid receptor



"Crystal structure of the μ-opioid receptor bound to a morphinan antagonist", A. Manglik, A. C. Kruse, T. S. Kobilka, F. S. Thian, J. M. Mathiesen, R. K. Sunahara, L. Pardo, W. I. Weis, B. K. Kobilka & S. Granier, Nature, 485 321 (2012)

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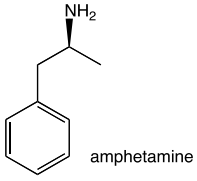
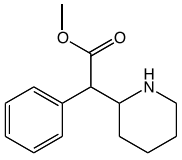
Psychostimulants, Examples

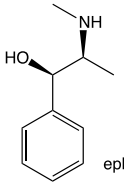


amphetamine

- **Amphetamines**: *d* or *l*
- **Methylphenidate** (Ritalin)

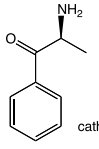


ritalin (methylphenidate)

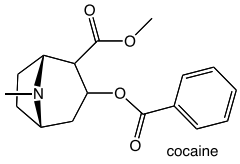


ephedrine

- **Ephedrine, ma-huang** (Ephedra vulgaris)
- **Cocaine** (Erythroxylum coca)
- **Cathinone** (Catha edulis) khat, tscaht, miraa



cathinone



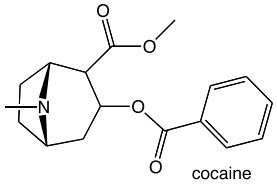
cocaine

Cocaine

A stimulant of the central nervous system, an appetite suppressant, and a topical anesthetic.

It is a serotonin–norepinephrine–dopamine reuptake inhibitor (also known as a triple reuptake inhibitor (TRI)), which mediates functionality of these neurotransmitters.

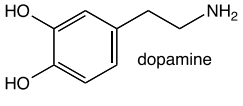
It acts simultaneously as a reuptake inhibitor for
serotonin (5-HT)
norepinephrine (noradrenaline, NA)
and dopamine (DA)
by blocking the action of
serotonin transporter (SERT),
norepinephrine transporter (NET)
and dopamine transporter (DAT)



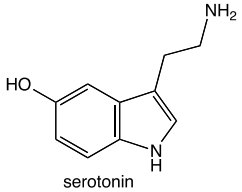
cocaine

Leading to increased extracellular concentrations of these neurotransmitters and, therefore, an increase in serotonergic, noradrenergic or adrenergic, and dopaminergic neurotransmission.

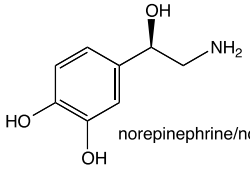
DAT action is what is addictive



dopamine

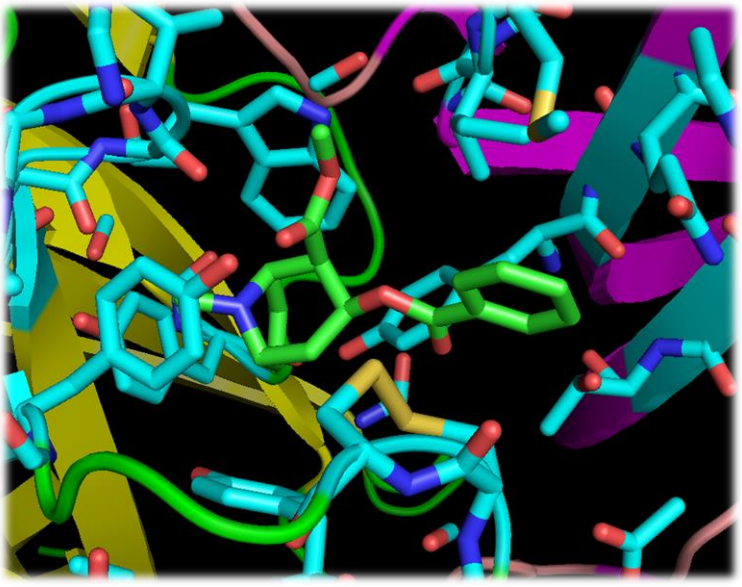


serotonin



norepinephrine/noradrenaline

Cocaine in Acetylcholine Binding Protein



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Methamphetamine Mechanisms of Action

Methamphetamine increases synaptic levels of the neurotransmitters

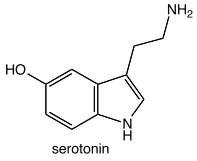
dopamine
serotonin (5-HT)
norepinephrine/noradrenaline,
has α and β adrenergic agonist effects.

Norepinephrine is responsible for methamphetamine's alerting, anorectic, locomotor and sympathomimetic effects.

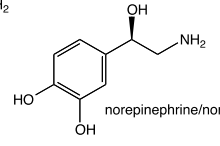
Dopamine stimulates locomotor effects, psychosis, and perception disturbances.

Serotonin (5HT) is responsible for delusions and psychosis.

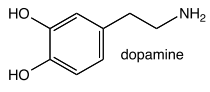
Methamphetamine's effects are similar to cocaine but its onset is slower and the duration is longer. Racemic amphetamine and d-amphetamine have similar chemical properties and actions to methamphetamine but are less potent.



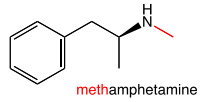
serotonin



norepinephrine/noradrenaline



dopamine



methamphetamine

Julien's Primer of Drug Action | Thirteen Edition | Advokat•Comaty•Julien Worth Publishers

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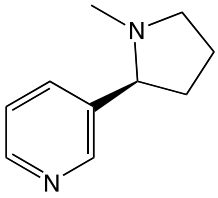
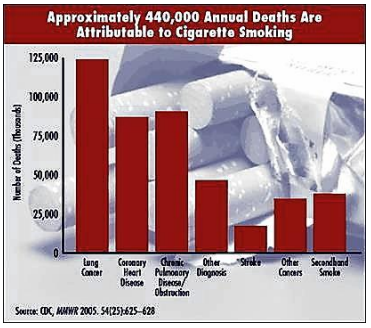
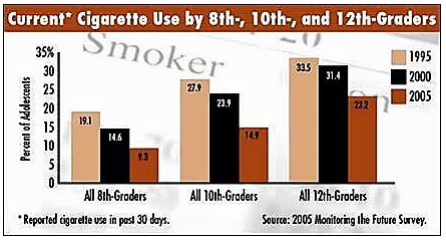
Nicotine



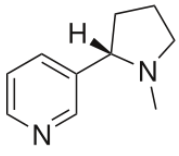
- **One** of the 3 most widely used psychoactive drugs.
- **Most preventable cause** of disease and premature death.
- Current use fell from ~50 percent in 1965 to **~25 percent in 1998**.
- Average starting age for people is declining; 9 out of 10 are addicted by age 21.



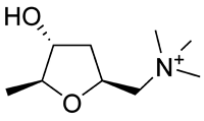


Nicotine

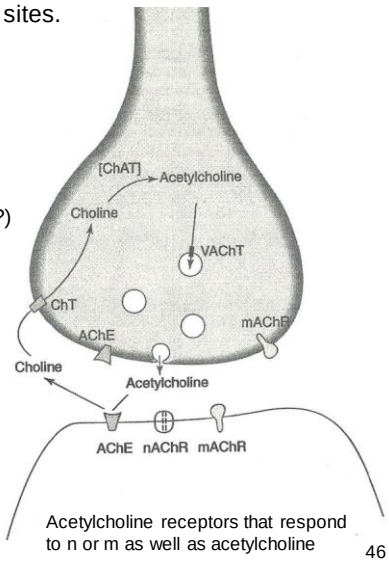
- Mechanism of action: Indirect activation of the sympathetic system.
  - Occupies and activates nicotinic cholinergic sites. (Ionotropic)
  - Low doses stimulate the receptors.
  - High doses block the receptors.
  - Causes release of:
    - Dopamine (reinforcement?)
    - Acetylcholine and glutamate (memory?)



nicotine

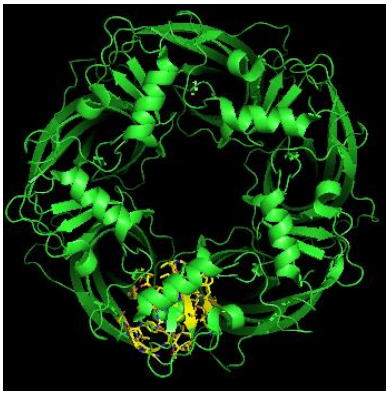


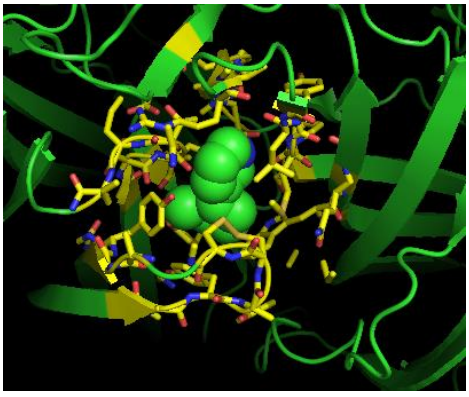
Muscarine (from mushrooms)



Acetylcholine receptors that respond to n or m as well as acetylcholine

Acetylcholine binding protein+nicotine





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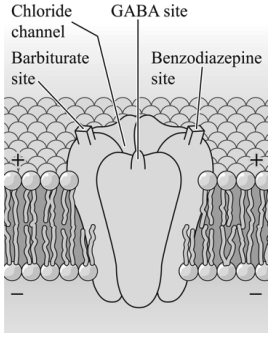
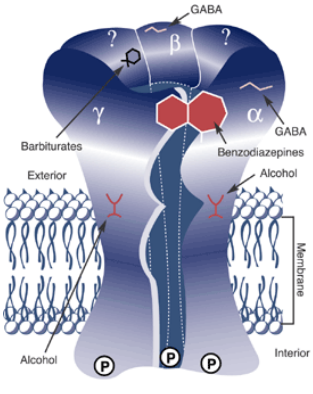
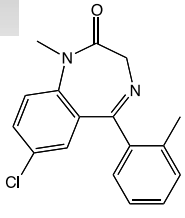
Sedatives / Barbiturates / Benzodiazepines

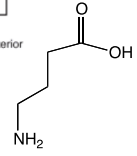
Alcohol, Barbiturates & Benzodiazepines are GABA receptor allosteric agonists
Bind to nearby sites and facilitate GABA, "flooding" neurons with Cl^-, inhibiting neural actions







Valium (Diazepam)



Gamma aminobutyric acid

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Audience Survey Question

ANSWER THE QUESTION ON BLUE SCREEN IN ONE MOMENT

Alcohol (ethanol) disrupts/interferes with the action of which of the following receptors:

- GABA and Glutamate
- Serotonin
- Opioid
- Cannabinoid
- All of the above

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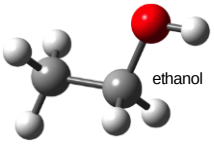
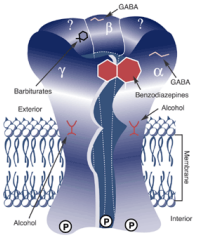
Alcohol, Pharmacodynamics

Alcohol disturbs synaptic activity of neurotransmitters (especially glutamate and GABA) and various intracellular transduction processes.

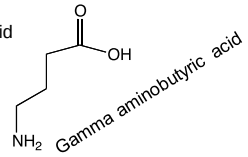


- **Glutamate Receptors**
  - Ethanol inhibits responsiveness of NMDA receptors to glutamate.
  - Exacerbated by enhancement of inhibitory GABA transmission.
  - *Acamprosate* (structural analog of glutamate) used to maintain abstinence in alcohol-dependent patients.



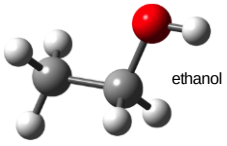
- **GABA Receptors**
  - Ethanol activates GABA-mediated increase in chloride ion flow → neural inhibition.
  - Results in sedation, muscle relaxation, inhibition of cognitive and motor skills, antianxiety effects.
  - Ultimately leads to augmentation of dopaminergic projections from VTA to nucleus accumbens (reward circuits).

- **Opioid Receptors**
  - Alcohol-dependent people may have (genetic) dysfunction in brain's opioid system.
  - Ethanol may trigger opioid release, triggering DA response in reward circuitry.
  - *Naltrexone* (opioid antagonist) may reduce alcohol craving.



Alcohol, Pharmacodynamics

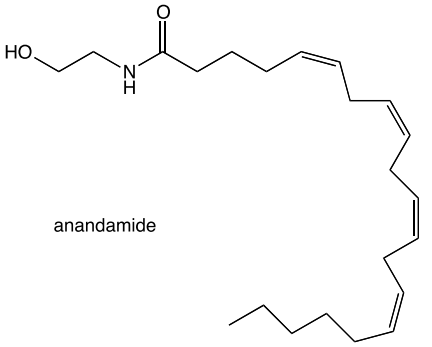
Serotonin Receptors



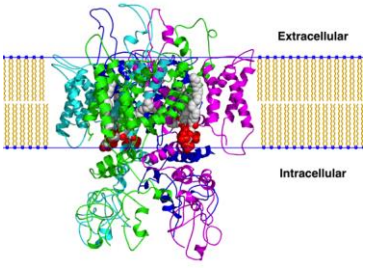
- Chronic alcohol use augments serotoninergic activity.
  - Serotonin dysfunction may play a role in some types of alcoholism.
- Emphasis in 5-HT_2 and 5-HT_3 receptors (located on dopaminergic neurons in nucleus accumbens).
- Serotonin reuptake-inhibiting antidepressants (e.g., setraline [Zoloft]) more effective in reducing drinking in lower-risk alcohol males.

Cannabinoid Receptors

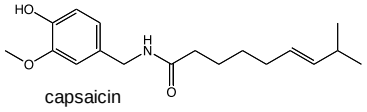
- Chronic alcohol use stimulates formation of endogenous cannabinoid transmitter *anandamide*.
  - Leads to down regulation of cannabinoid receptors, disinhibiting nucleus accumbens.
  - Cessation of drinking → hyperactive endocannabinoid reaction → alcohol craving



Cannabinoids & Endocannabinoids

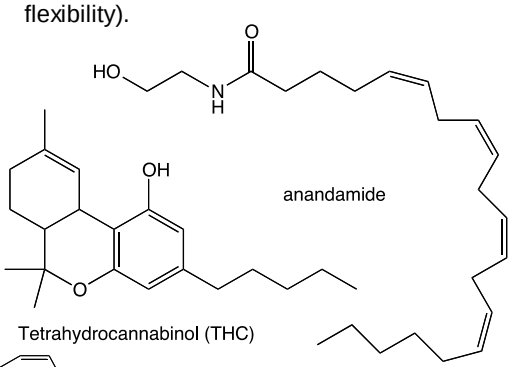


transient receptor potential cation channel subfamily V member 1 (**TrpV1)**

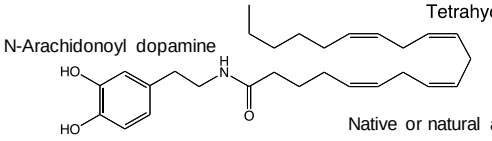


capsaicin

- The term ***anandamide*** is derived from the Sanskrit word for bliss (ananda).
- Anandamide is an ***endogenous cannabinoid agonist;*** interestingly, it is only a weak agonist at its receptors (likely due to conformational flexibility).



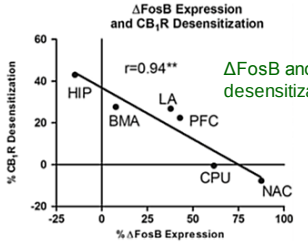
Tetrahydrocannabinol (THC)



Native or natural agonist (what is in us that let's us feel heat)

Cannabinoids and Opioids Share Several Pharmacological Properties

- **Analgesia,** Sedation, Catalepsy, Hypotension, Hypothermia
- Indeed, cannabinoids and opioids can therapeutically be used together (e.g., for analgesia for a "**morphine-sparing effect**").
- Stimulation of the CB1 receptor activates mesolimbic dopamine reward pathway; sharing a common action with such drugs as cocaine, morphine, and alcohol.
- Exposure to one abusing drug (THC) can precipitate relapse to another (cocaine).
- A cannabinoid antagonist blocks or prevents relapse to other drugs (alcohol, cocaine, or heroin).
- Thus, CB-1 receptors are thought to be involved in opioid-induced reward.



ΔFosB and CB1 receptor desensitization inversely correlated

Lazenka, M F; Selley, D E; Sim-Selley L J Neuropharmacology 2014, 77 224-233.

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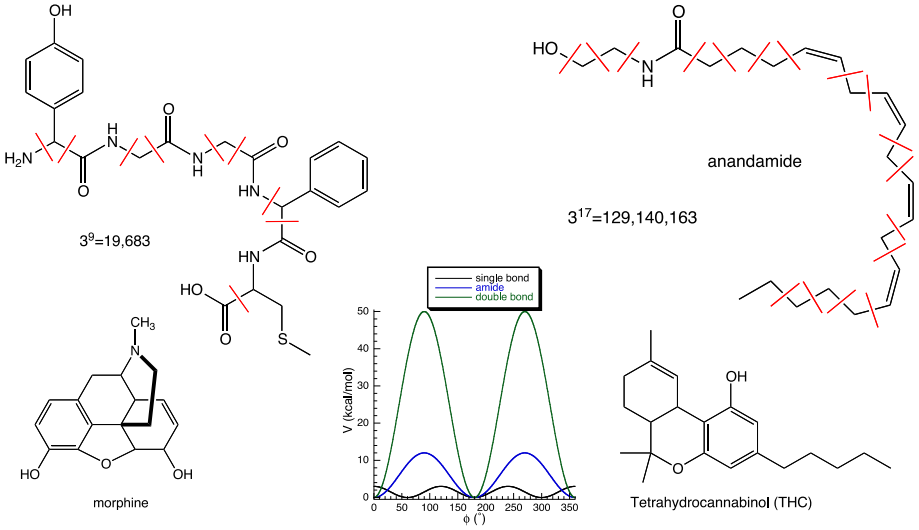
Similarities between Cannabinoid and Opioid Systems

Natural agonists are weaker acting than exogenous agonists.
Natural agonists are conformationally more flexible than exogenous agonists.



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Chemistry of Addiction

- **Neuronal synaptic transmission involves**
  neurotransmitter presynaptic release, receptor binding,
  neurotransmitter binding site release,
  and neurotransmitter degradation/reuptake

- **Drug molecules "look like" natural substrates**
  bind to receptor, transporter, or enzyme active sites

- **Impact of drug depends upon:**
  -Strength of binding, which depends upon:
  -shape & positioning of functional groups
  -hydrogen bonding, salt bridges
  -π-stacking, π-cation interactions
  -hydrophobic contacts
  -conformational rigidity
  -Ability to pass through hydrophobic blood-brain barrier

- **Addiction correlates with neuroplasticity and ΔFosB accumulation which involves chronic use, at least partially due to rapid reward.**

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Chemistry of Addiction

Acknowledgements:

Colorado State University

College of Natural Sciences

The materials for this presentation accrued from the development effort for a Chemistry of Addictions course envisioned for Chemistry majors as well as CSU's Psychology Department's Concentration in Addiction Counseling students. The course is in its 4th year.

Initial development was carried out by Michael Gardner, a Hendrix College undergraduate at the time and now 4th year Medical Student at the University of Arkansas for Medical Sciences.

Support for the development effort was provided by the Department of Chemistry and the College of Natural Sciences.

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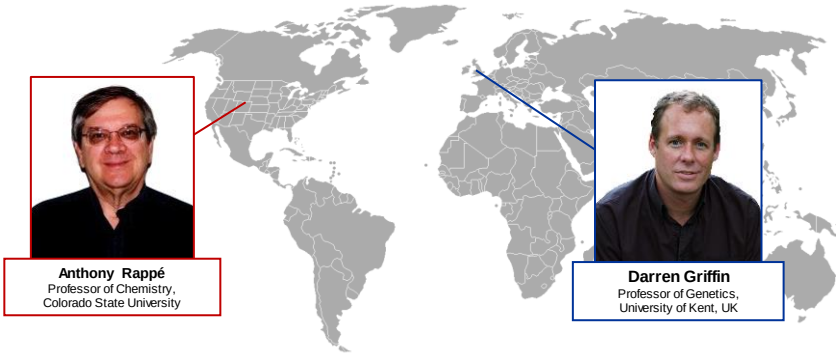

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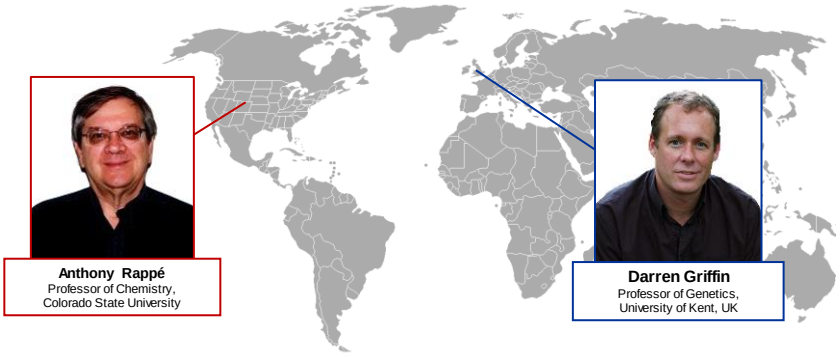

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