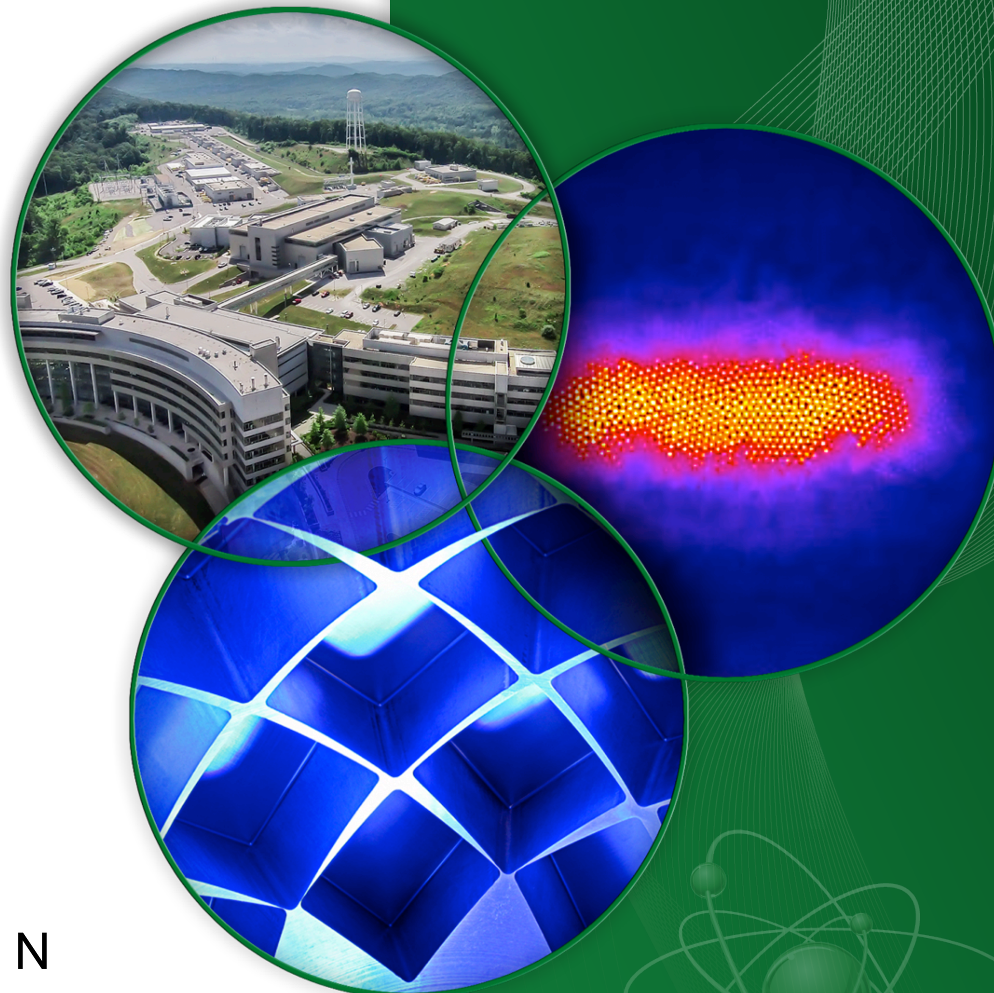
# ICE-MAN and VirtuES, the Integrated Computational Environment-Modeling & Analysis for Neutrons at ORNL



**Timmy Ramirez-Cuesta,**
Luke Daemen, YQ Cheng, E Mamontov, N Jalarvo, Eric Novak, Naresh Osti, NScD, ORNL
Robert Smith, Jay Jay Billings
CSD, ORNL

ORNL is managed by UT-Battelle
for the US Department of Energy

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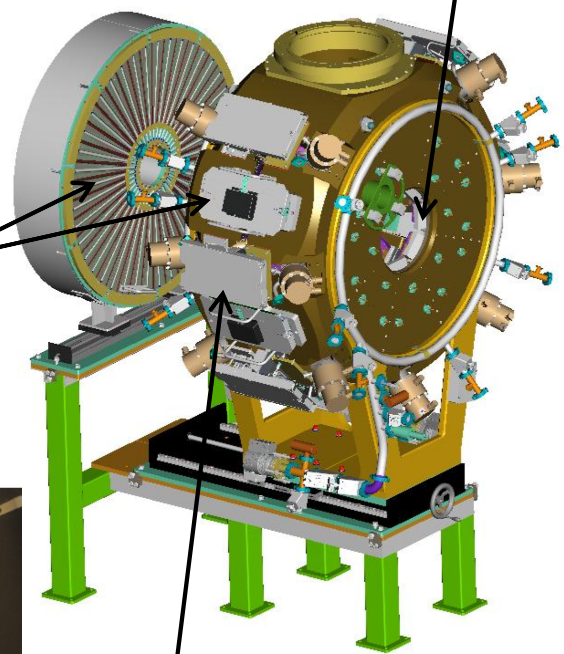
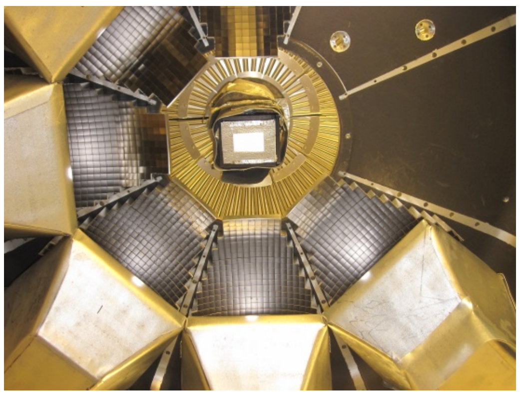
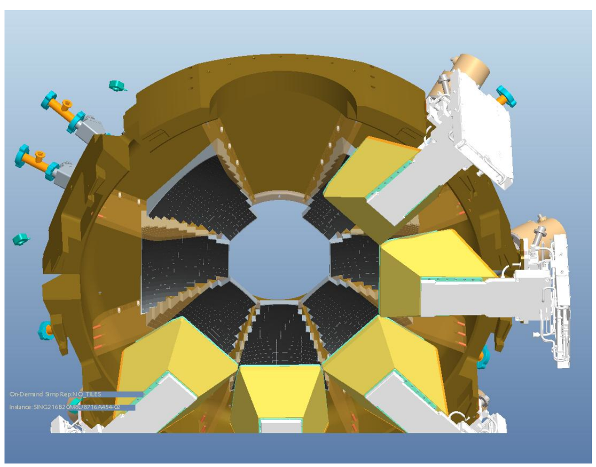
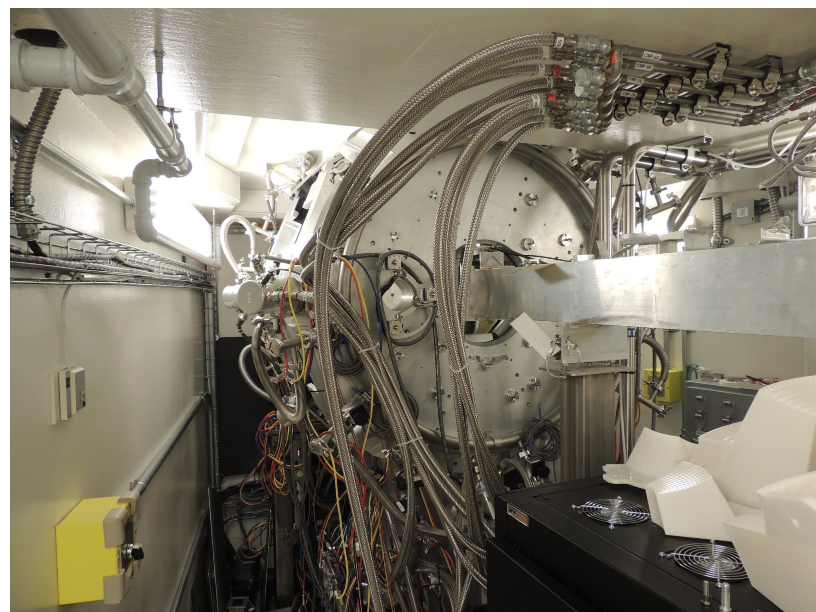
# VISION



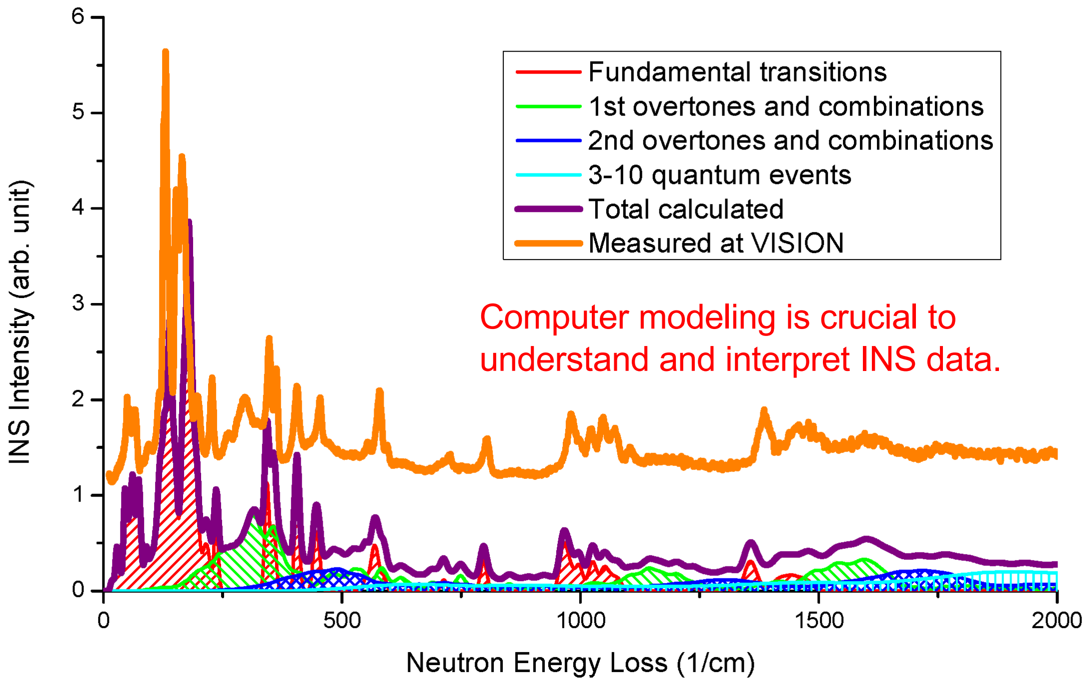

- Indirect geometry spectrometer optimized to study chemical systems
- High flux/throughput
- Broadband (-2 to 1000 meV)
- Constant dE/E (~1.5%)
- Elastic line HMFW ~150 μeV
- Simultaneous diffraction







# Integrated modeling for data interpretation

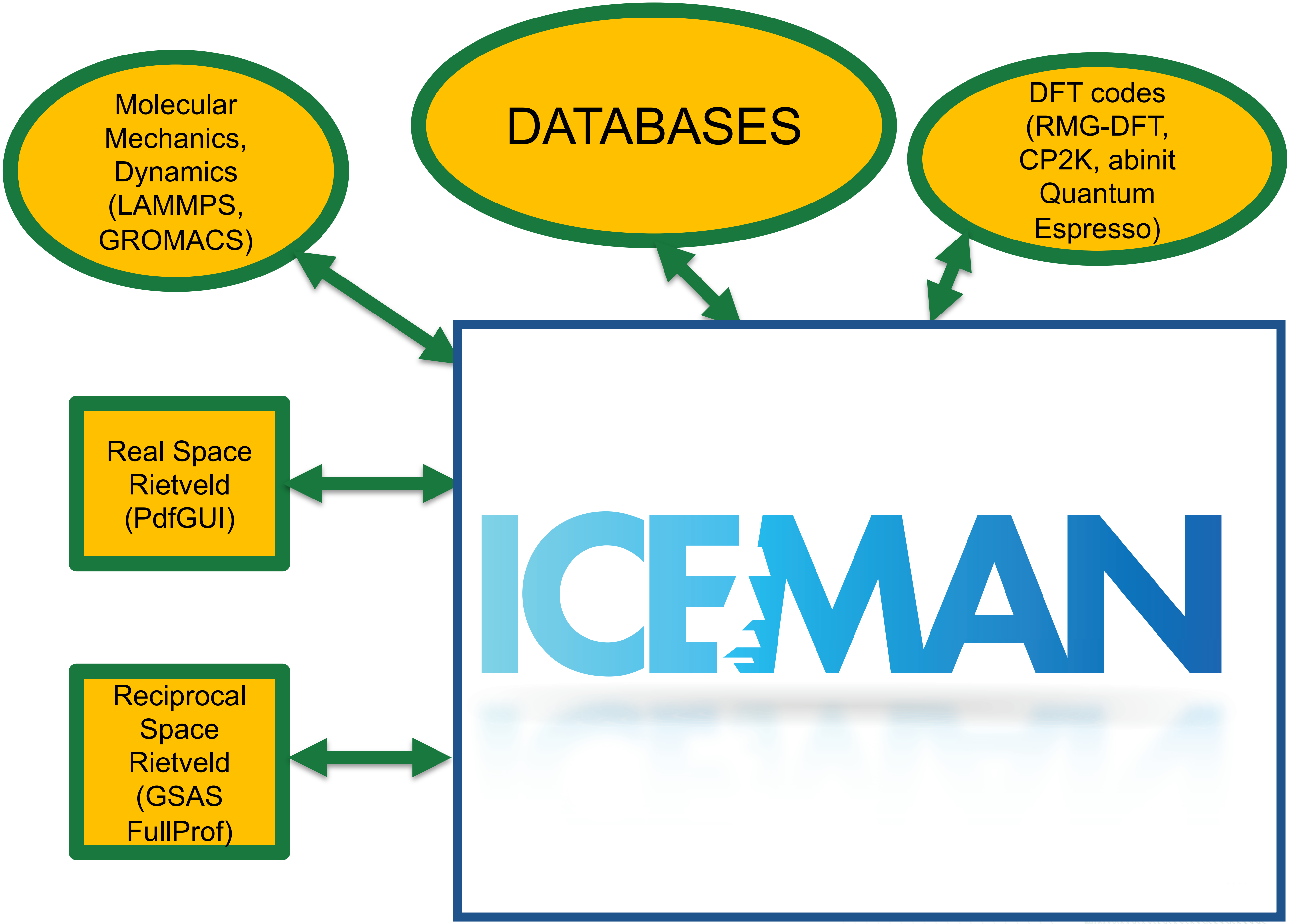


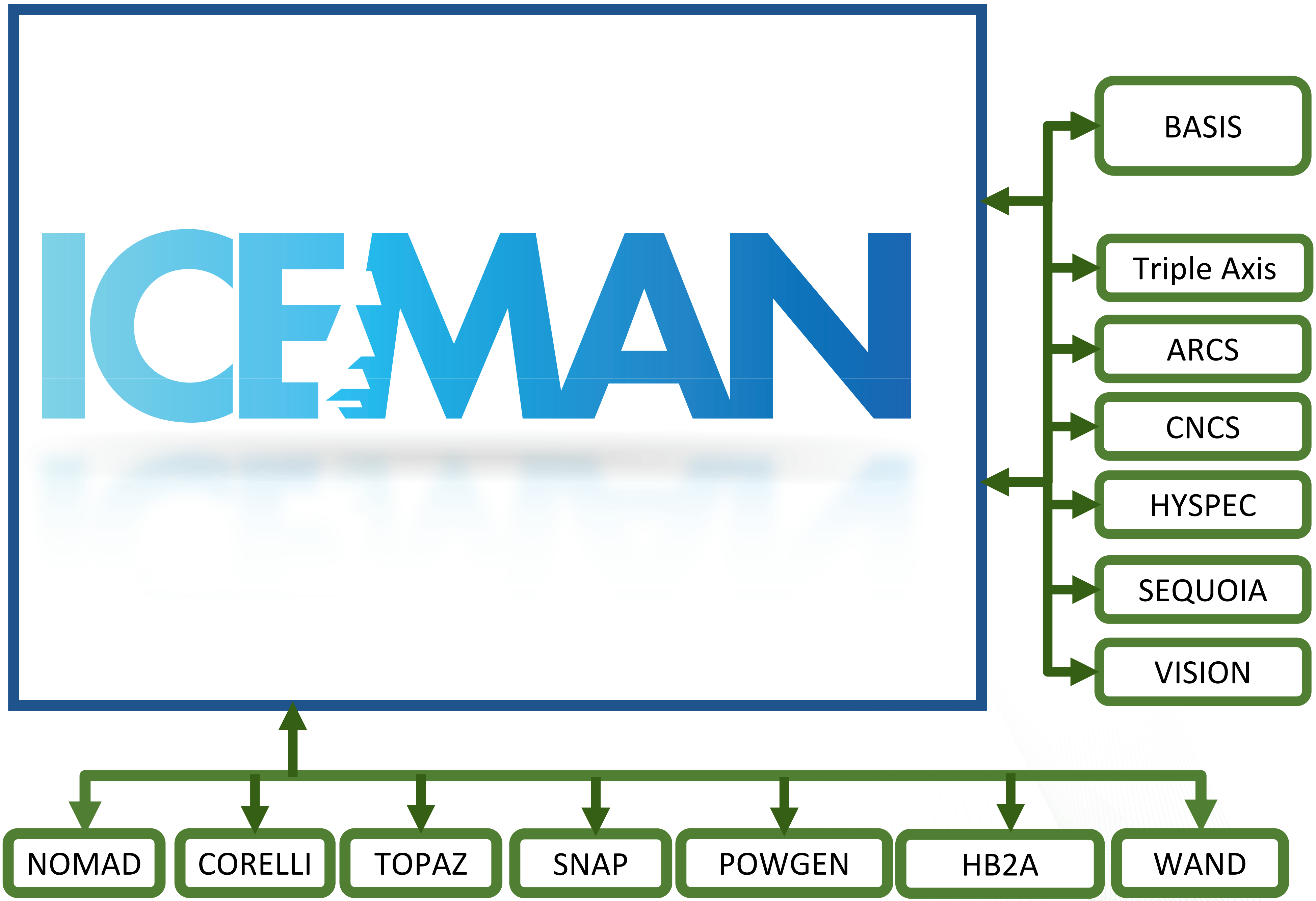

- Dual 16 core Intel Haswell E5-2698v3 3.2 GHz Processors per node
- 50 compute nodes, 1600 (non-hyperthreaded) cores
- 128 GB memory/node, 6.4 TB Total memory
- Each node has 10Gbe and Infiniband networking for connectivity.
- Installed as part of the ORNL Compute and Data Environment for Science (CADES)

**VirtuES cluster**

# Integrated Computational Environment, for Modeling and Analysis of Neutron data

Acknowledgement: Laboratory Directed Research and Development program at ORNL


Molecular Mechanics, Dynamics (LAMMPS, GROMACS)
DATABASES
DFT codes (RMG-DFT, CP2K, abinit Quantum Espresso)
Real Space Rietveld (PdfGUI)
Reciprocal Space Rietveld (GSAS FullProf)
ICEMAN


ICEMAN
BASIS
Triple Axis
ARCS
CNCS
HYSPEC
SEQUOIA
VISION
NOMAD
CORELLI
TOPAZ
SNAP
POWGEN
HB2A
WAND

# Why do we need simulations for NVS (or INS in general)?

- Interpret neutron data
  - assigning peaks to vibrational modes
- Obtain insight on fundamental properties
  - understanding interatomic interactions, anharmonicity, complex excitations, phase transitions, chemical reactions
- Connect theory and experiment
  - simulation is a virtual experiment and an *in silico* implementation of theory

We can measure it. → We do understand it.

# What to simulate for INS?

- Double differential cross-section

$$\frac{d^2\sigma}{d\Omega\, dE'} = \text{(number of neutrons scattered per second into a small solid angle } d\Omega \text{ in the direction } \theta, \phi \text{ with final energy between } E' \text{ and } E' + dE')/\Phi\, d\Omega\, dE',$$

- Fermi's golden rule

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right)_{\lambda\to\lambda'} = \frac{k'}{k}\left(\frac{m}{2\pi\hbar^2}\right)\boxed{|\langle \boldsymbol{k}'\lambda'|V|\boldsymbol{k}\lambda\rangle|^2}\delta(E_\lambda - E_{\lambda'} + \hbar\omega) \propto \frac{k'}{k}S(Q,\omega)$$

*V*: potential describing the interaction between neutrons and the system
$\hbar\omega$ : fundamental excitation in the system

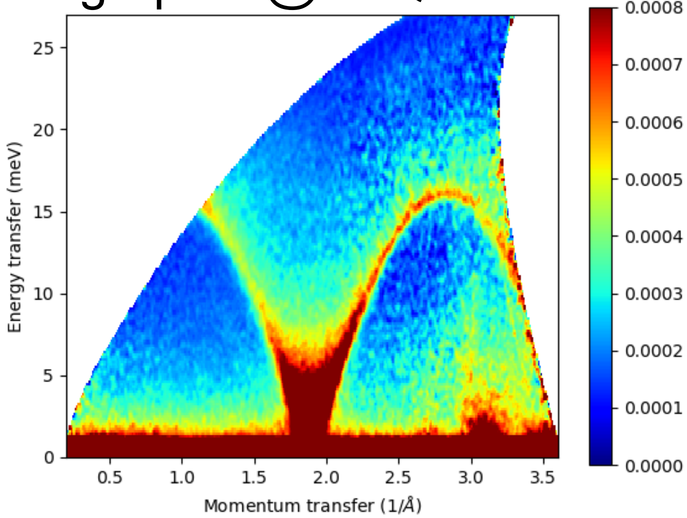
- The goal is to formulate the interaction between neutrons and the system, so that *S*(*Q*,*ω*) can be expressed by the excitations of interest.

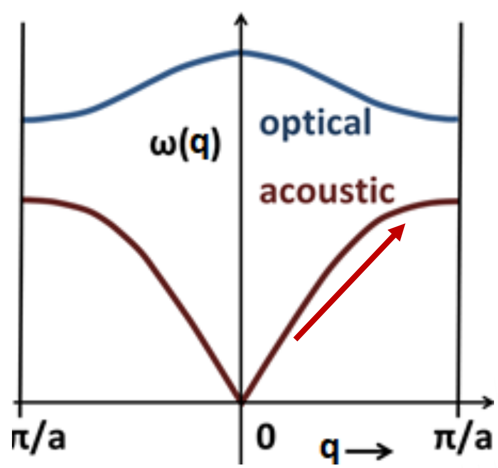
# Coherent inelastic scattering

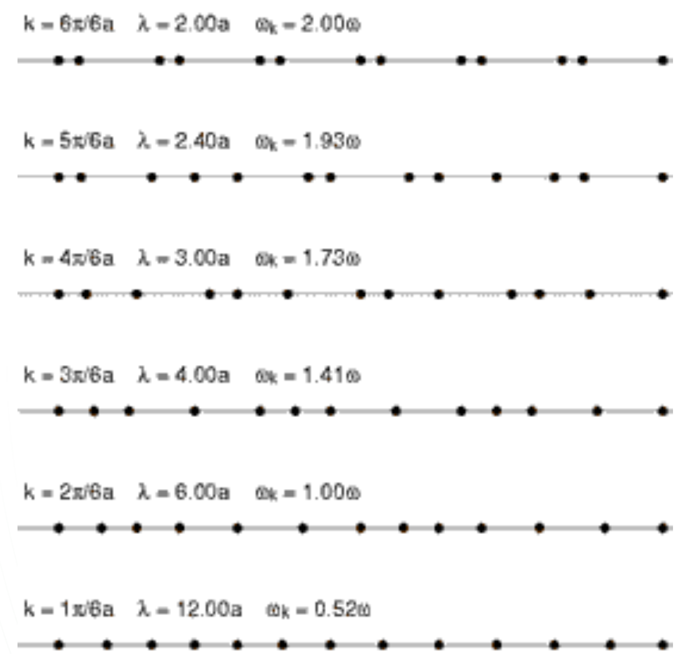
- One-phonon $S(Q,\omega)$

$$S_{coh\pm1}(\boldsymbol{Q},\omega) = \frac{1}{2N}\sum_{s}\sum_{\boldsymbol{\tau}}\frac{1}{\omega_s}\left|\sum_{d}\frac{\bar{b}_d}{\sqrt{m_d}}\exp(-W_d)\exp(i\boldsymbol{Q}\cdot\boldsymbol{r}_d)\,(\boldsymbol{Q}\cdot\boldsymbol{e}_{ds})\right|^2 \times\langle n_s+\frac{1}{2}\pm\frac{1}{2}\rangle\,\delta(\omega\mp\omega_s)\delta(\boldsymbol{Q}\mp\boldsymbol{q}-\boldsymbol{\tau})$$

graphite@SEQUOIA



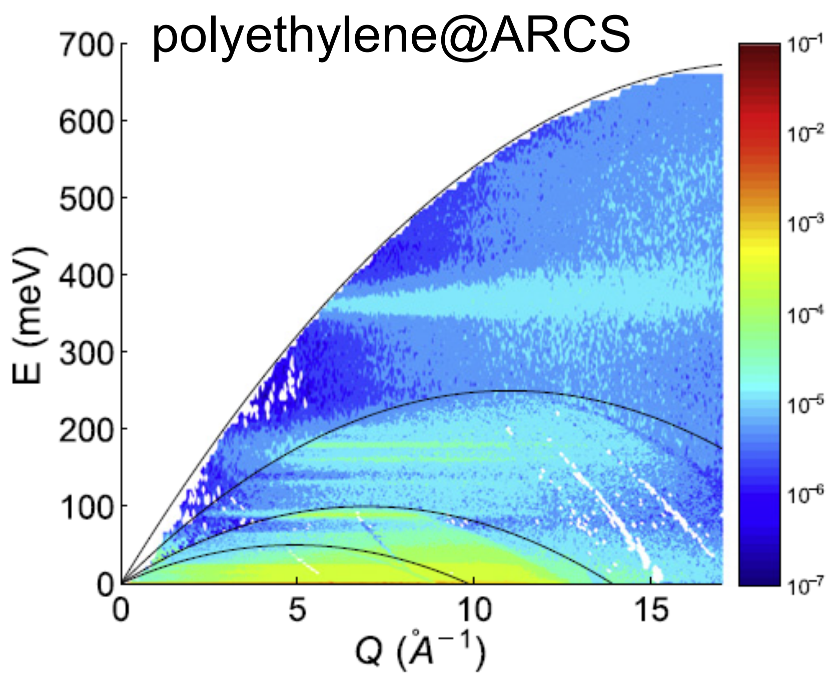

From: wikipedia

- **Peak position in energy depends on *Q*.**
- **Total intensity determined by not only how each atom moves, but also their relative phase.**

# Incoherent inelastic scattering

- One-phonon $S(Q,\omega)$

$$S_{inc\pm1}(\boldsymbol{Q},\omega) = \sum_d \frac{1}{2m_d}\left\{\bar{b}_d^2 - \left(\bar{b}_d\right)^2\right\}\exp(-2W_d)\sum_s \frac{|\boldsymbol{Q}\cdot\boldsymbol{e}_{ds}|^2}{\omega_s}\langle n_s + \frac{1}{2} \pm \frac{1}{2}\rangle\,\delta(\omega \mp \omega_s)$$




C.M. Lavelle et al. / Nuclear Instruments and Methods in Physics Research A 711 (2013) 166–179

Coherent

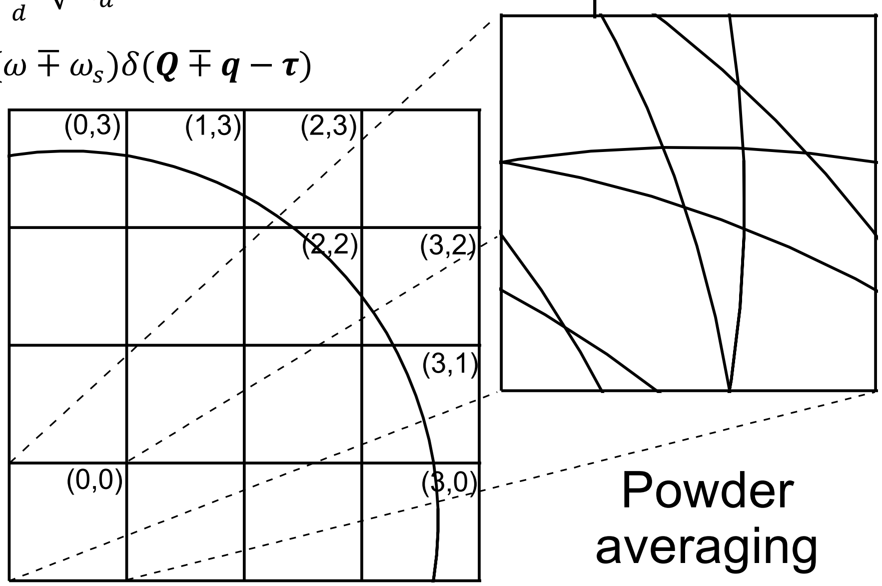
Incoherent

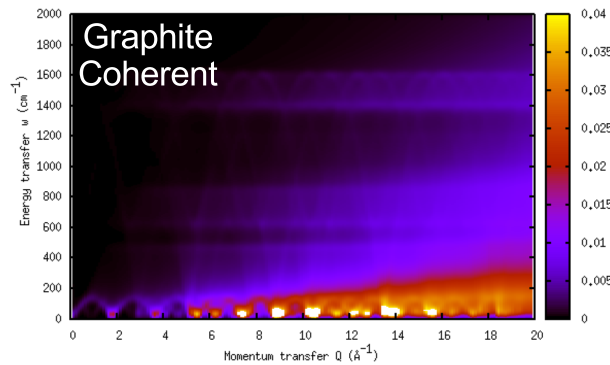
- **Peak position in energy does not depend on *Q***
- **Each atom contributes to the total intensity independently.**

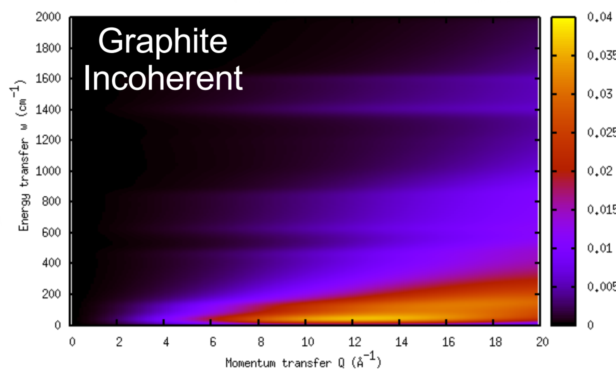
# Incoherent approximation

- When and why
  - Elements/isotopes with large incoherent scattering cross-section (e.g., hydrogen, vanadium) – The scattering itself is intrinsically incoherent.
  - High Q or large unit cell (small Brillouin zone), e.g. in low symmetry or disordered structure – The scattering may be coherent, but the ruler is too big for the pattern to be resolved.

$$S_{coh\pm1}(\boldsymbol{Q},\omega) = \frac{1}{2N}\sum_s\sum_{\boldsymbol{\tau}}\frac{1}{\omega_s}\left|\sum_d \frac{\bar{b}_d}{\sqrt{m_d}}\exp(-W_d)\exp(i\boldsymbol{Q}\cdot\boldsymbol{r}_d)\,(\boldsymbol{Q}\cdot\boldsymbol{e}_{ds})\right|^2 \times\langle n_s+\frac{1}{2}\pm\frac{1}{2}\rangle\,\delta(\omega\mp\omega_s)\delta(\boldsymbol{Q}\mp\boldsymbol{q}-\boldsymbol{\tau})$$




$$S(\boldsymbol{Q},n\omega_s) = \frac{(\boldsymbol{Q}\cdot\boldsymbol{U}_s)^{2n}}{n!}\exp[-(\boldsymbol{Q}\cdot\boldsymbol{U}_{total})^2]$$

$$\boldsymbol{U}_s = \sqrt{\frac{\hbar}{2m\omega_s}}\,\boldsymbol{e}_{ds}$$

# Development of OCLIMAX

- Started 2016
- First version released 2017
- Paper published 2019
- Used to analyze data from VISION and multiple other neutron spectrometers

Features:

- ❖ Incoherent and coherent scattering
- ❖ Powders and single crystals
- ❖ Temperature effects
- ❖ Multiphonon excitations
- ❖ Arbitrary instrument geometry and resolution
- ❖ Arbitrary cuts in 4-dimensional Q-E space
- ❖ Interface with atomistic modeling tools (e.g. DFT codes)
- ❖ Interface with INS data analysis tools (e.g. DAVE and Mantid)
- ❖ User-friendly (multiple platform, easy to use, fast on PCs)

JCTC Journal of Chemical Theory and Computation
Article
Cite This: J. Chem. Theory Comput. 2019, 15, 1974–1982
pubs.acs.org/JCTC

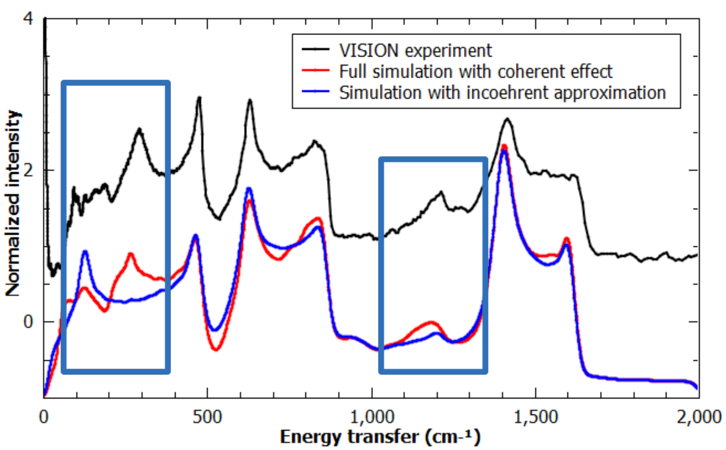
**Simulation of Inelastic Neutron Scattering Spectra Using OCLIMAX**

Y. Q. Cheng,* L. L. Daemen, A. I. Kolesnikov, and A. J. Ramirez-Cuesta*
Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge Tennessee 37831, United States

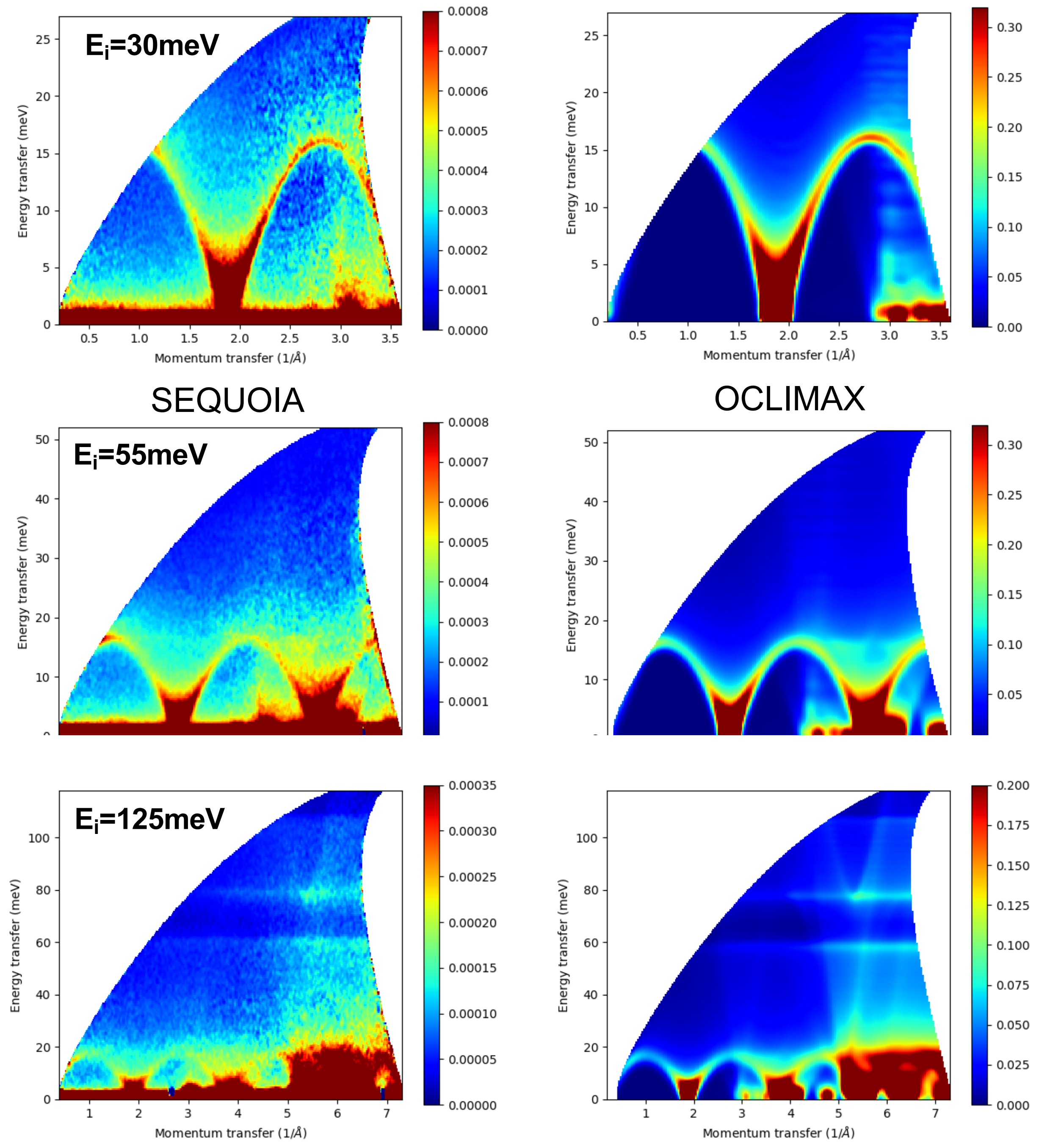
# OCLIMAX example: graphite

- Coherent scattering
  - Powders
  - Single crystal
- Kinematics
  - Option to generate masks in the map

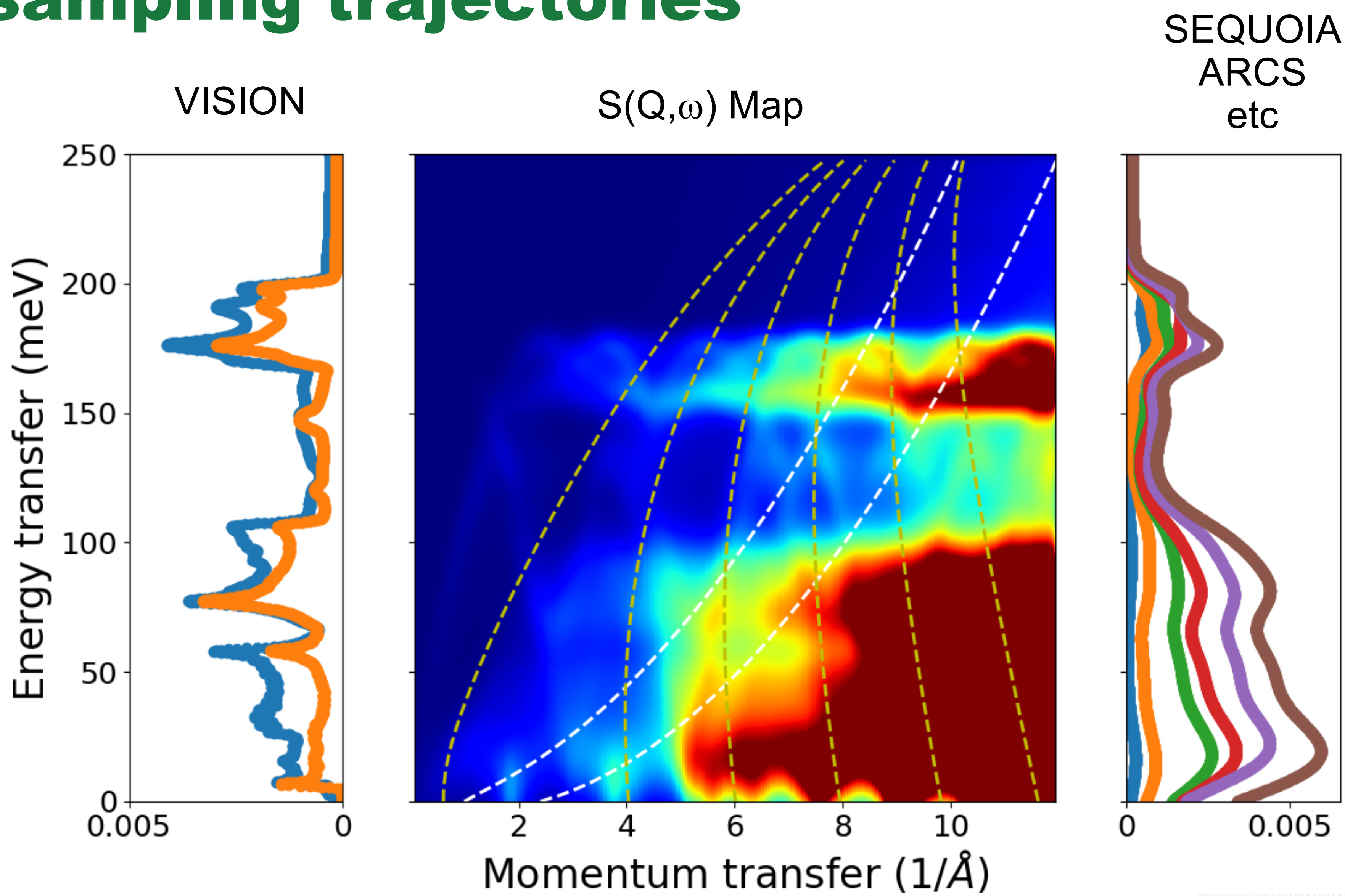
VISION




Full calculation versus incoherent approximation

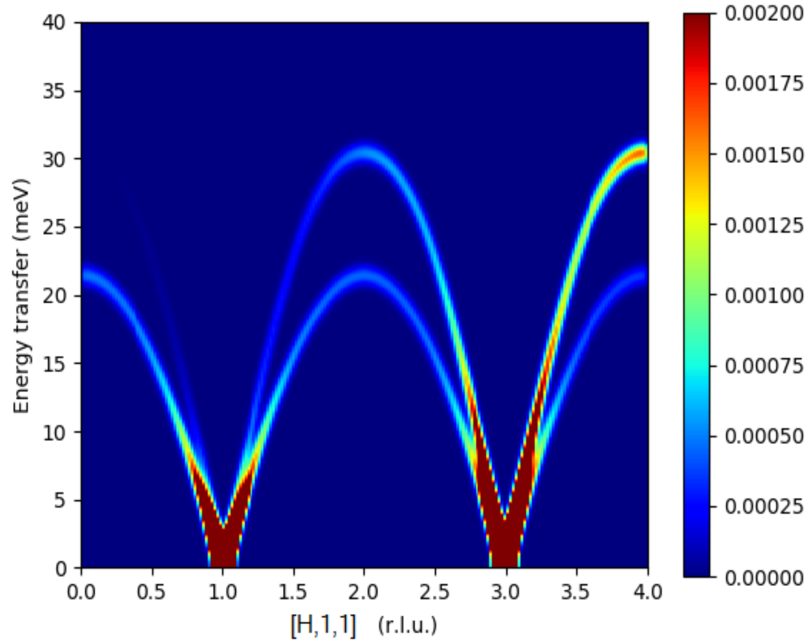


# Calculated S(Q,ω) map and various sampling trajectories

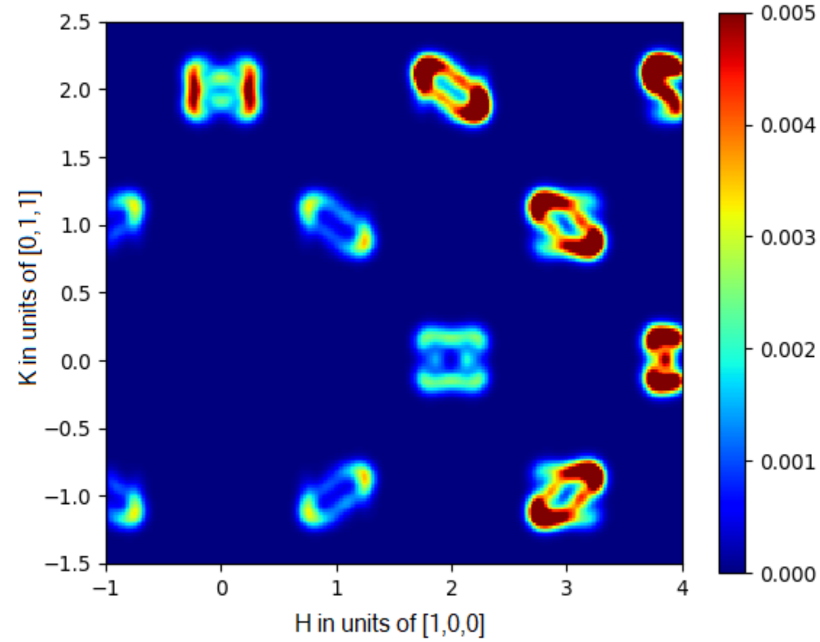


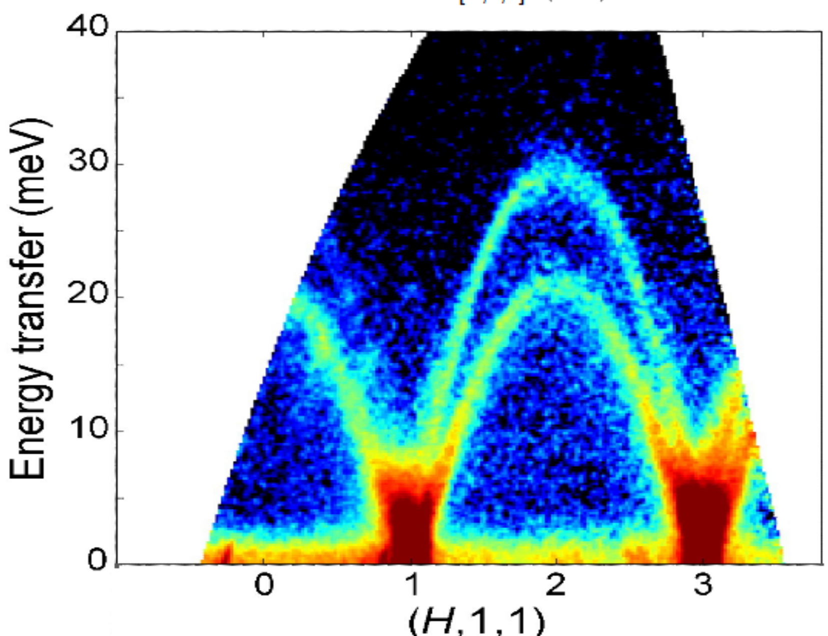

Cheng Y.Q., Daemen L.L., Kolesnikov A.I., Ramirez-Cuesta A.J., "Simulation of inelastic neutron scattering spectra using OCLIMAX", Journal of Chemical Theory and Computation, 15, 3, 1974-1982 (2019).

# OCLIMAX example: single crystal



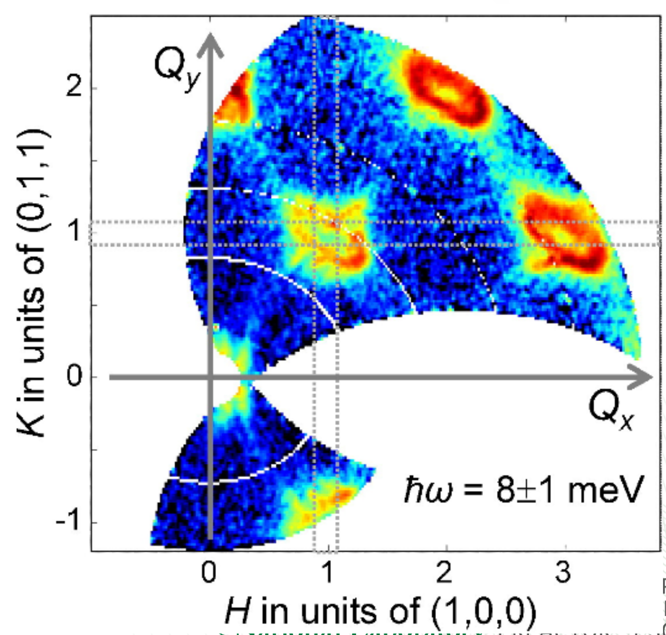

**OCLIMAX**



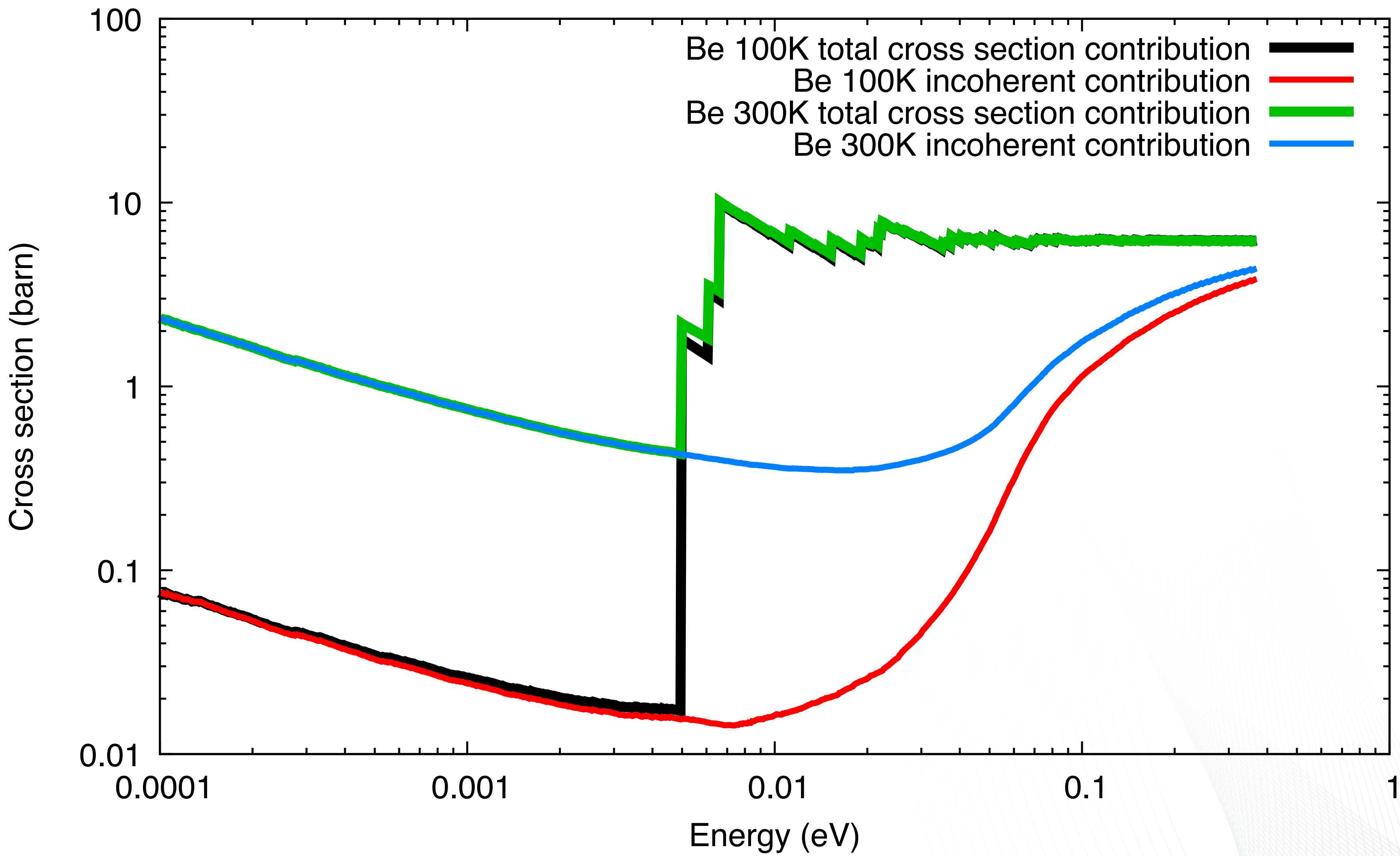

**4SEASONS @J-PARC**

- ✓ **Validating phonon frequencies, polarization vectors, and force constants**
- ✓ **Understanding phonon anomalies.**




H. Seto et al. Biochimica et Biophysica Acta, **1861**, 3651-3660 (2017).

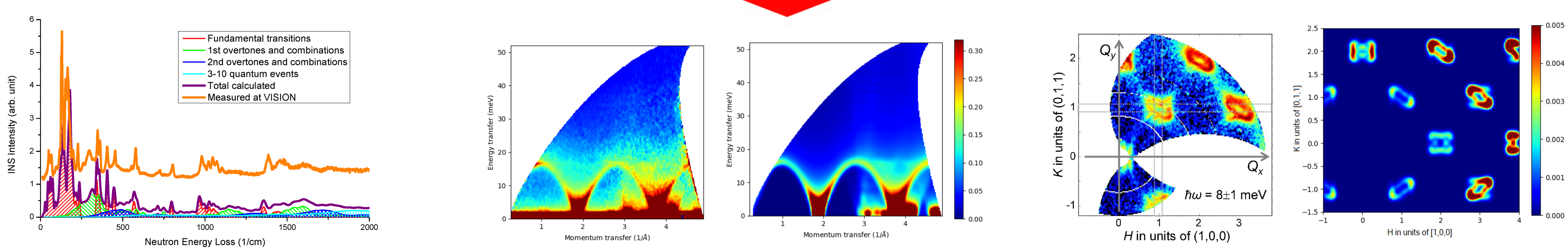
# Total cross sections for solids from first principles calculations



# OCLIMAX bridges theory and INS experiments

QUANTUM ESPRESSO · VASP (Vienna Ab-initio Simulation Package) · CASTEP · CP2K · CRYSTAL (a computational tool for solid state chemistry and physics) · ORCA · NWCHEM (HIGH-PERFORMANCE COMPUTATIONAL CHEMISTRY SOFTWARE) · RMG · phonopy 2.0.0

docker · OCLIMAX · ICEMAN




VISION, CNCS, HYSPEC, SEQUOIA, ARCS and many other neutron spectrometers.


ICEMAN
Triple Axis
ARCS
CNCS
HYSPEC
SEQUOIA
VISION


ICEMAN
BASIS
Triple Axis
ARCS
CNCS
HYSPEC
SEQUOIA
VISION

# Traditional methods

- Minimization, least squares algorithms
- Sequentially fitting QENS functions to data
- Parameters collected
- Plot parameters vs Q
- Fit parameters to functionality

- Laborious, time consuming
- Tedious, error prone method
- It is very difficult to track what's been done to the data

$$I(Q,E) = [f_{eisf}(Q)\frac{1}{\pi}\frac{\Gamma_n(\mathrm{Q})}{\Gamma_n(\mathrm{Q})^2 + E^2} + (1 - f_{eisf}(Q))\frac{1}{\pi}\frac{\Gamma_b(\mathrm{Q})}{\Gamma_b(\mathrm{Q})^2 + E^2}]$$
$$\otimes R(Q,E) + (C_1(Q)E + C_2(Q))$$

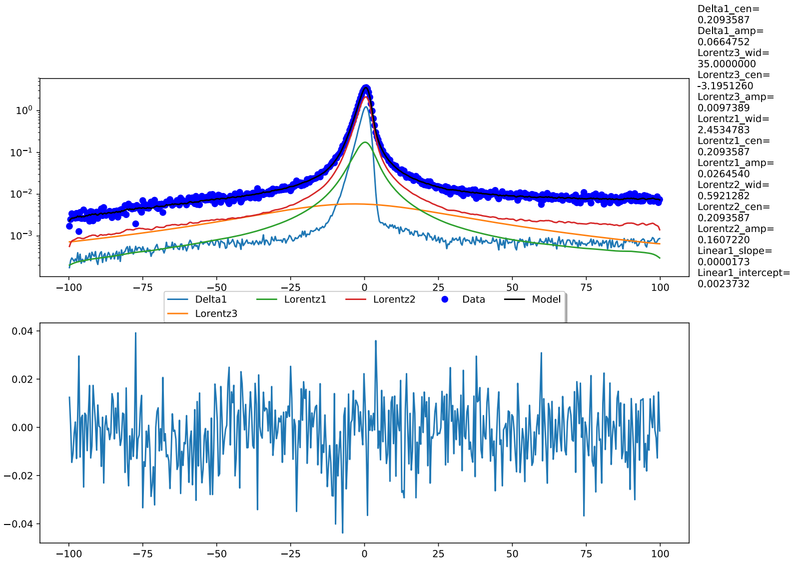
QClimax

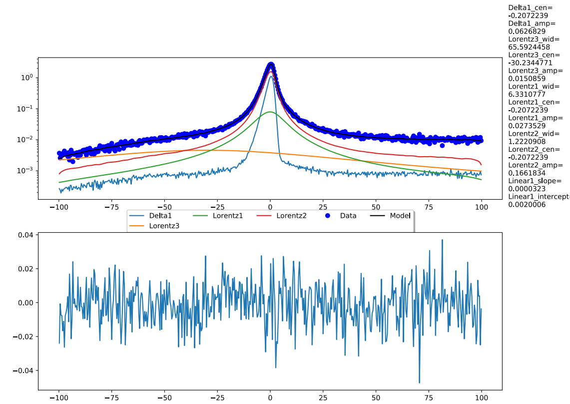
## Non-Linear Least-Squares Minimization and Curve-Fitting for Python

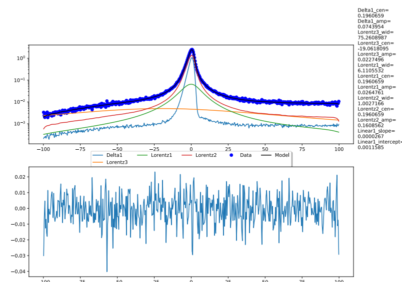
*Release 0.9.6*

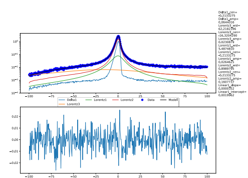
Matthew Newville, Till Stensitzki, and others

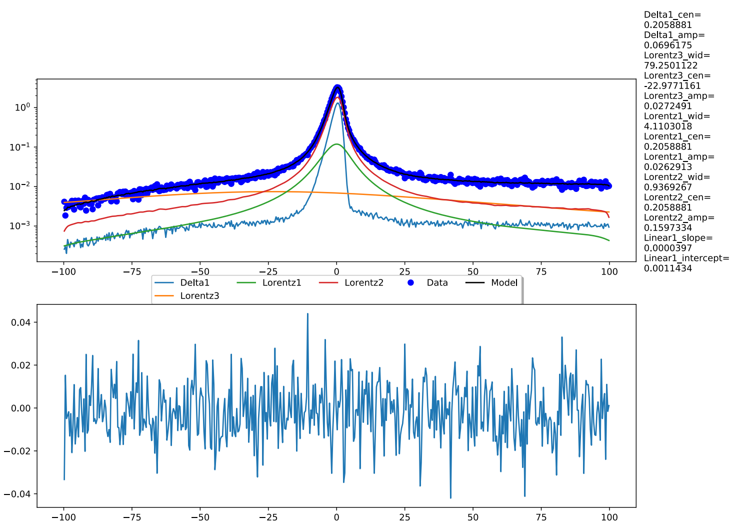
# Qclimax







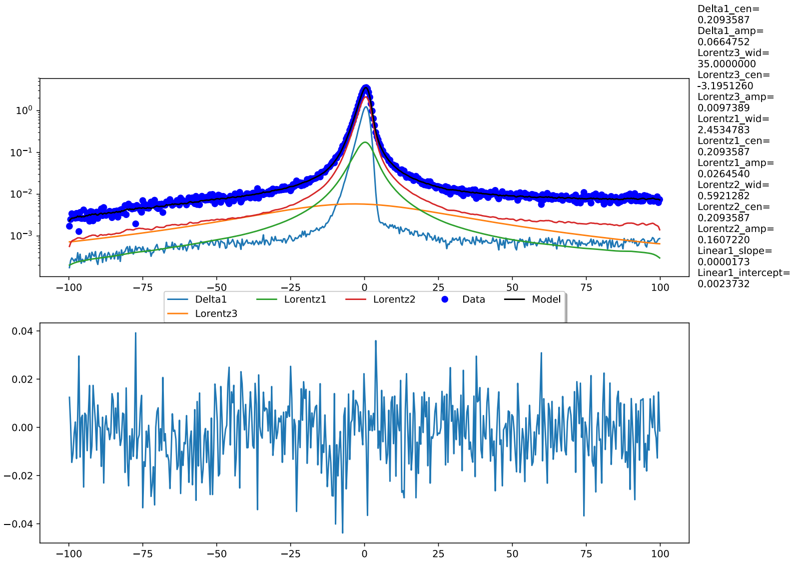

Global fitting of the data

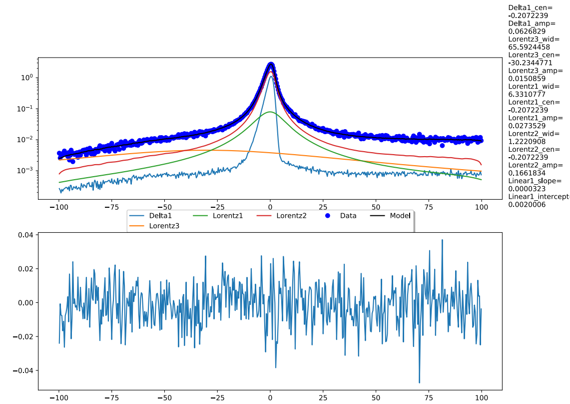
Fitting data using parameter while imposing constraints overt this parameter:

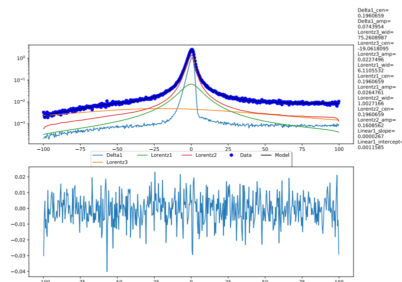
$$FWHM = f(\alpha, \beta, \gamma, \cdots, \omega, \boldsymbol{Q})$$

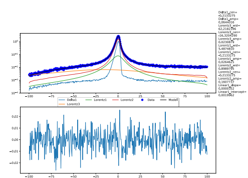
Returning best fit values for $\alpha, \beta, \gamma \cdots$ and so forth

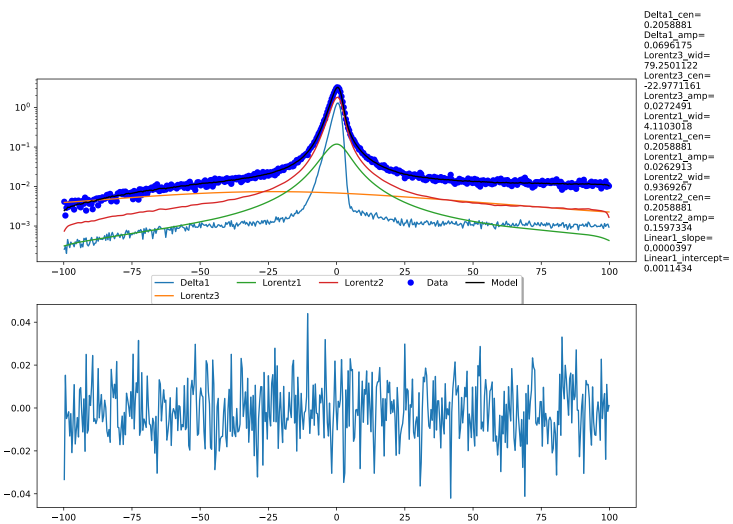
# Qclimax





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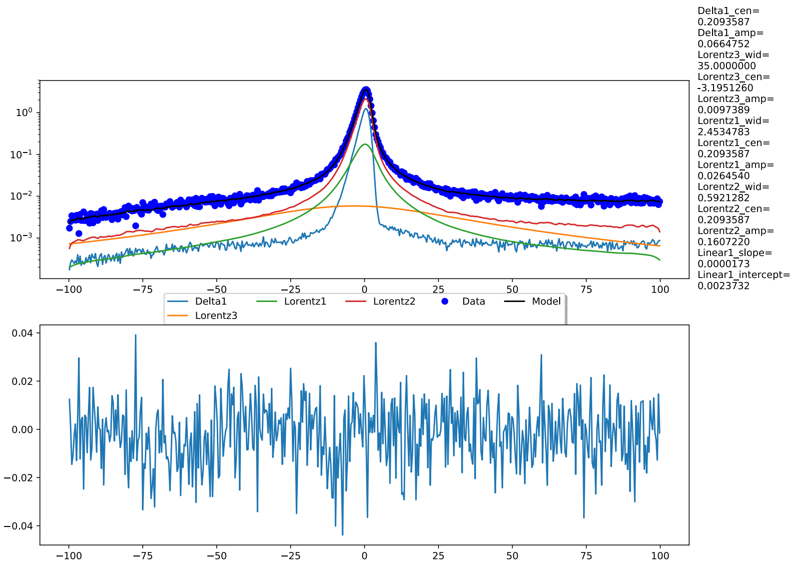
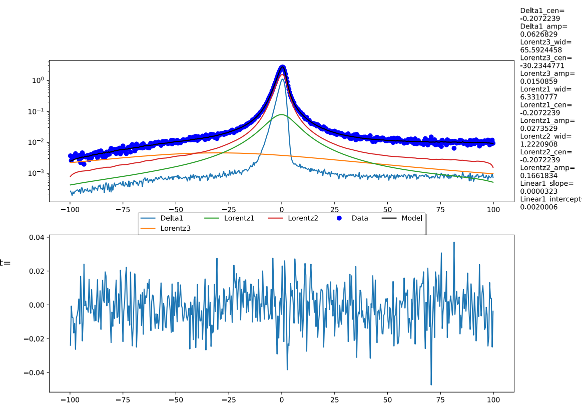
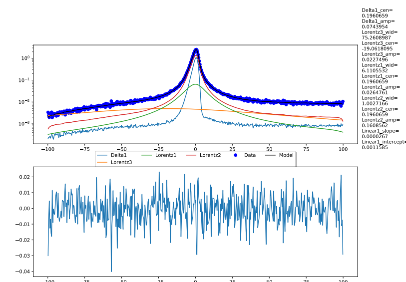
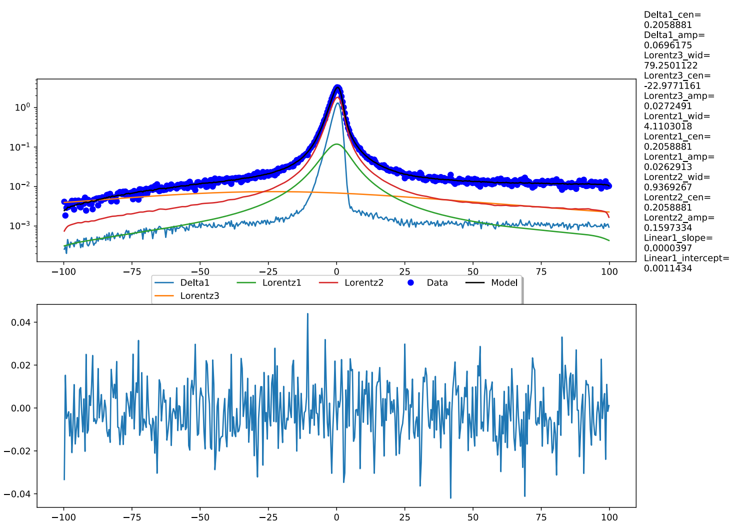
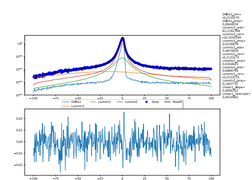
# Qclimax



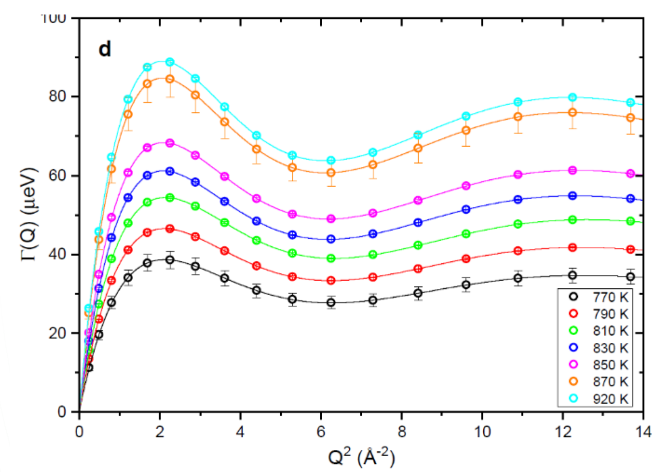





Example:
Chudley-Elliot fit to hydrogen in metal hydrides



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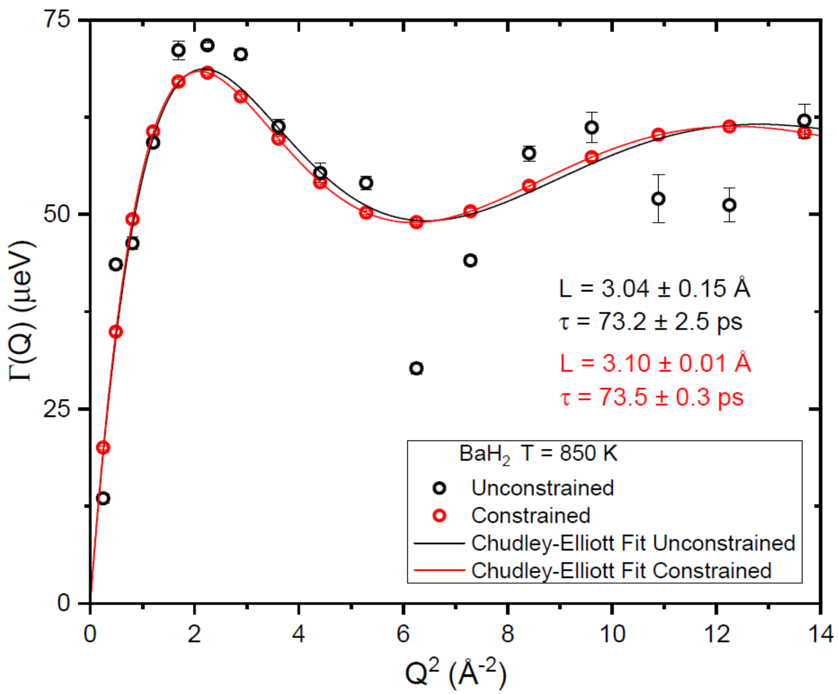
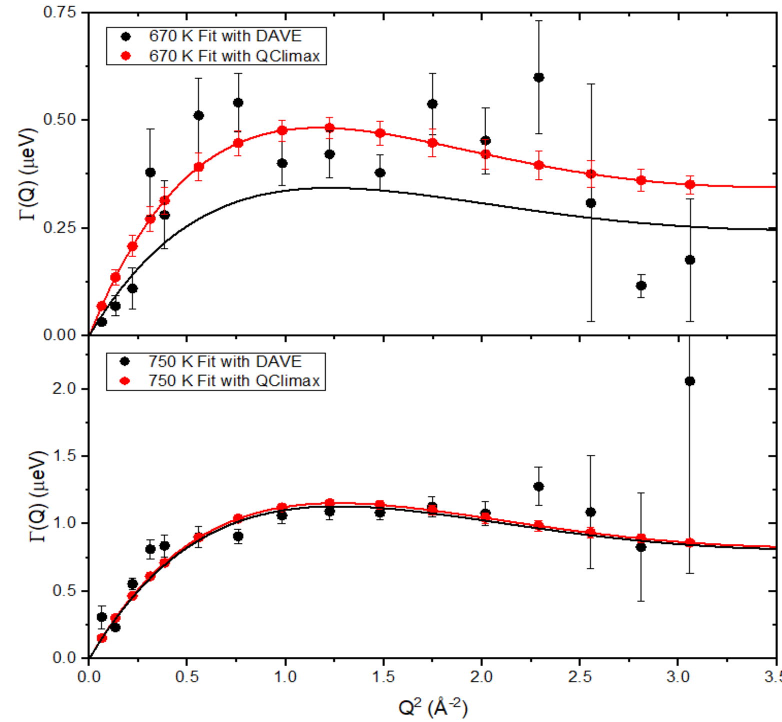
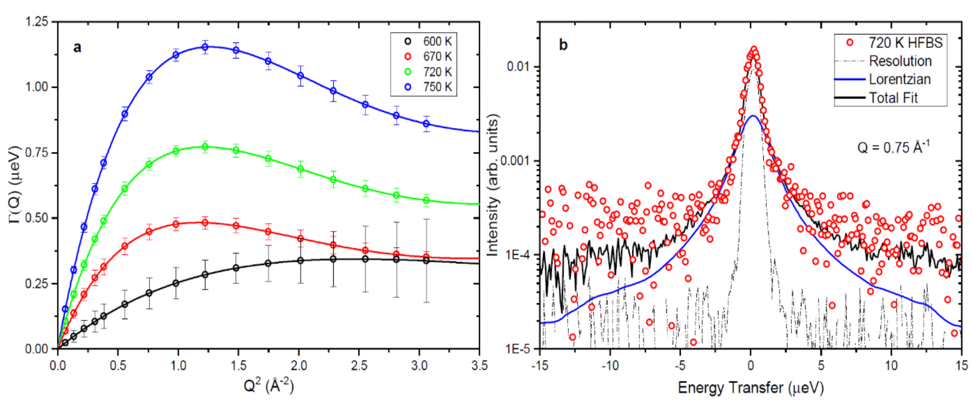
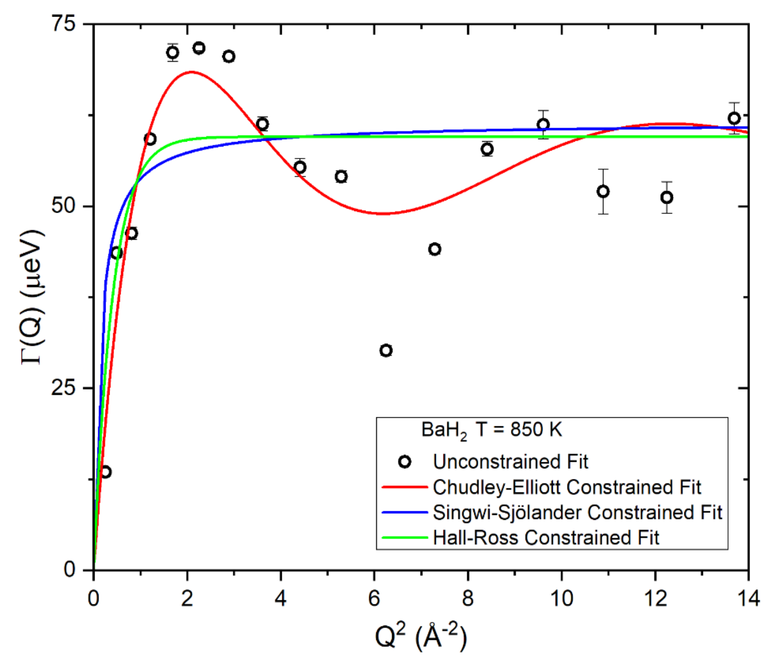
# Bayesian behaviour



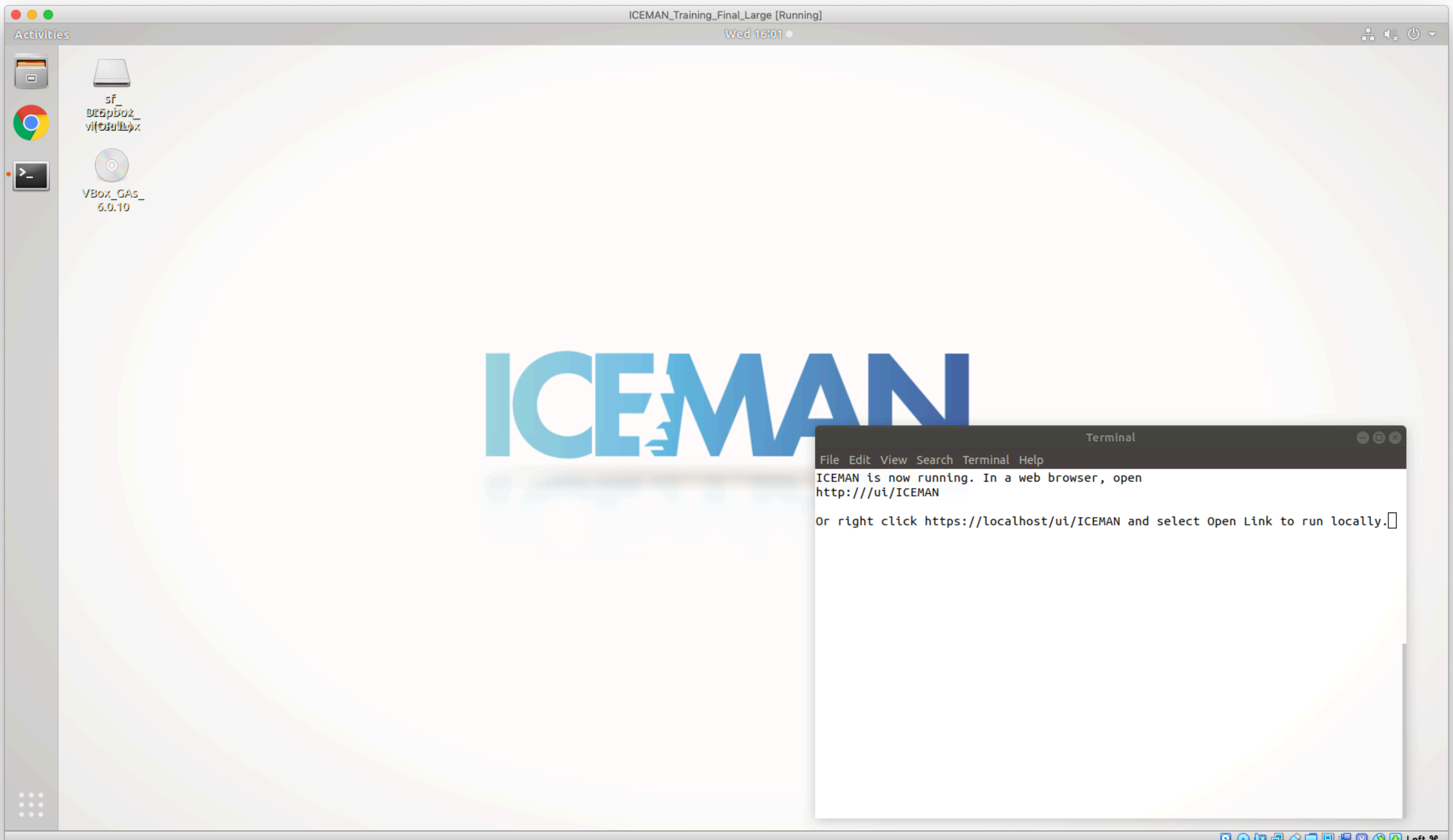



# Many flavours

- Docker container

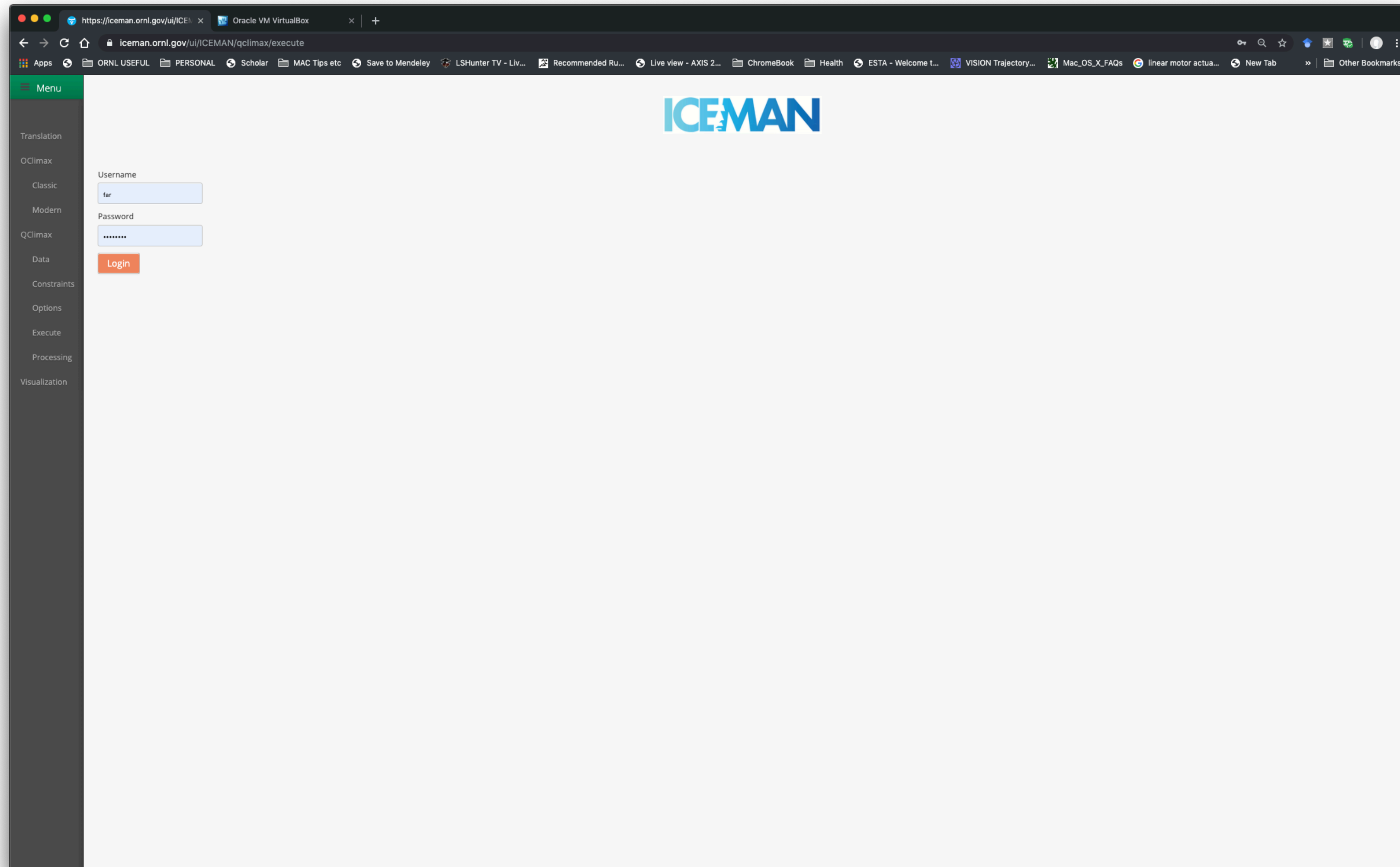
# Many flavours

- Virtual machine

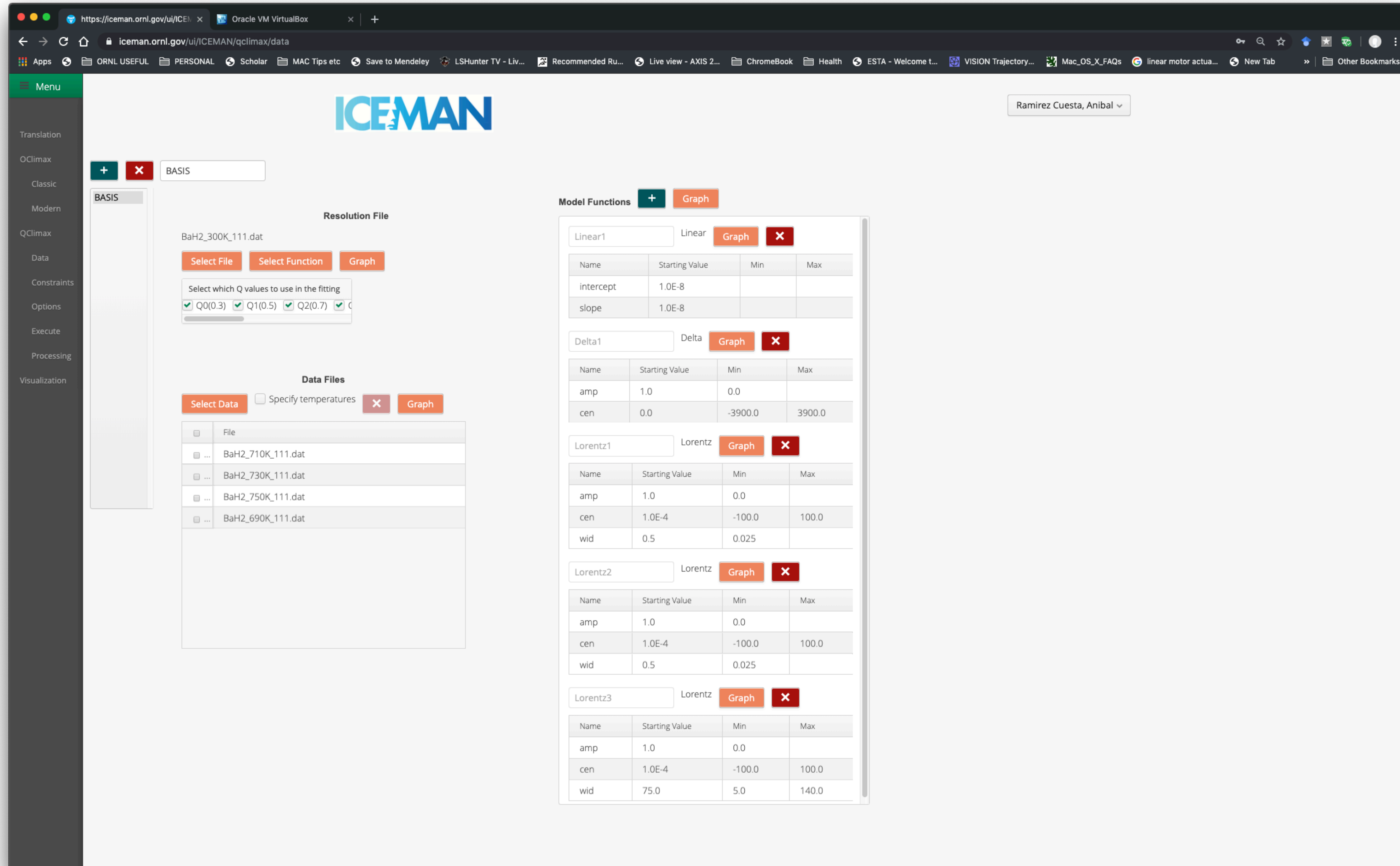


# Many flavours

- Web access (hosted at ORNL, CADES)



# Simple interface



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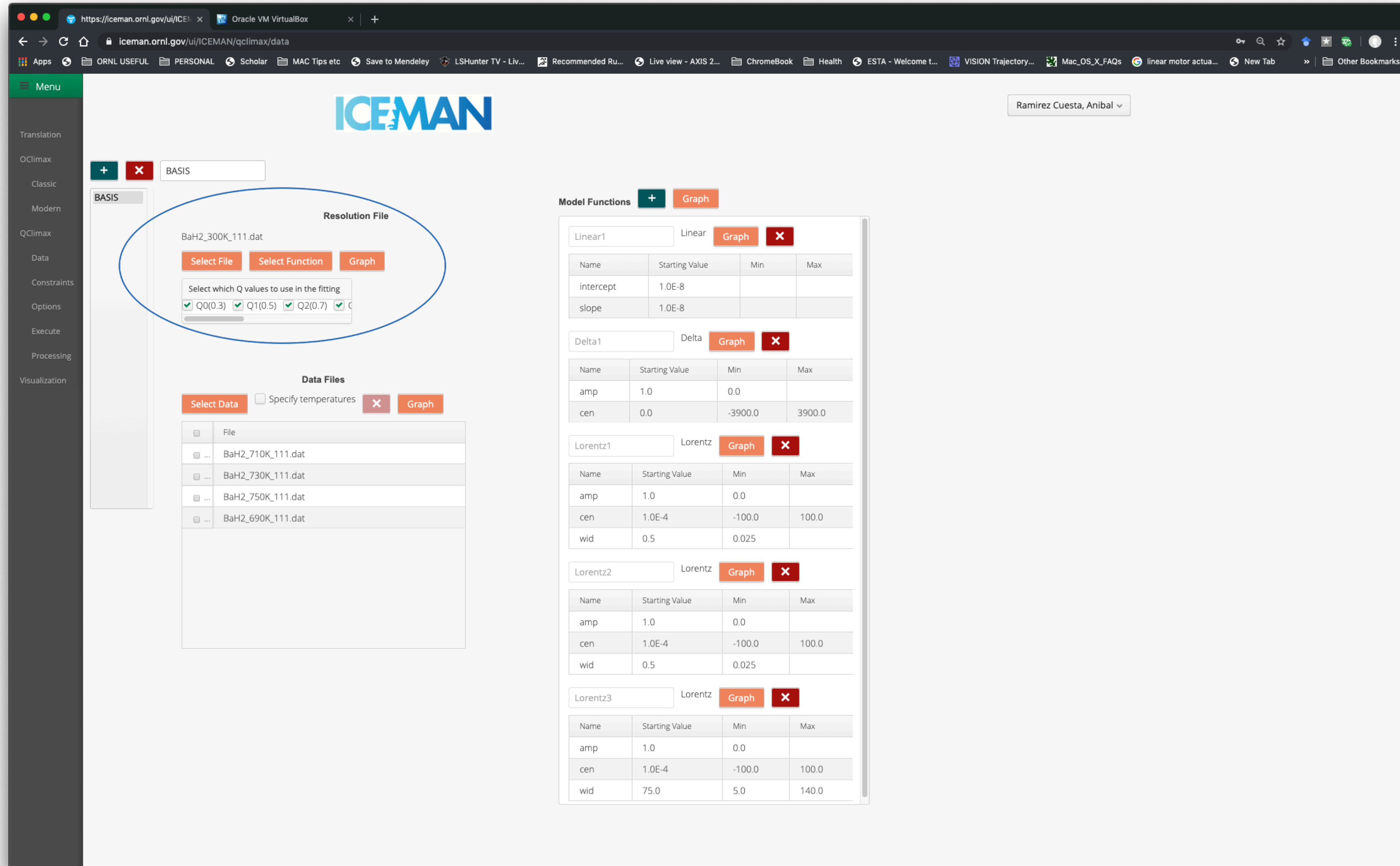

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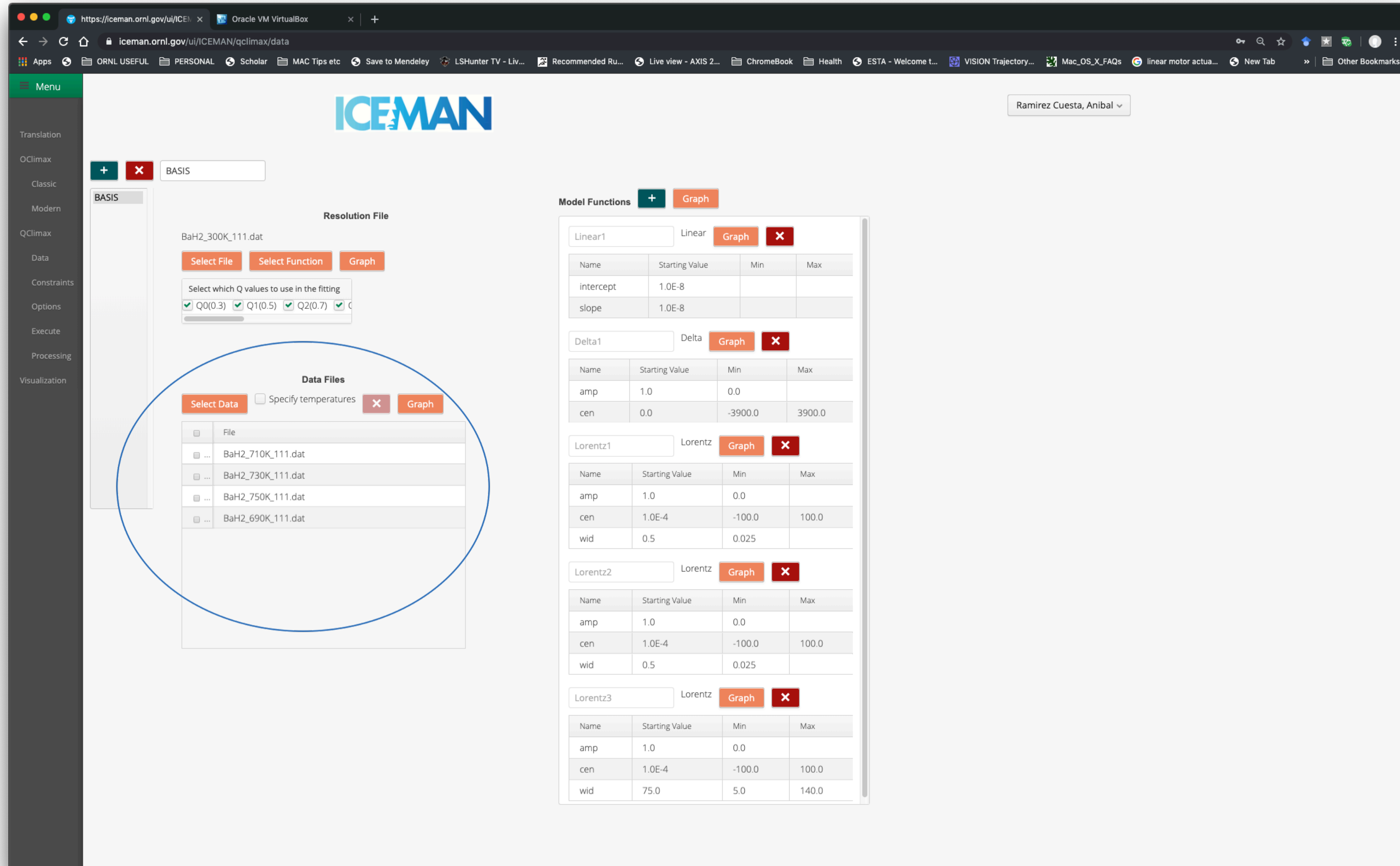

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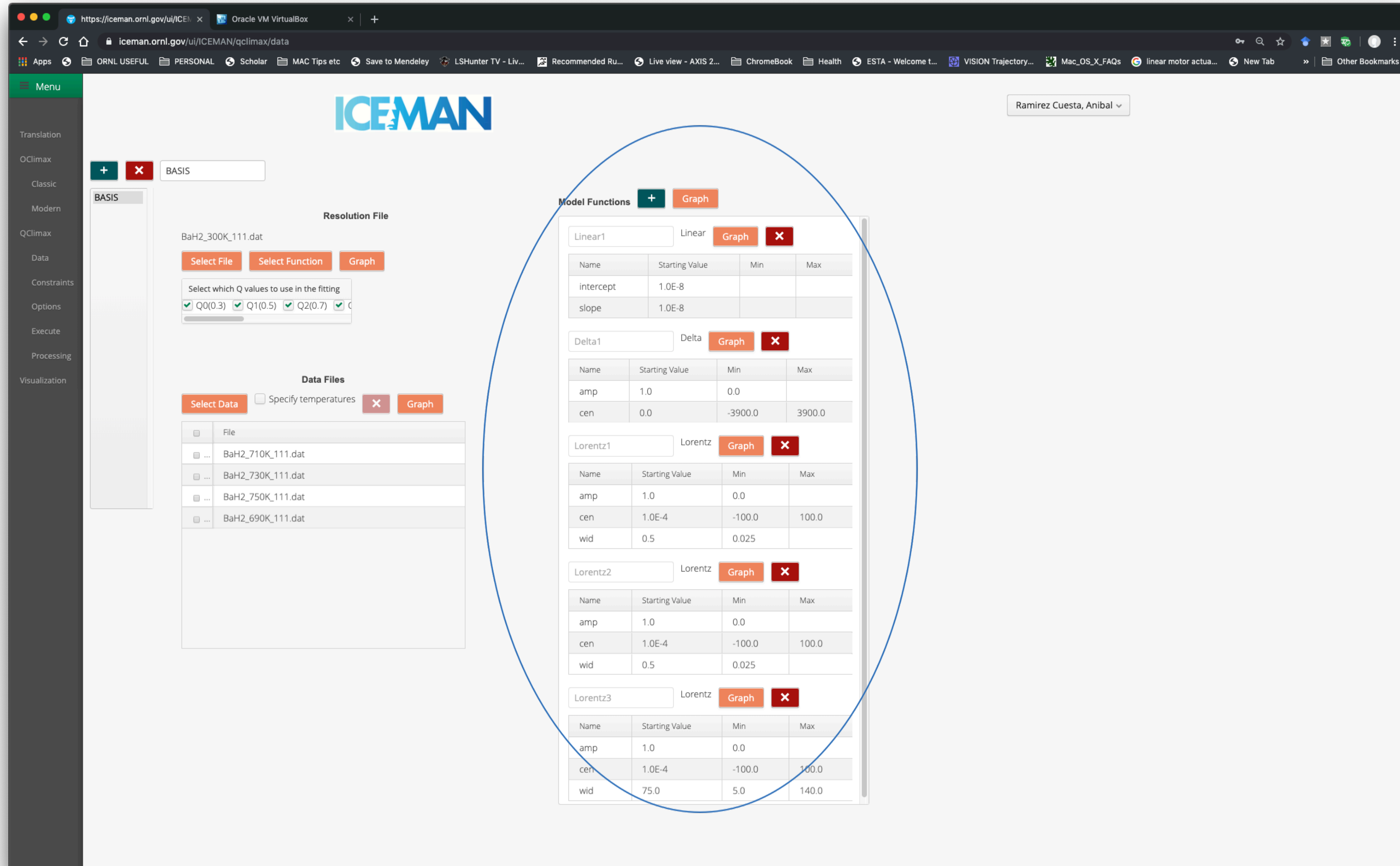
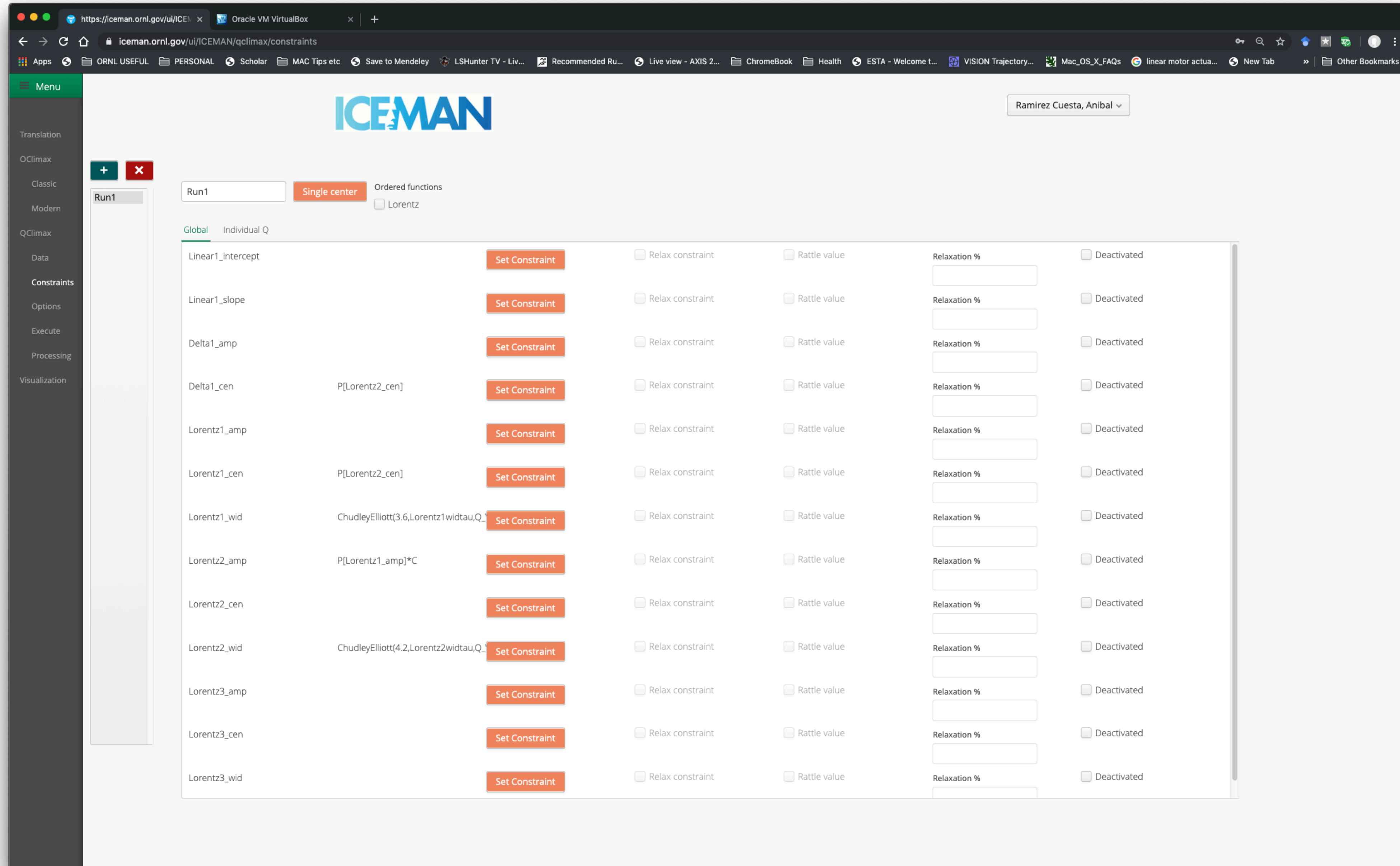
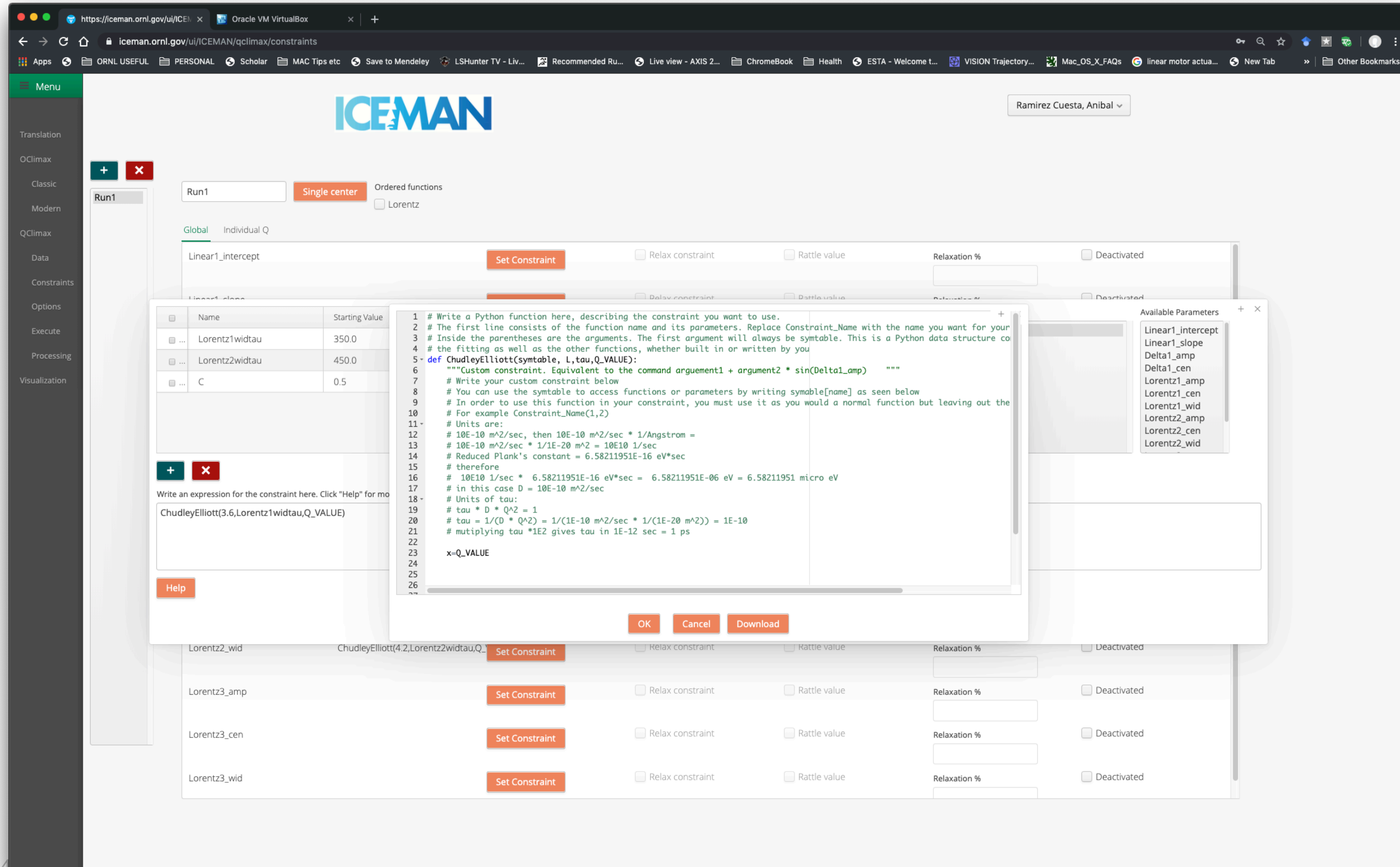


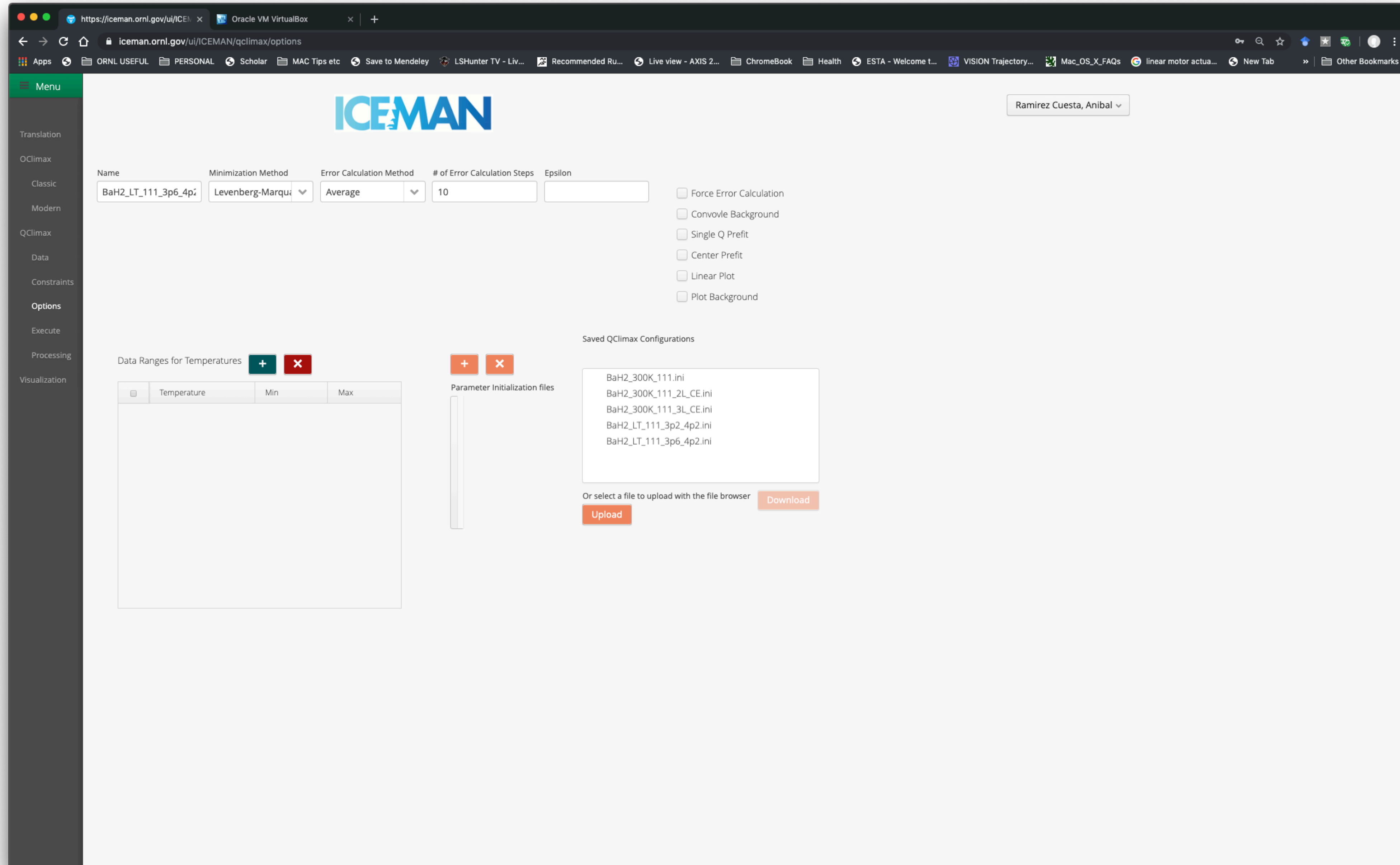
# Use your constraints



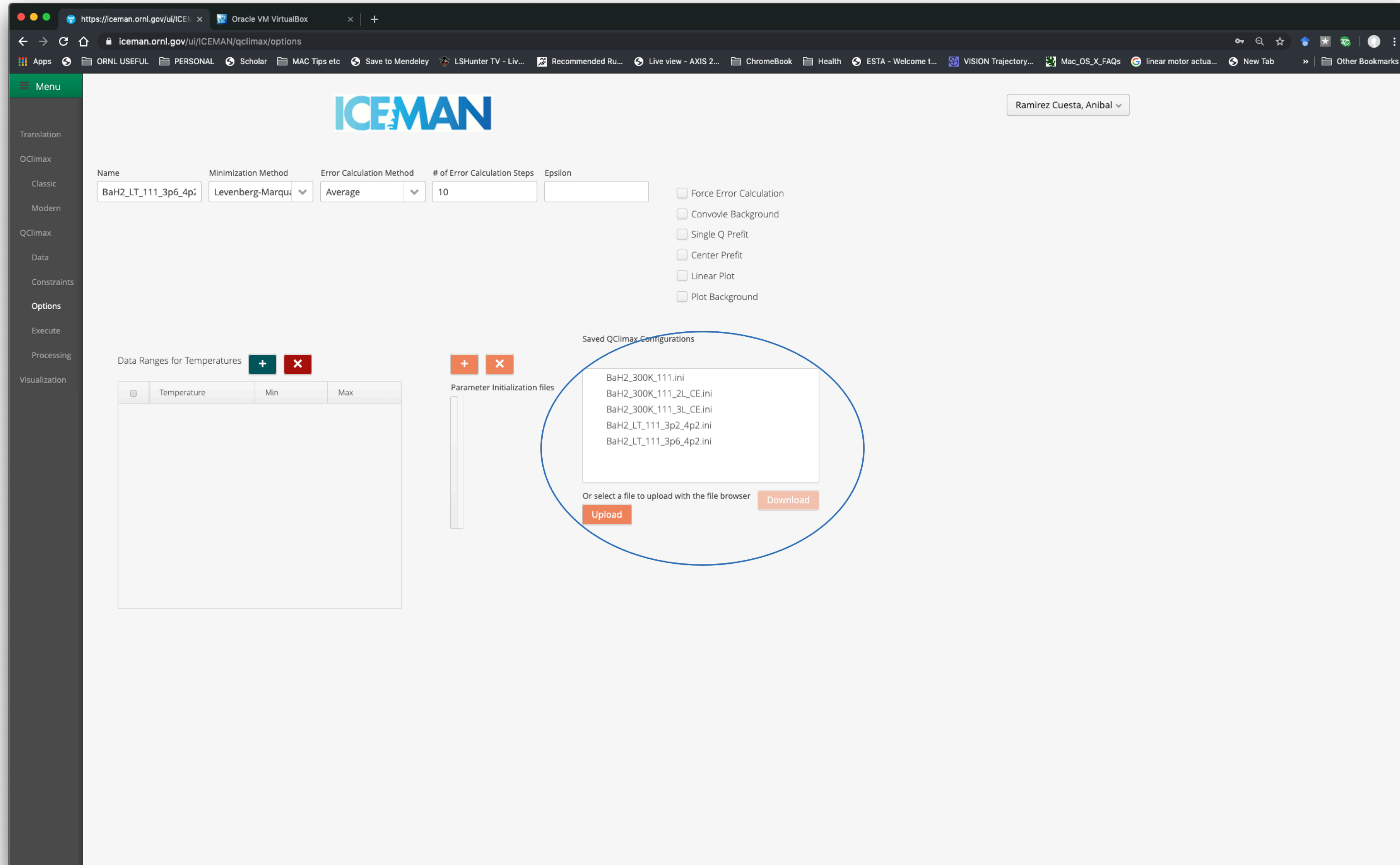
# Constraints not there?
# No problem write them in python!



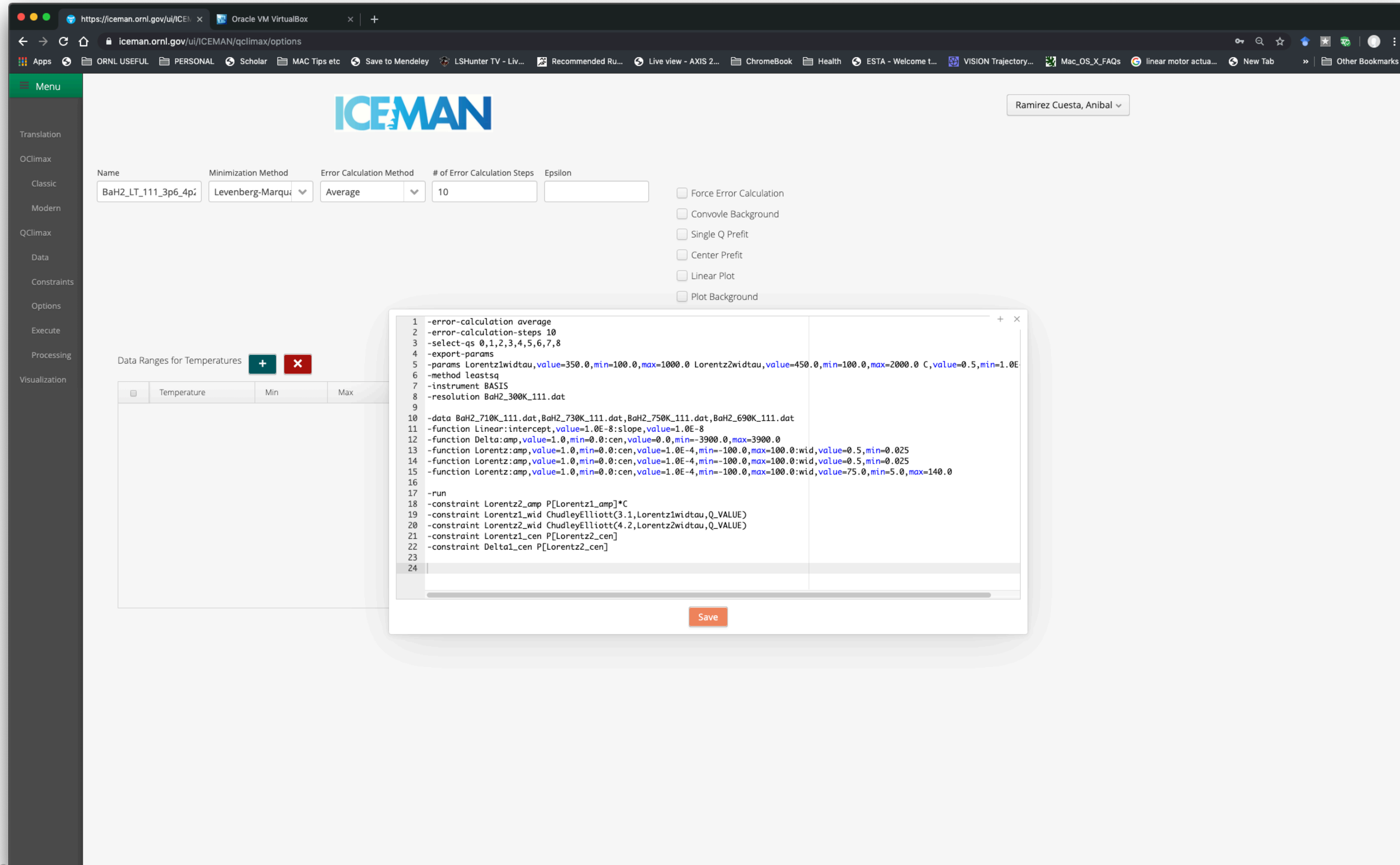
# Select methods



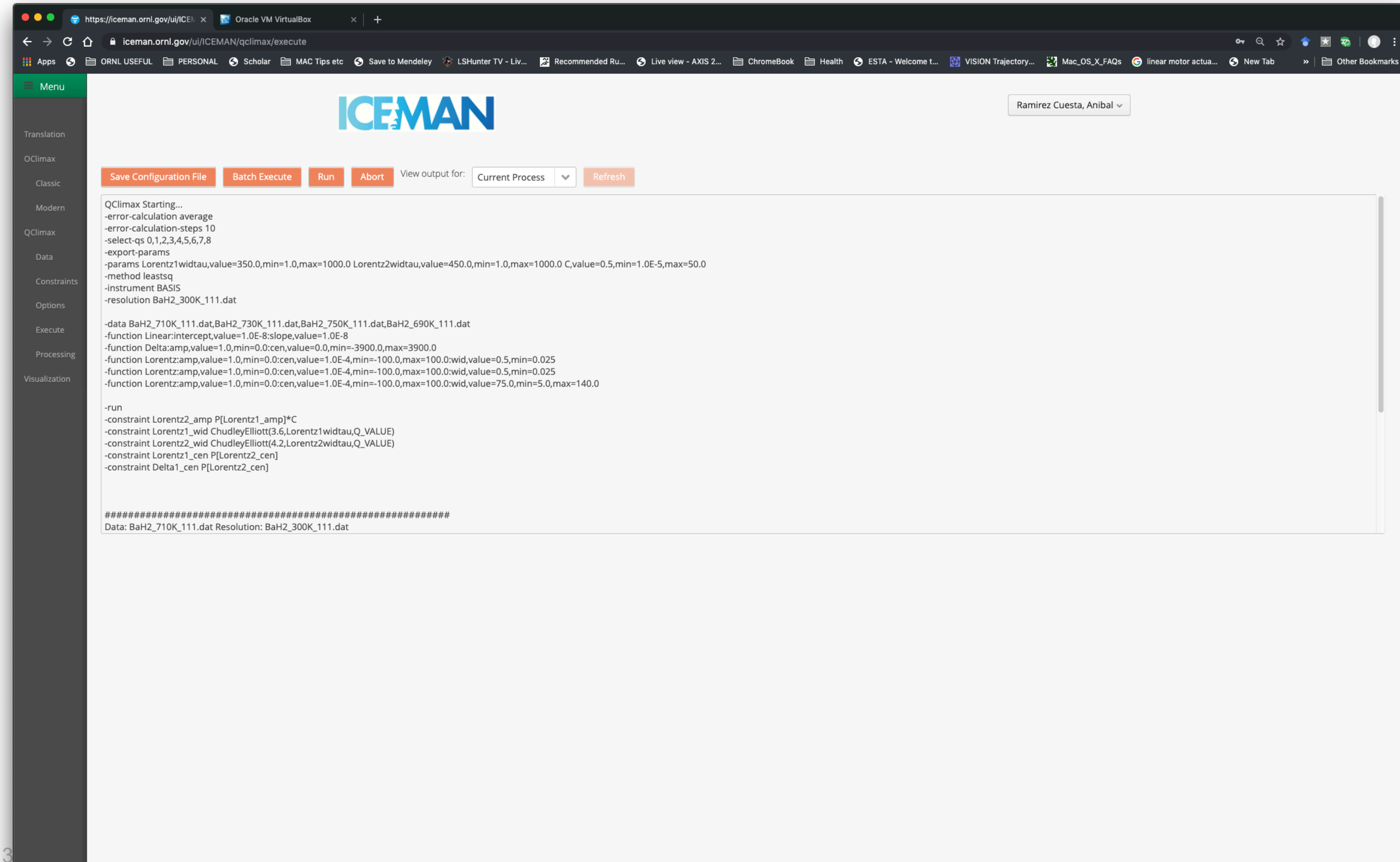
# Repeat old runs



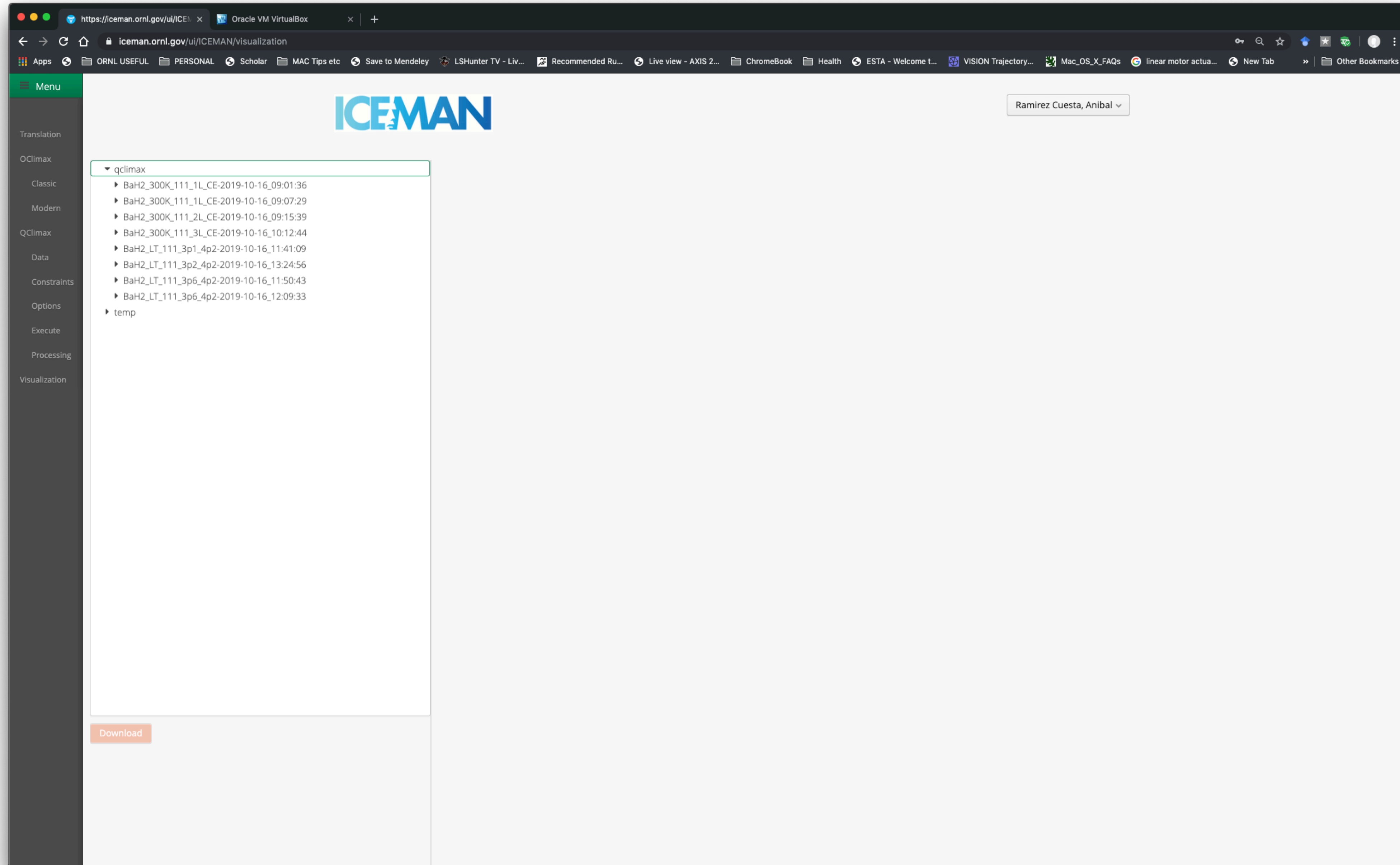
# Modify old runs, make expert changes



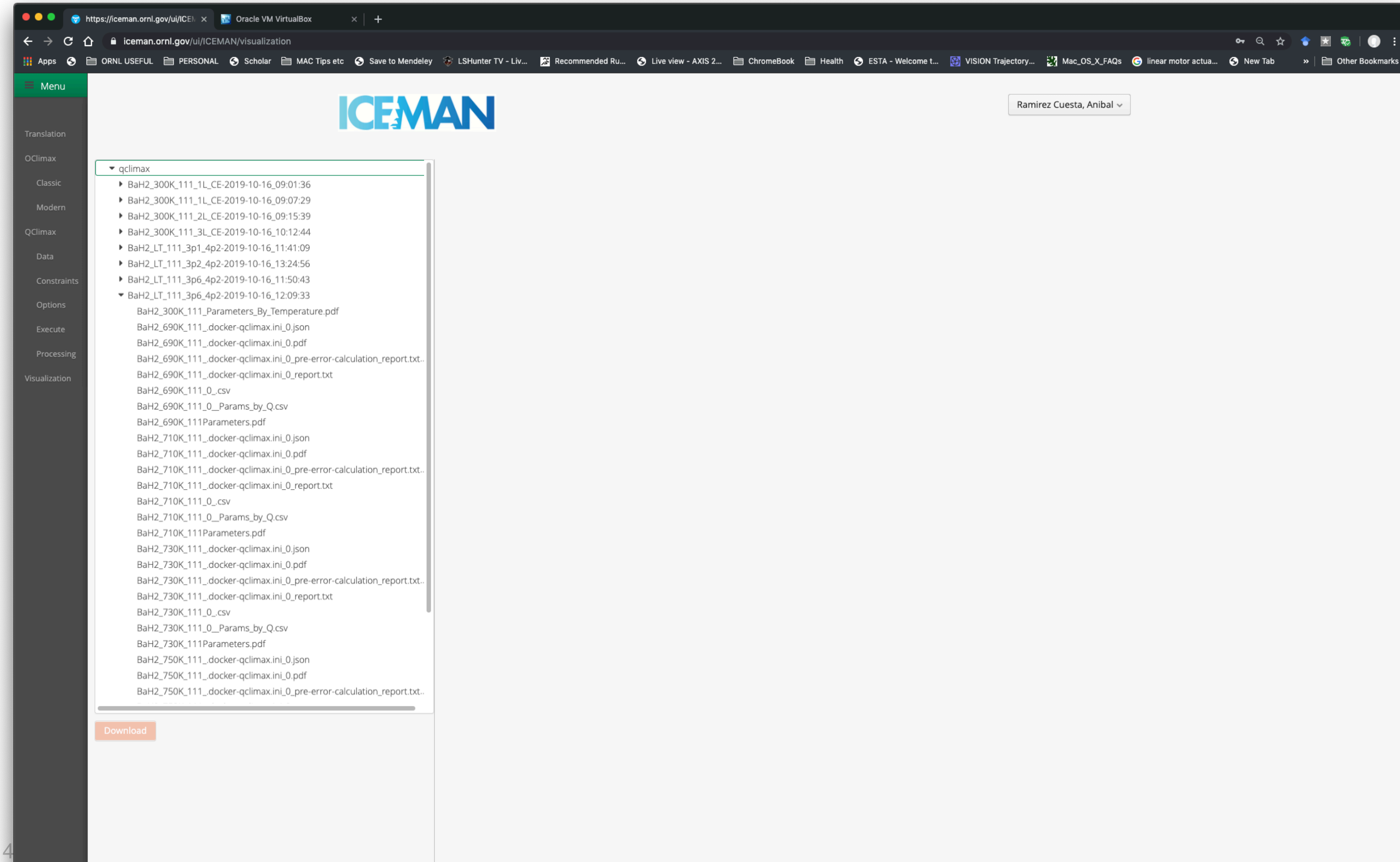
# Run fittings



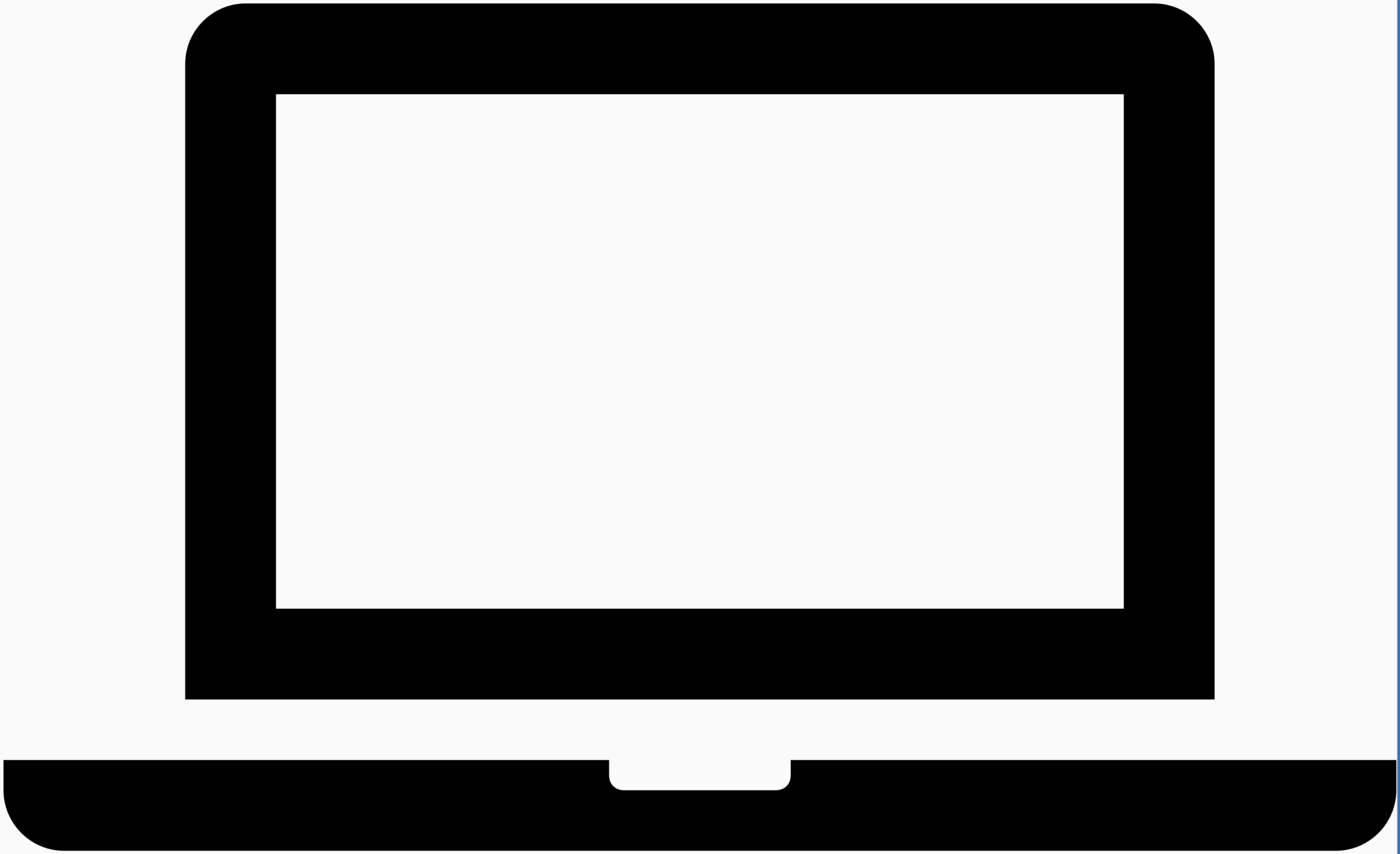
# Visualize, edit results, get new starting configurations, download etc.



# Visualize, edit results, get new starting configurations, download etc.



# Structure of heterogeneous software



User computer Linux, mac, PC

# Structure of heterogeneous software


User computer Linux, mac, PC

# Structure of heterogeneous software

docker

VirtualBox

User computer Linux, mac, PC

# Structure of heterogeneous software

Docker Container
Translations PYTHON

Docker Container
Translations PERL

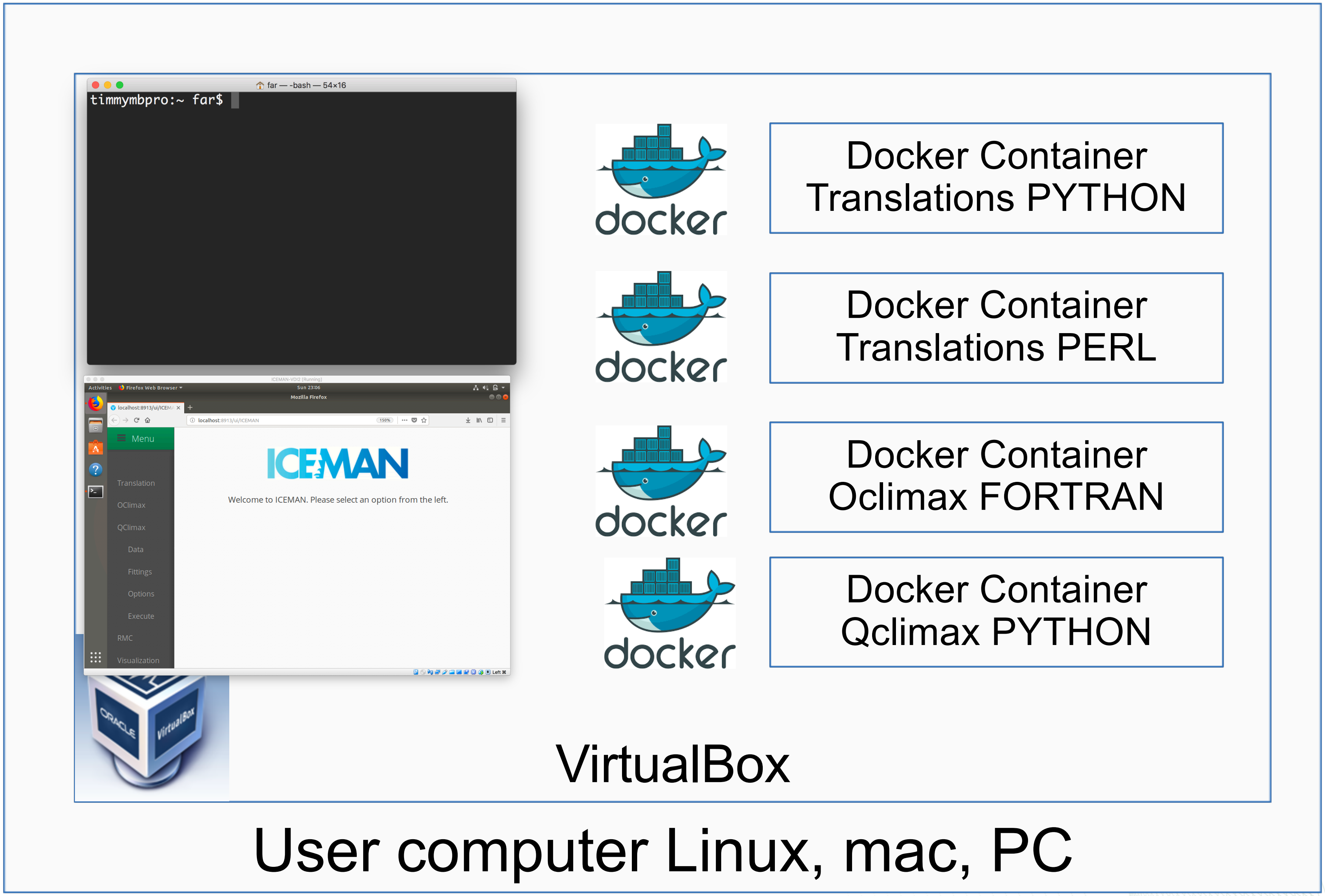
Docker Container
Oclimax FORTRAN

Docker Container
Qclimax PYTHON

VirtualBox

User computer Linux, mac, PC

# Structure of heterogeneous software



# Conclusion

- Instrumental needs in VISION at SNS require respectable computer resources (1600+ cores cluster today ) to help interpret the data
- The VirtuES cluster is available for users to run mostly DFT
- Software to generate inelastic neutron spectra from calculations to directly compare with experimental data has been developed (Oclimax)
- Software to analyze QENS data using global fitting and easy interface and extensibility in the fitting functions and constraints is also available.
- Virtual machine technologies, VirtualBox and Docker are used to produce software that is easy to maintain, expand and that can be operated from the command line and a web interface if desired.
- Secondary objective for software is to automate processes by exchanging file formats and generate a number of input/output files for different codes
- And much more…
- These codes provide the basis for a different approach to automation and reproducibility of neutron data analysis as well as routine integration of computer modeling in neutron scattering.
- Questions?