Cleaning coulomb explosion data via random coincidence subtraction

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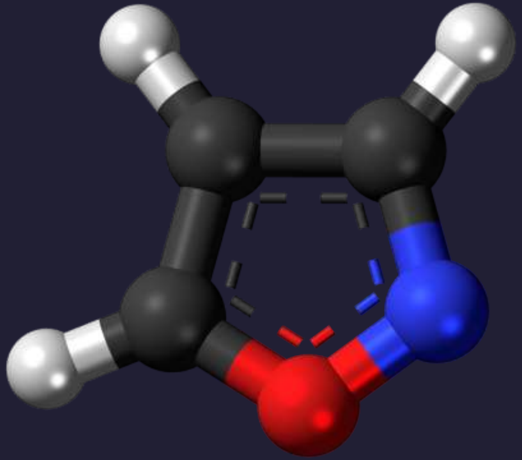
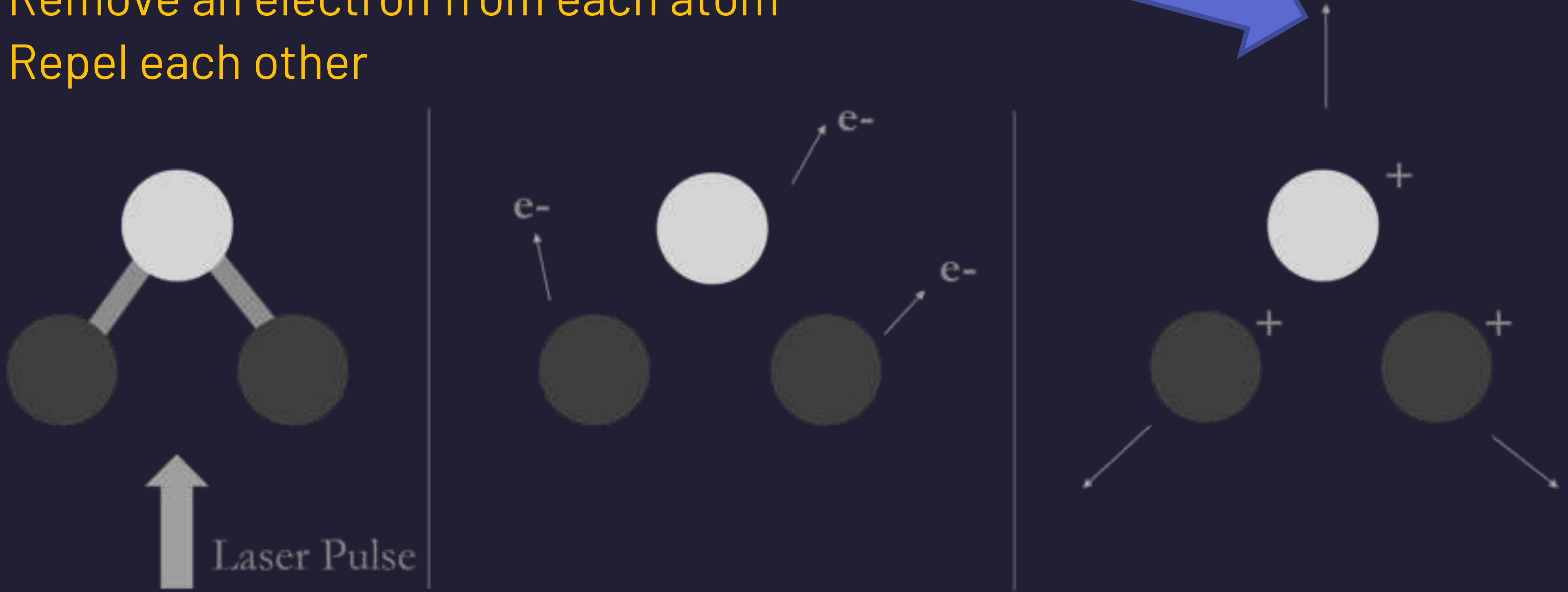


Coulomb Explosion Imaging

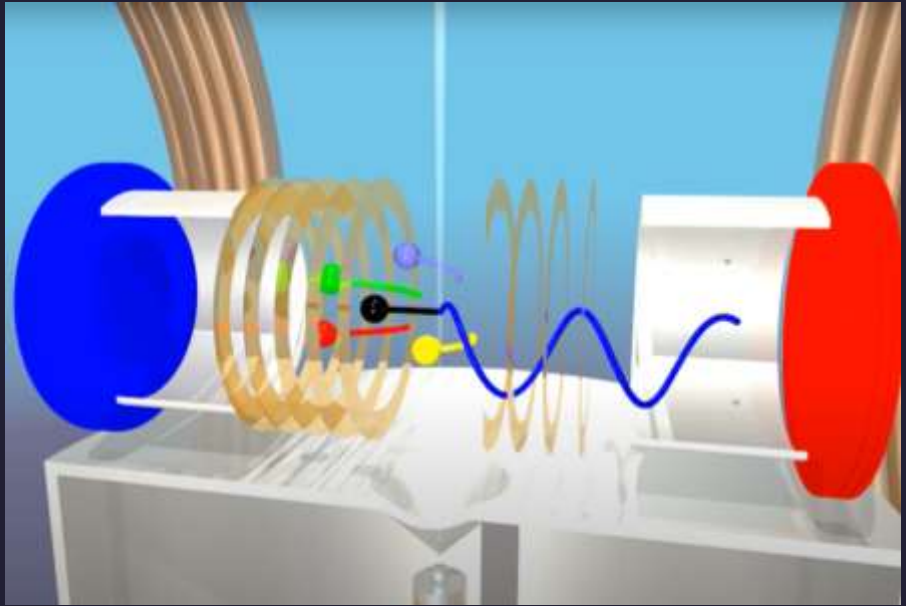
We measure these
(insight into the structure)

- Ionize a sample
    - Remove an electron from each atom
    - Repel each other



Coulomb Explosion Imaging

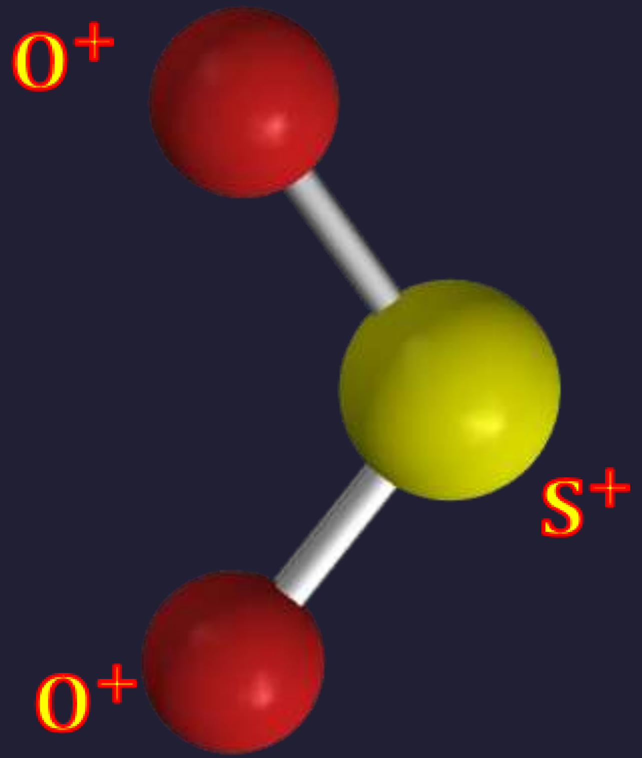
- Atoms now have charge and fall to the detector via the electric field
- Observe when and where the ions hit the detector
  - Get momenta



Pitzer, M., Fehre, K., Kunitski, M., Jahnke, T., Schmidt, L., Schmidt-Böcking, H., Dörner, R., Schöffler, M. Coulomb Explosion Imaging as a Tool to Distinguish Between Stereoisomers. J. Vis. Exp. (126), e56062, doi:10.3791/56062 (2017).

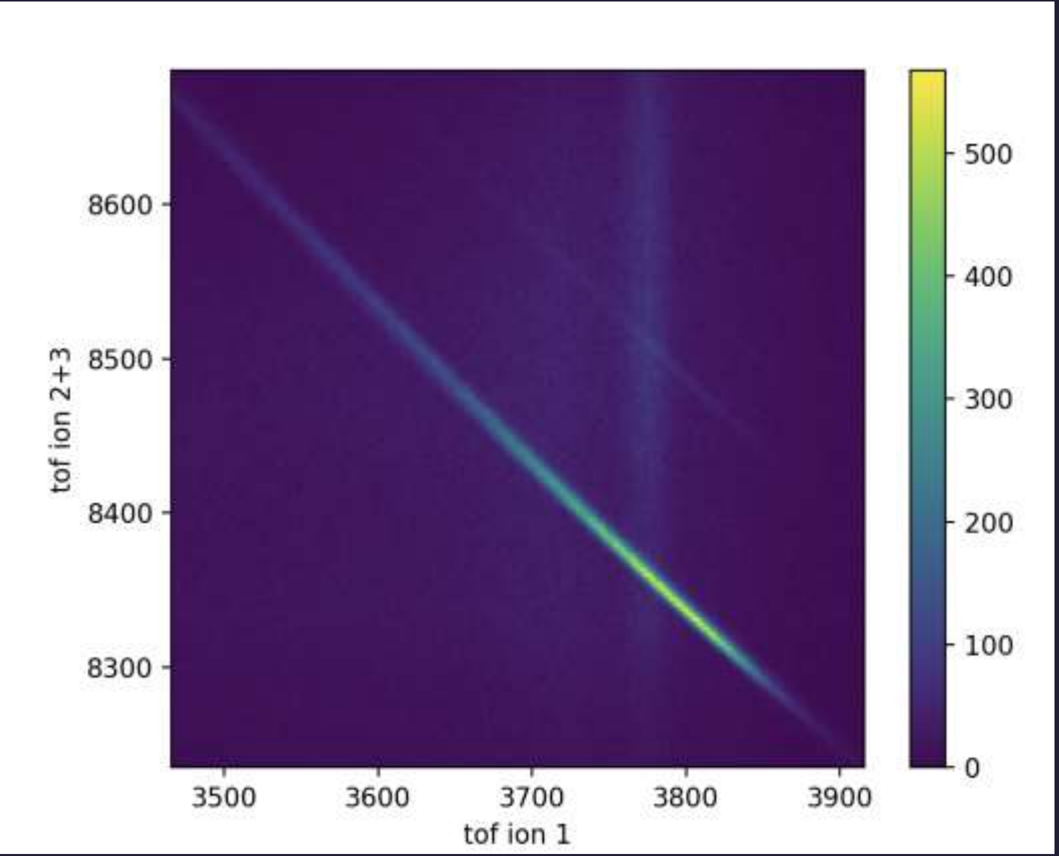
Sulfur Dioxide

Smaller Molecule



3-fold Coincidence for $SO_2 \longrightarrow$ S+ O+ O+

- "Complete Channel"
- Momentum conservation
- Easy to cut randoms
  - Leads to "clean" images



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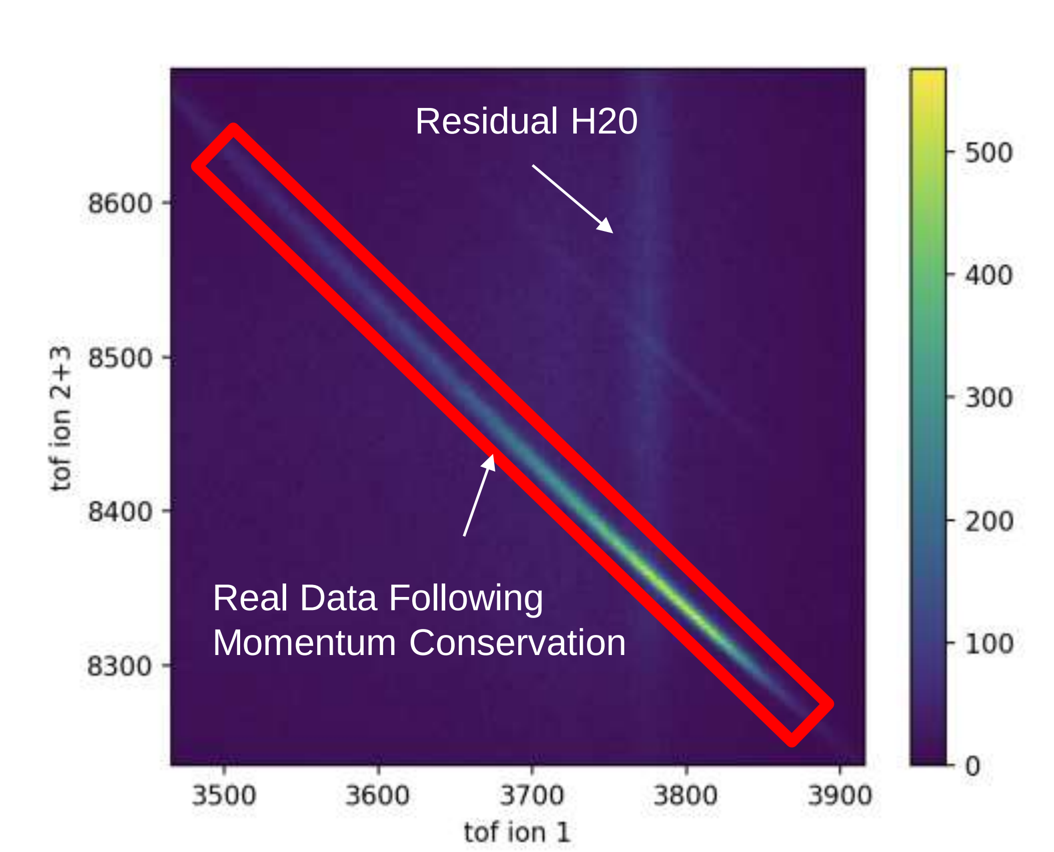
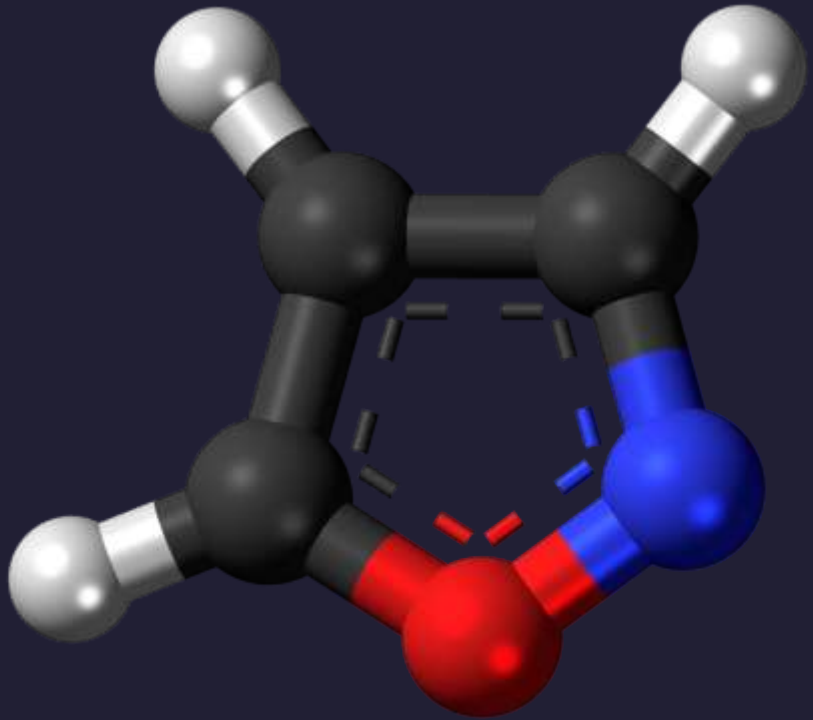


Isoxazole C_3H_3NO

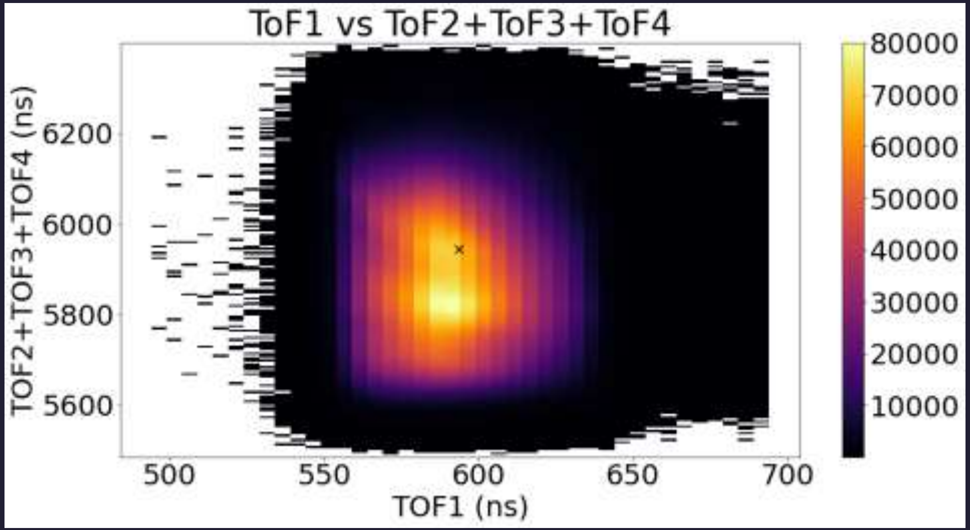
Larger Molecule



File:Isoxazole-3D-balls.png. (2020, October 30). *Wikimedia Commons*. Retrieved 19:53, July 21, 2023 from https://commons.wikimedia.org/w/index.php?title=File:Isoxazole-3D-balls.png&oldid=507693222.

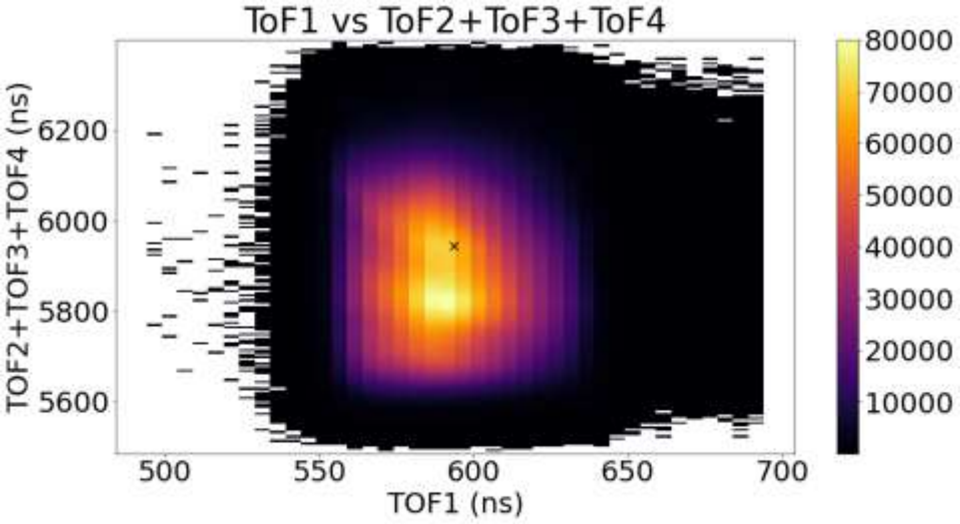
4-fold Coincidence Plot for Isoxazole H+ C+ N+ O+ Channel

- “Incomplete Channel”
- Only getting 1/2 of the ions, so can’t find momentum conservation
    - **No clear coincidence line**



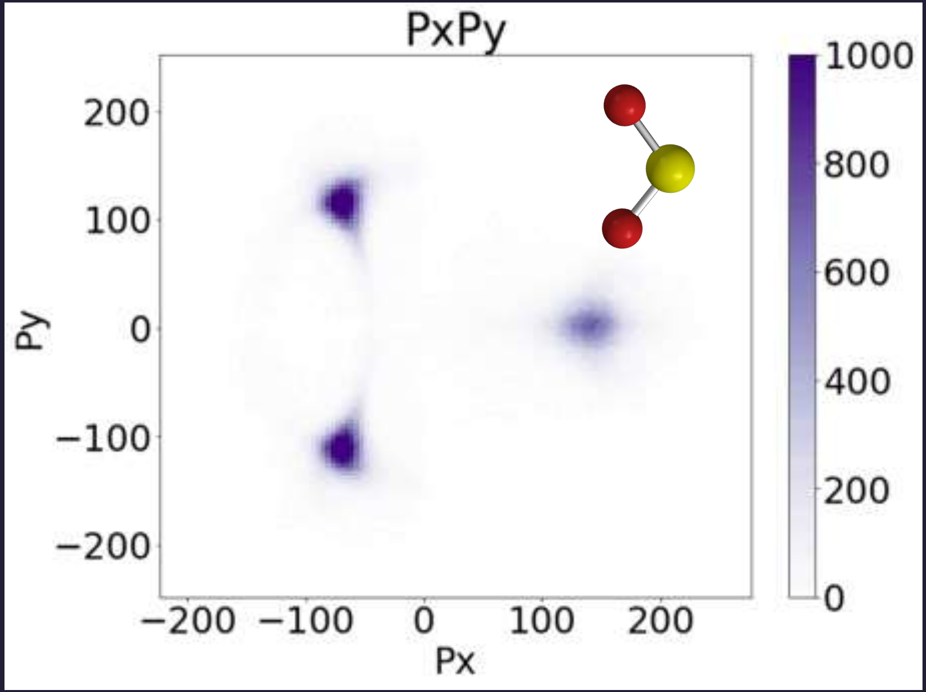
Harder to remove bad data in incomplete channels

- "Incomplete Channel"
- Only getting 1/2 of the ions, so can't find momentum conservation
  - **No clear coincidence line**

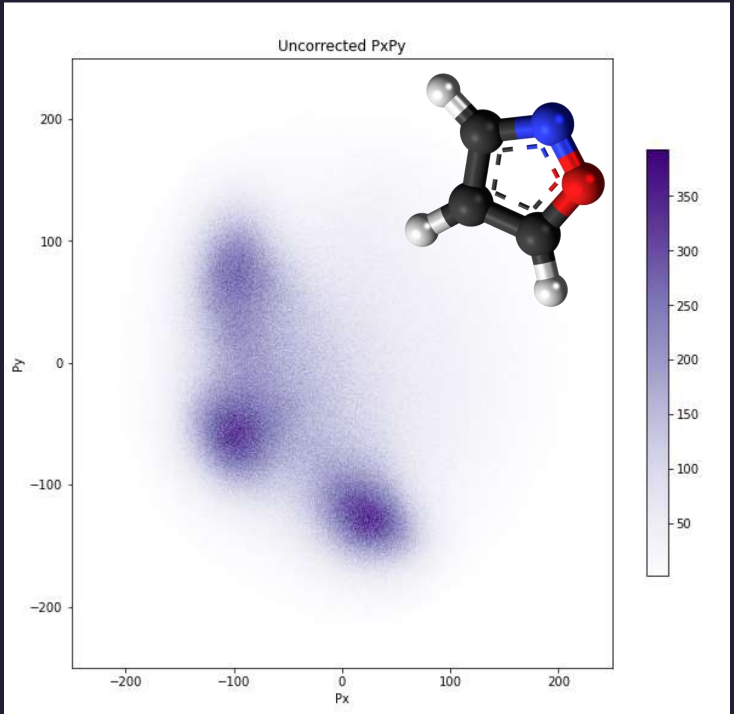


Incomplete Channels are less clean

Sulfur Dioxide

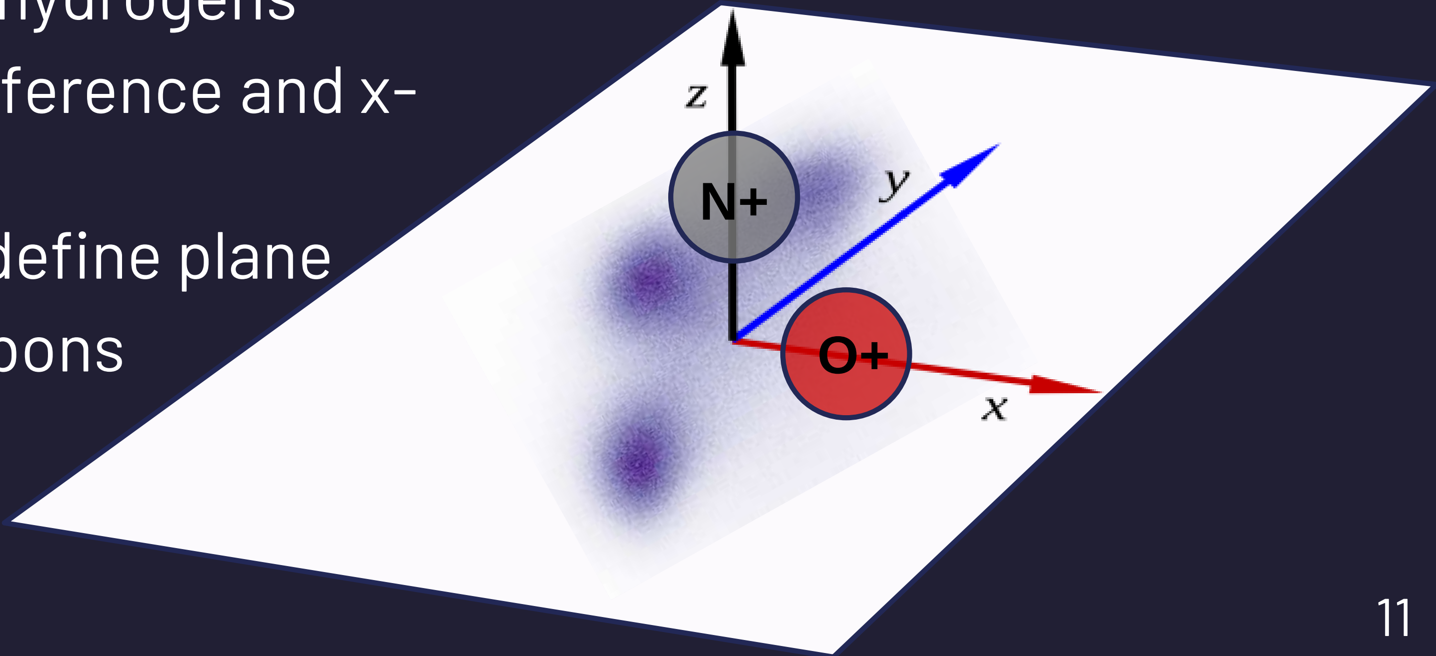


Isoxazole



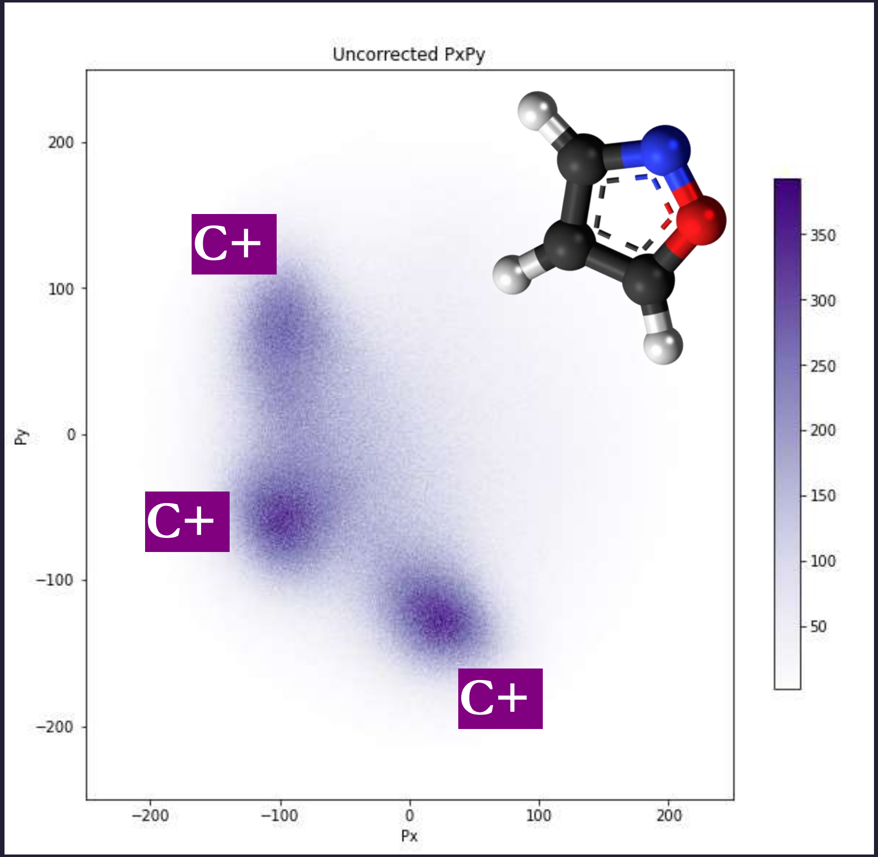
Newton Plot for Isoxazole C_3H_3NO
H+ C+ N+ O+ Channel

- Plotting momenta from the Coulomb Explosion
- Not plotting hydrogens
- Oxygen as reference and x-direction
- Nitrogen to define plane
- Dots are carbons



Motivation

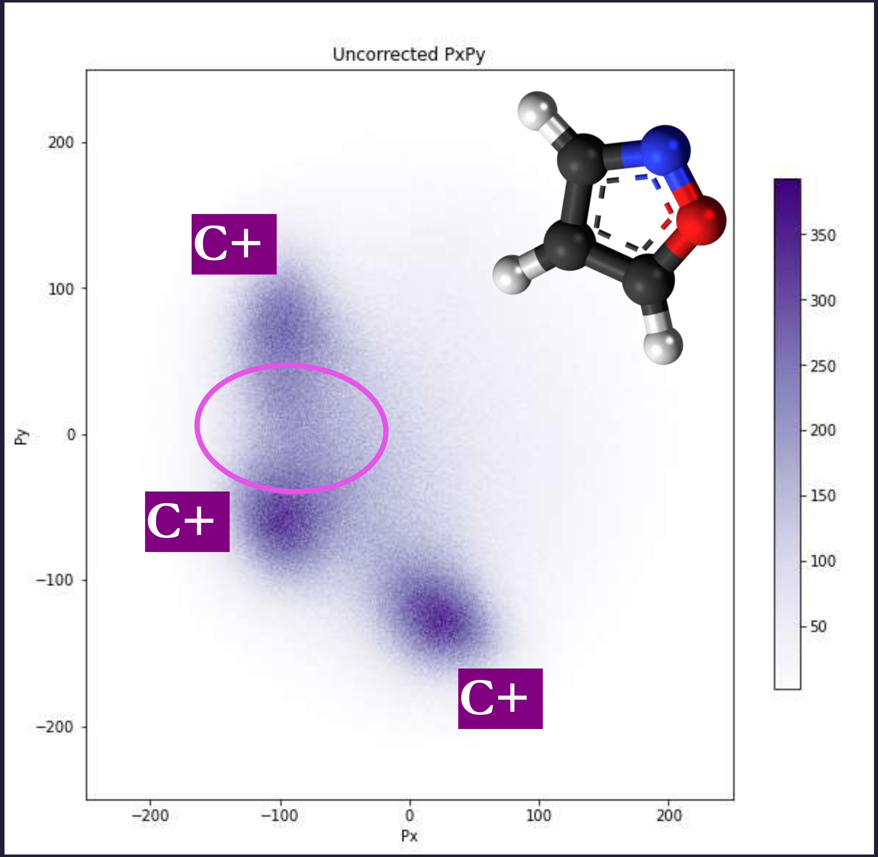
Are the structures real or from *'fuzziness'* ?



Motivation

Are the structures real or from *'fuzziness'* ?

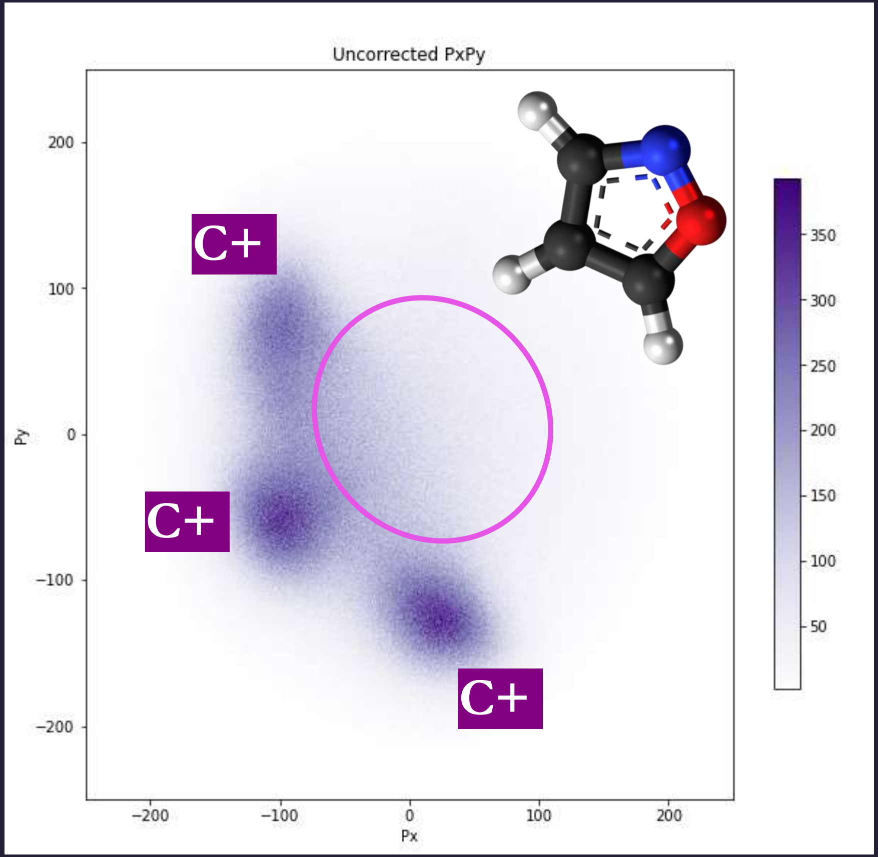
- Probability of carbons being here?



Motivation

Are the structures real or from *'fuzziness'* ?

- Probability of carbons being here?
- Probability of them being way out to the center & top right?

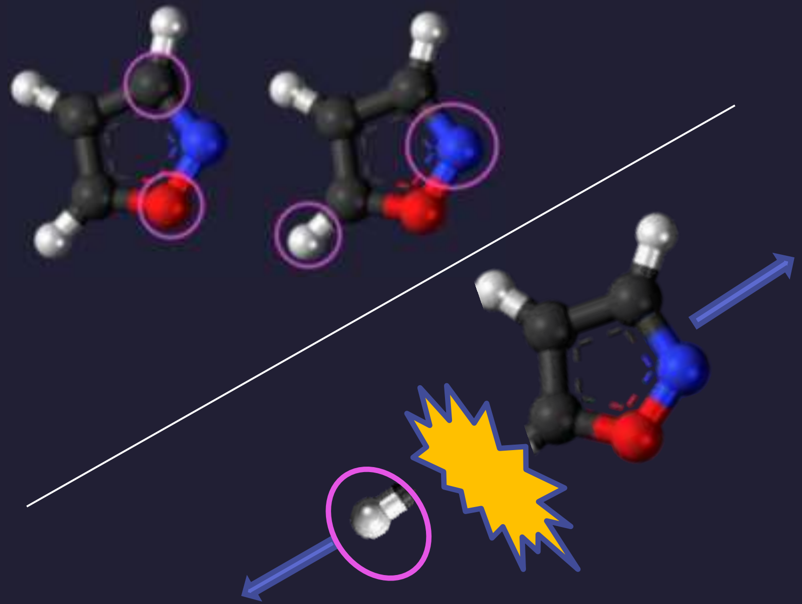


Remove “bad data” by identifying its components.

Modeling the Bad/Random Data Points

Contributions:

- Ionized more than one sample
    - Measured another molecule's ion
        - Ionized the whole molecule or just part
    - More likely because high power is needed for Isoxazole breakup
- Residual Gasses
    - Ex. H2O in the chamber
    - Not perfect vacuum



How many ways could this infiltrate our data?

How many ways can “Bad Data” be permutated among the four measured ions?

- T is a good datapoint (True)
- F is an incorrect datapoint (False)
- TTTF = first three ions are correct but the 4th is wrong.
- Combinations of ways an event could be invalid are:
  - If one is wrong: TTTF, TTFT...
  - If two are wrong: TTFF, TFFT...
  - If three are wrong: TFFF, FTFF...
  - All wrong: FFFF

“Boolean Model”

Modeling by Coincidence Shifting

What does it look like if the 2nd ion came from another source?

i.e. Incorrect Datapoint (False)

Event #	Ion 1	Ion 2	Ion 3	Ion 4
1	X, Y, T	X, Y, T	X, Y, T	X, Y, T
2	X, Y, T	X, Y, T	X, Y, T	X, Y, T
3	X, Y, T	X, Y, T	X, Y, T	X, Y, T
4	X, Y, T	X, Y, T	X, Y, T	X, Y, T
5	X, Y, T	X, Y, T	X, Y, T	X, Y, T

Modeling by Coincidence Shifting

Events are independent of each other, so it is replaced by a random Ion 2 data point

Event #	Ion 1	Ion 2	Ion 3	Ion 4
1	X, Y, T	X, Y, T	X, Y, T	X, Y, T
2	X, Y, T	X, Y, T	X, Y, T	X, Y, T
3	X, Y, T	X, Y, T	X, Y, T	X, Y, T
4	X, Y, T	X, Y, T	X, Y, T	X, Y, T
5	X, Y, T	X, Y, T	X, Y, T	X, Y, T

Modeling by Coincidence Shifting

Prob. replaced by residual gas

Prob. replaced by not fully ionized sample

Prob. replaced by fully ionized sample

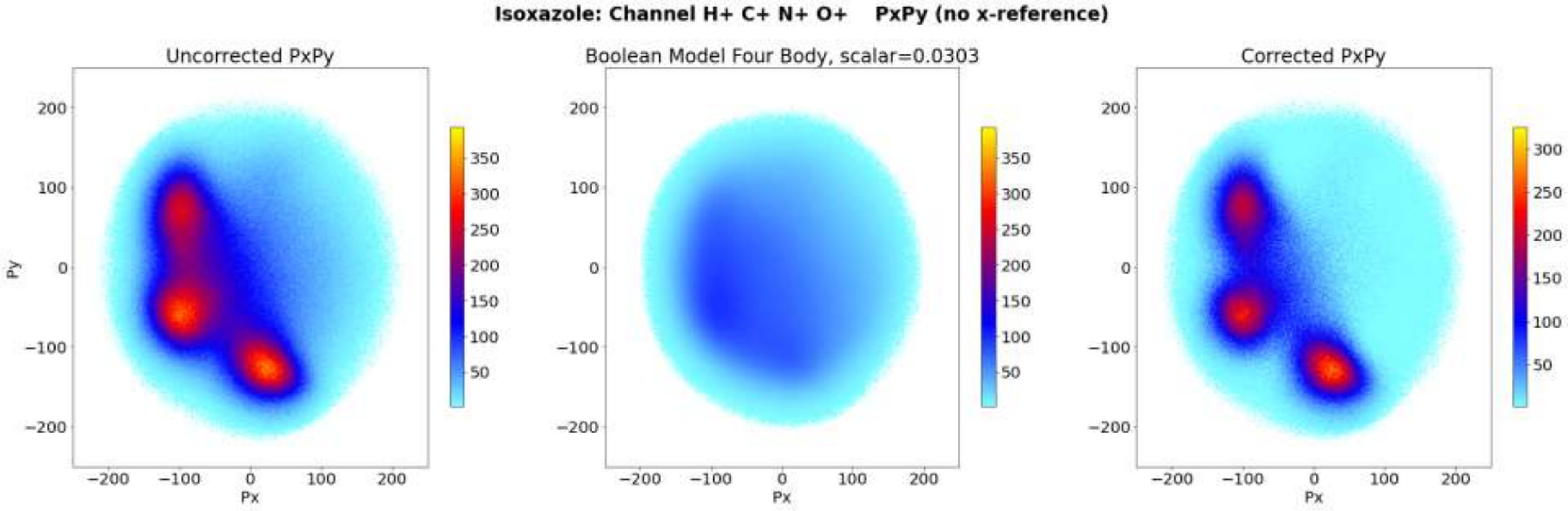
Event #	Ion 1	Ion 2	Ion 3	Ion 4
1	X, Y, T	X, Y, T	X, Y, T	X, Y, T
2	X, Y, T	X, Y, T	X, Y, T	X, Y, T
3	X, Y, T	X, Y, T	X, Y, T	X, Y, T
4	X, Y, T	X, Y, T	X, Y, T	X, Y, T
5	X, Y, T	X, Y, T	X, Y, T	X, Y, T

Include all permutations of invalid events

-> Model is complete!

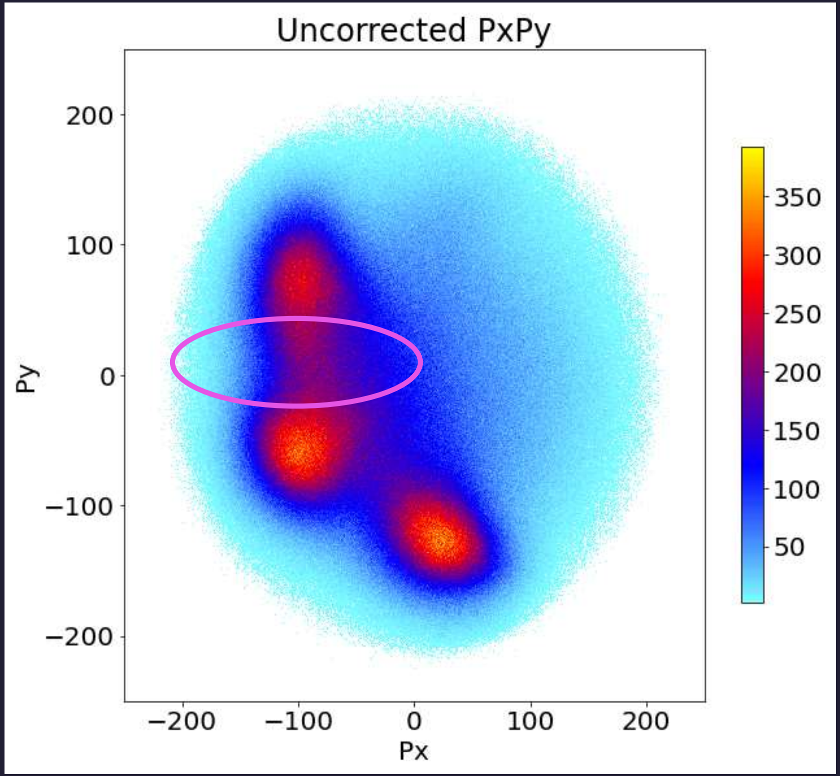
Results

Uncorrected Data | Model | Corrected

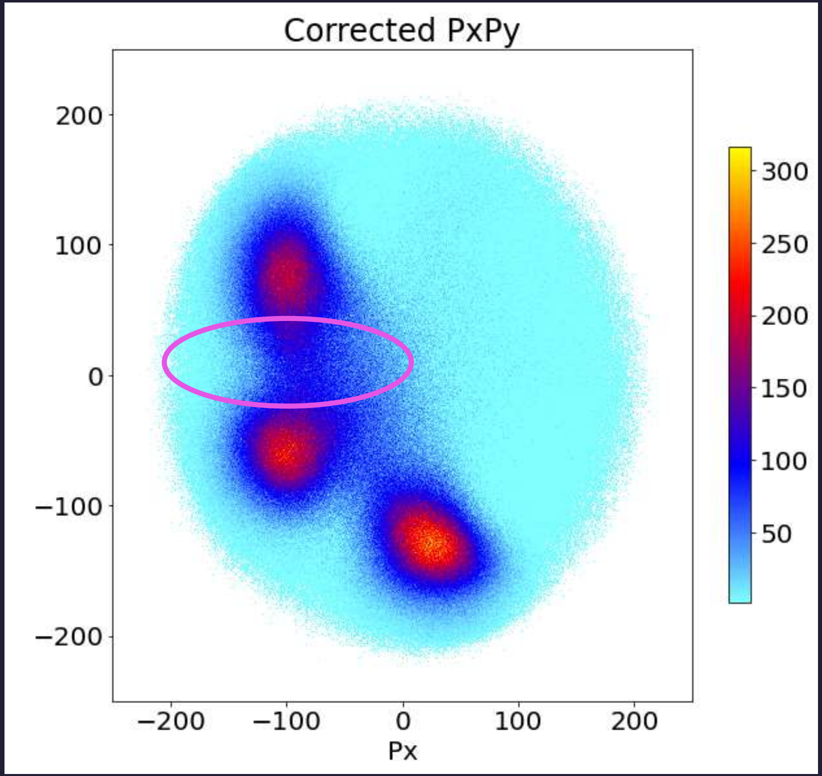


Results

Uncorrected Data

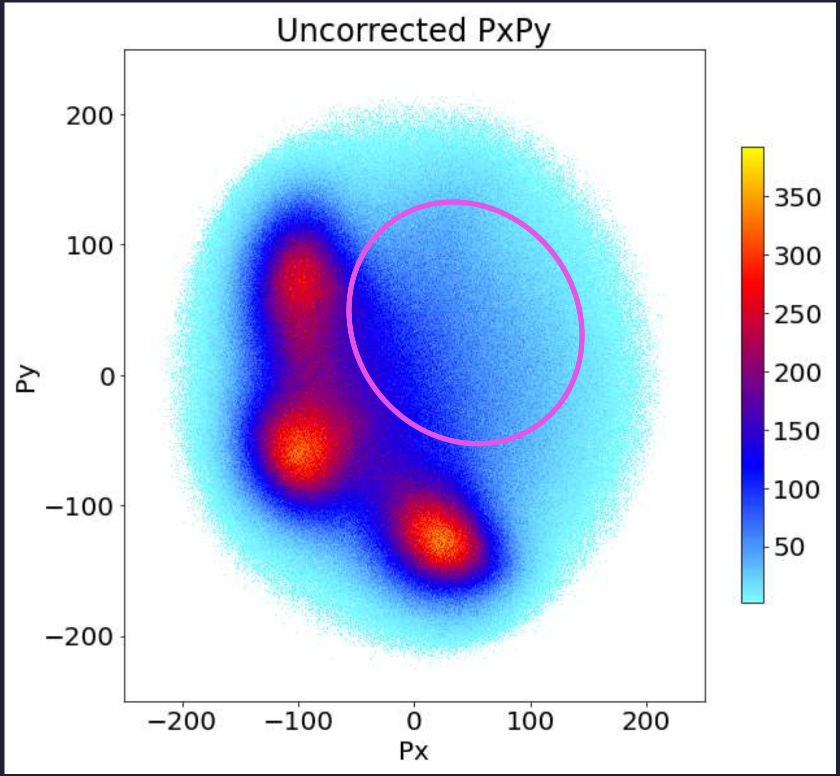


Corrected

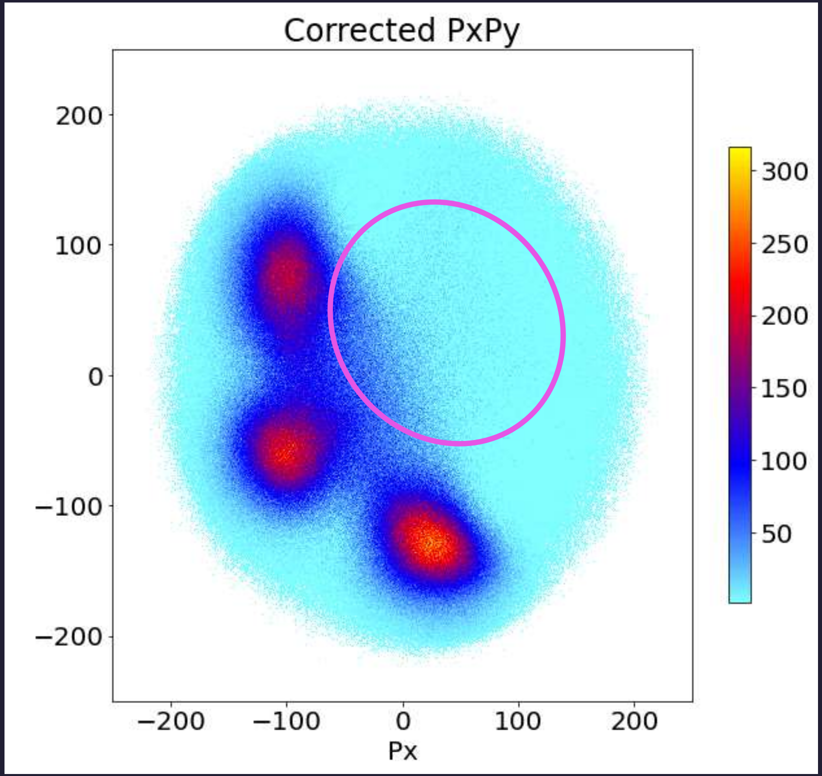


Results

Uncorrected Data

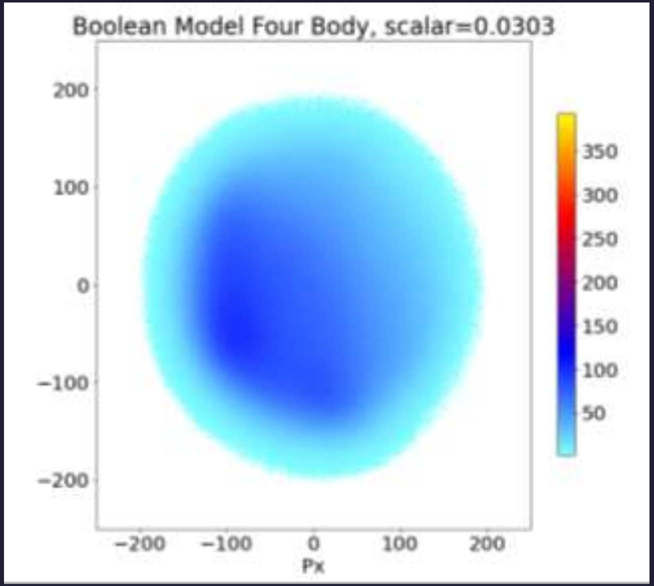


Corrected



Results

- Effective method for sharpening incomplete channel images & plots
- Helps determine real structures
- Can account for secondary molecules and some residual gasses
- Background terms are non-isotropic



Future outlook

- Shifting model is biased towards fully ionized contributions
  - Good data that is shifted == bad data from second fully ionized sample
    - Data: 70% good data
    - Model: >70% fully ionized contributions
    - Means it's better at this, but worse at others
- More likely that one is wrong than all four
- Applying this to more incomplete channel experiments
  - Ex. May work best with messy data with ions that include lots of residual gas points

Acknowledgements

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&

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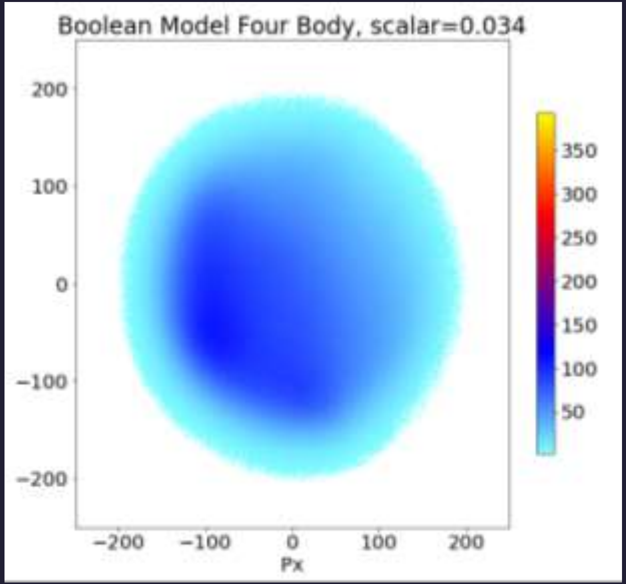
Questions?

- Can you correct things other than newton plots?
- What do you mean “the model is biased towards secondary contributions”?
- Could you go back to that one slide?



Bias Towards Total Ionization

Original Data

Prob. residual gas

Prob. not fully ionized sample

Prob. separate fully ionized sample

Prob. original fully ionized sample

Model

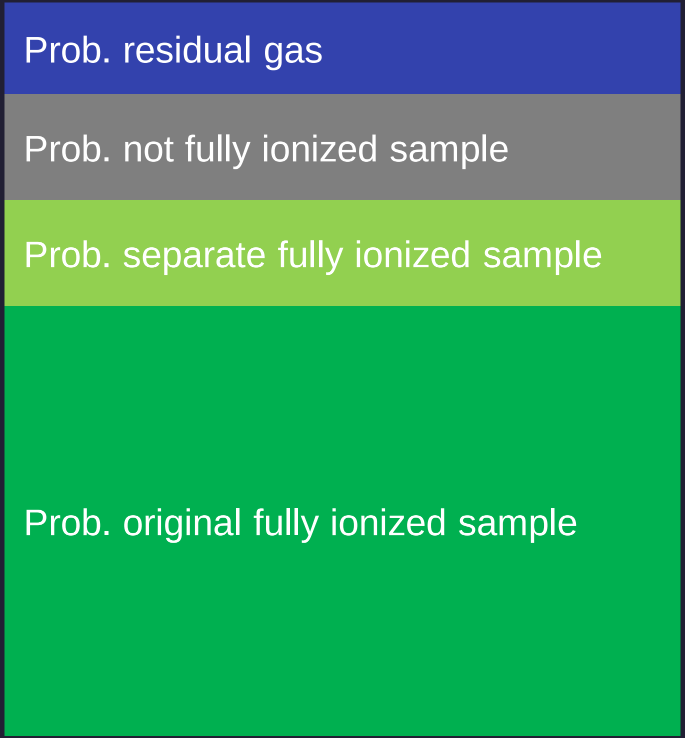
Prob. replaced by residual gas

Prob. replaced by not fully ionized sample

Prob. its from a separate fully ionized sample

Bias Towards Total Ionization

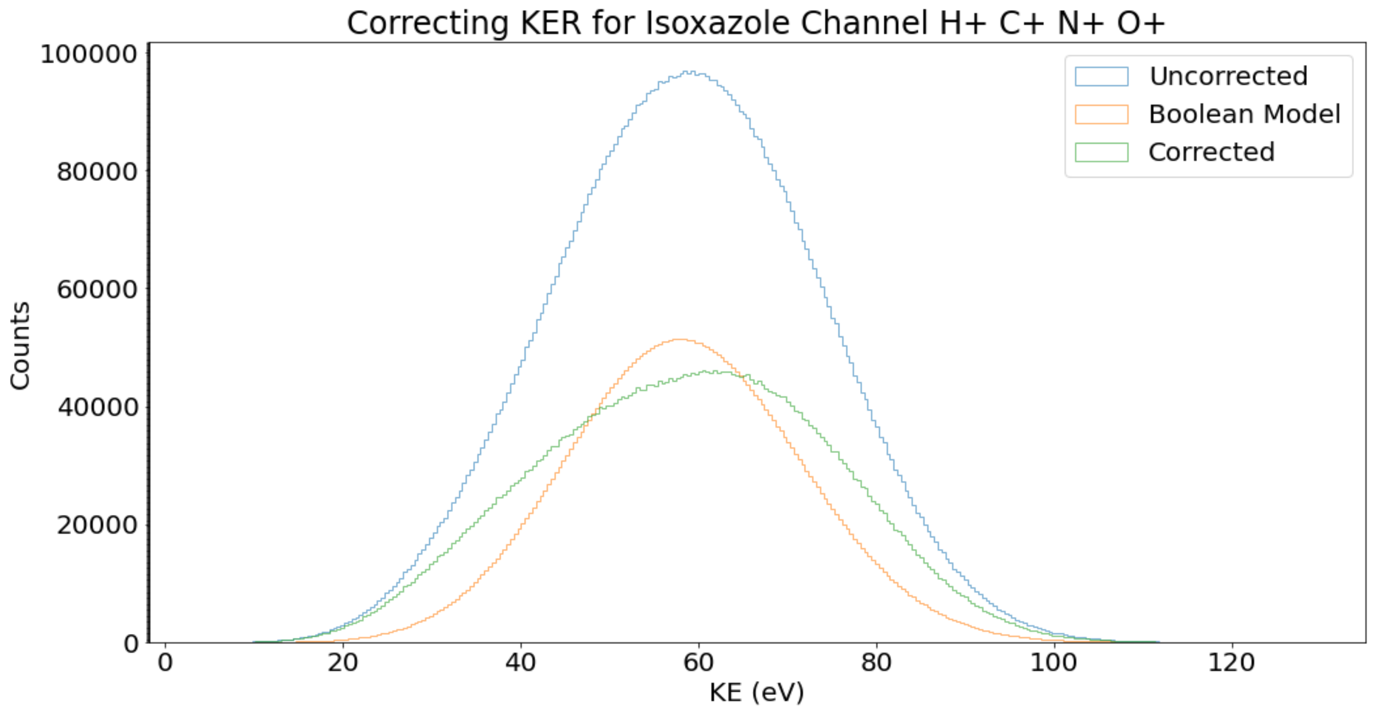
Original Data



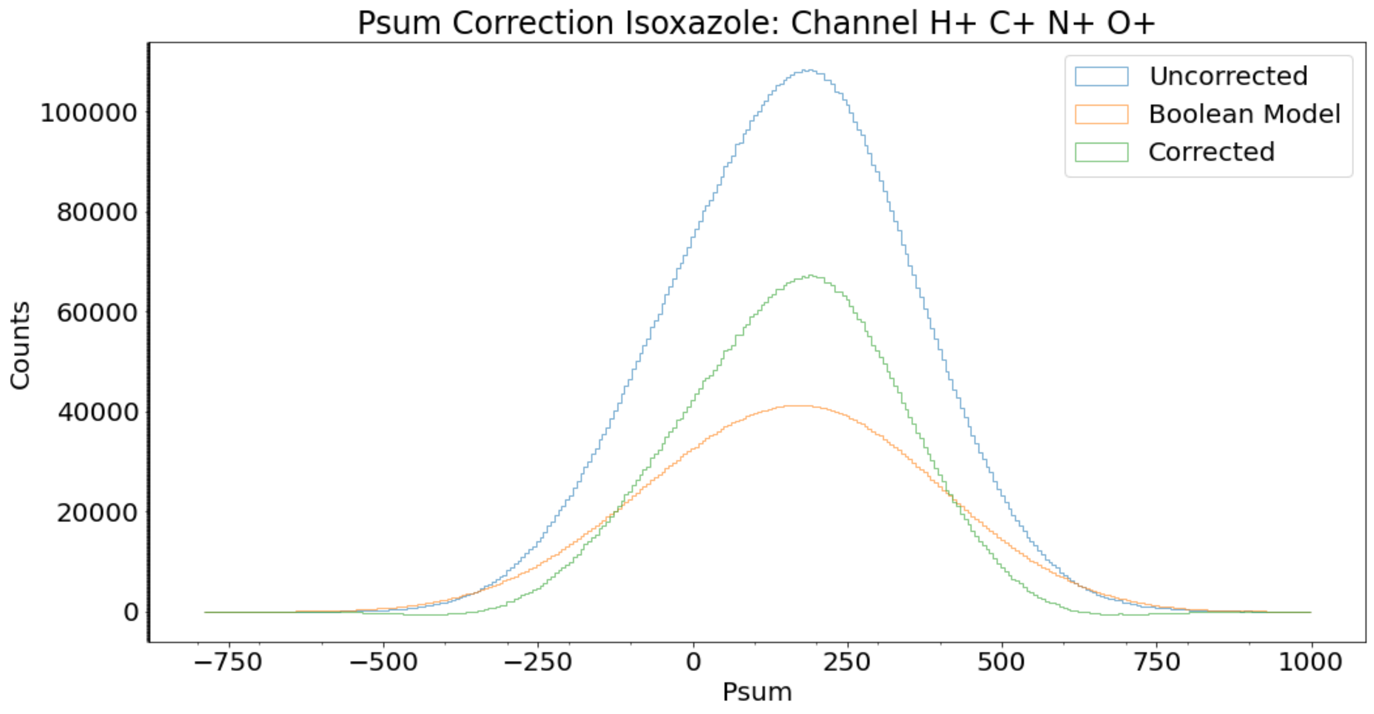
Scaled Down Model

- Still subtracts randoms, but it is more effective at removing separate fully ionized samples.
- Fundamental issue if you expect zero secondary fully ionized samples

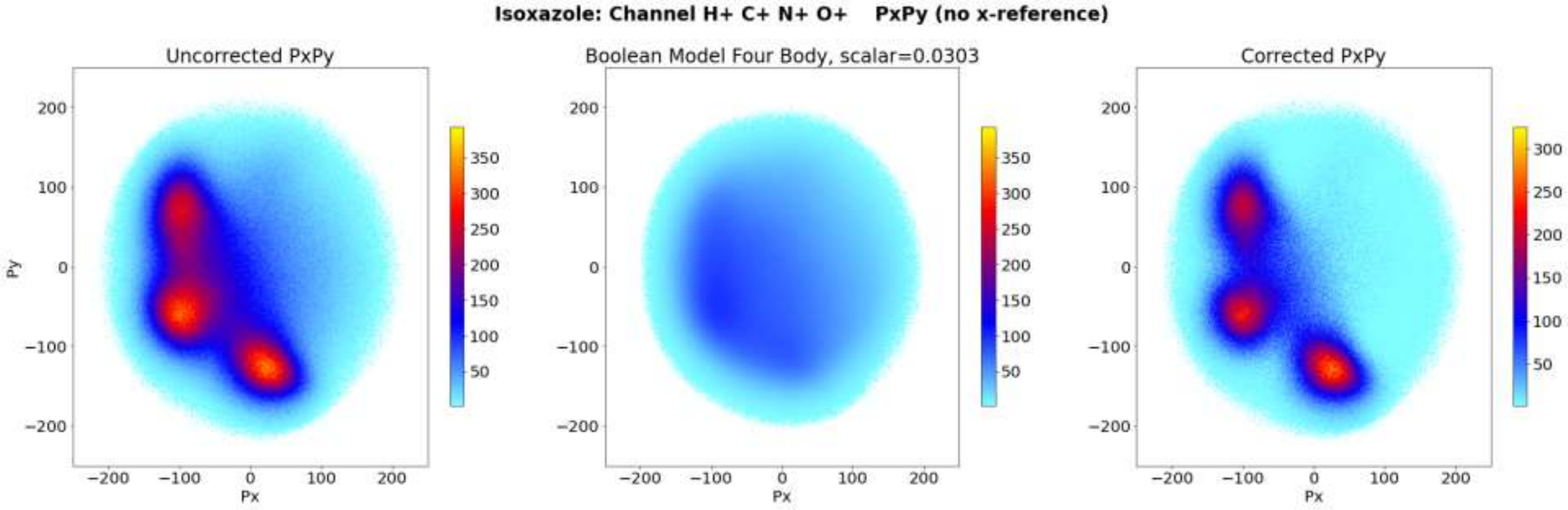
Corrected KER



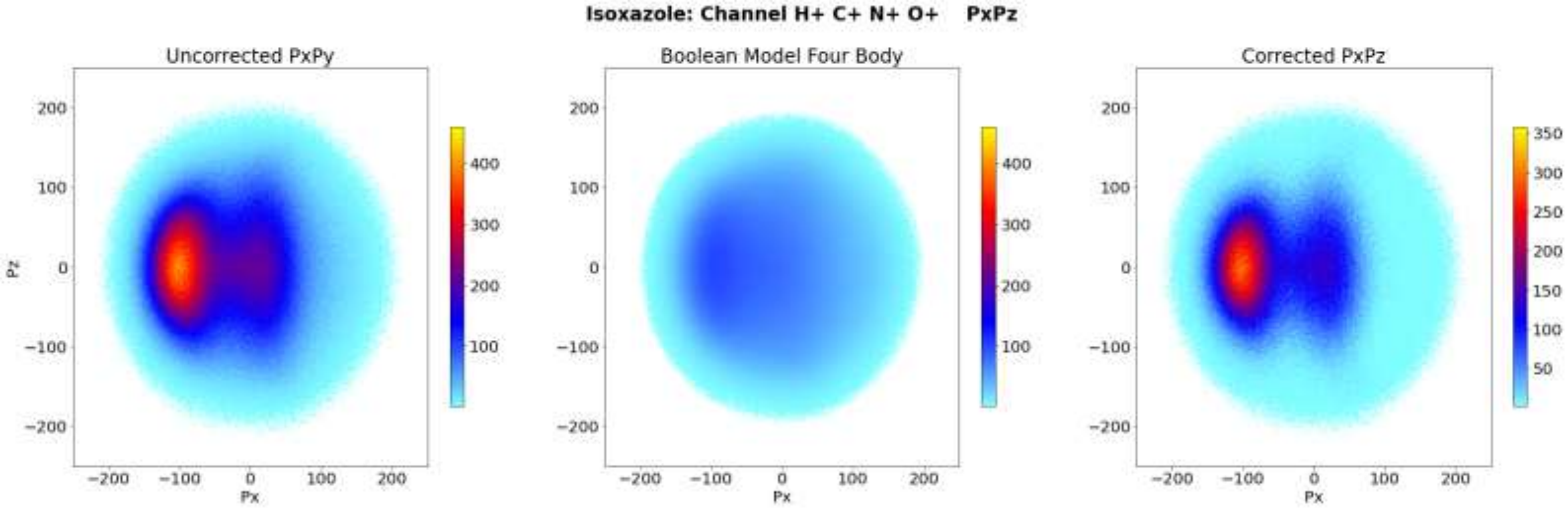
Corrected Psum



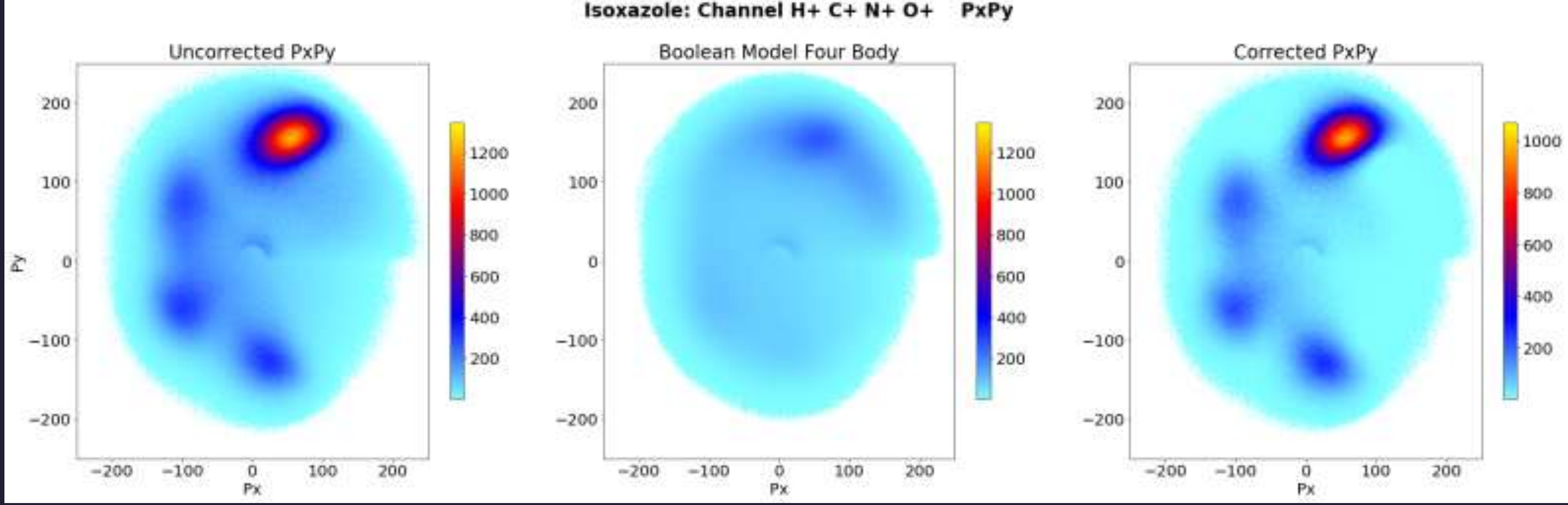
PxPy
(no x-reference)



PxPz

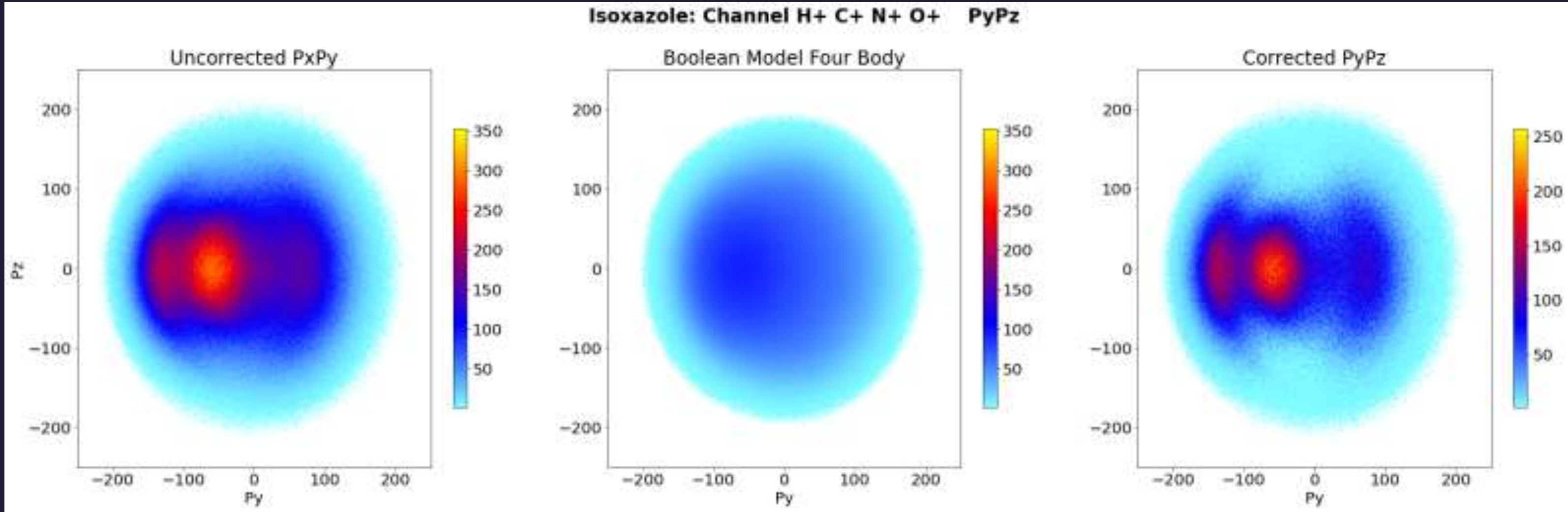


PxPy



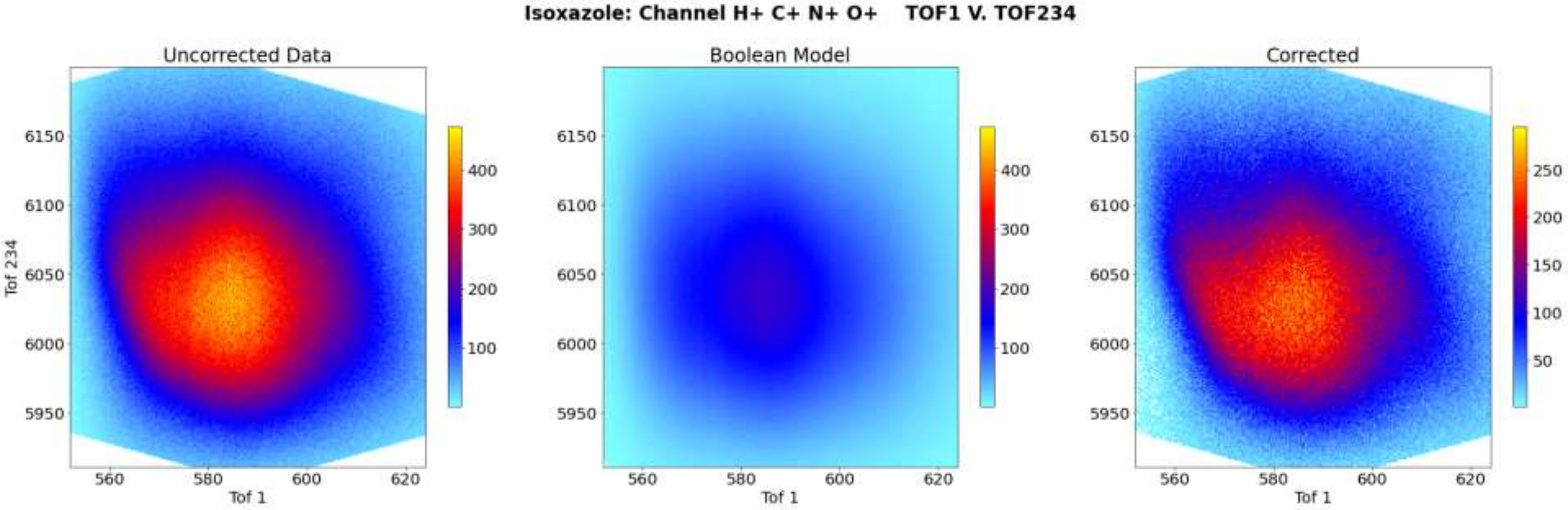
Isoxazole: Channel H+ C+ N+ O+ PxPy
Uncorrected PxPy
Boolean Model Four Body
Corrected PxPy
Px
Py

PyPz

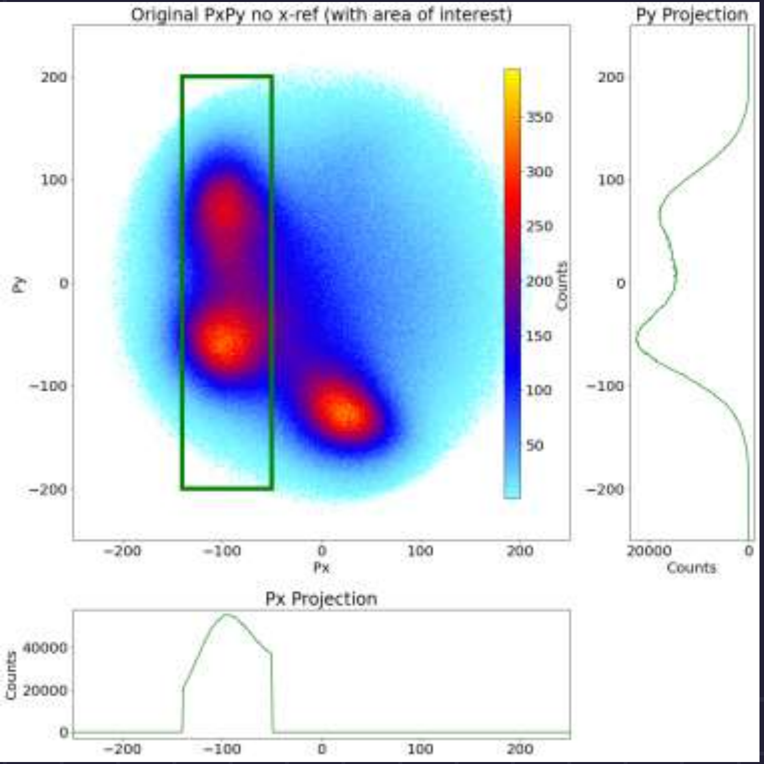


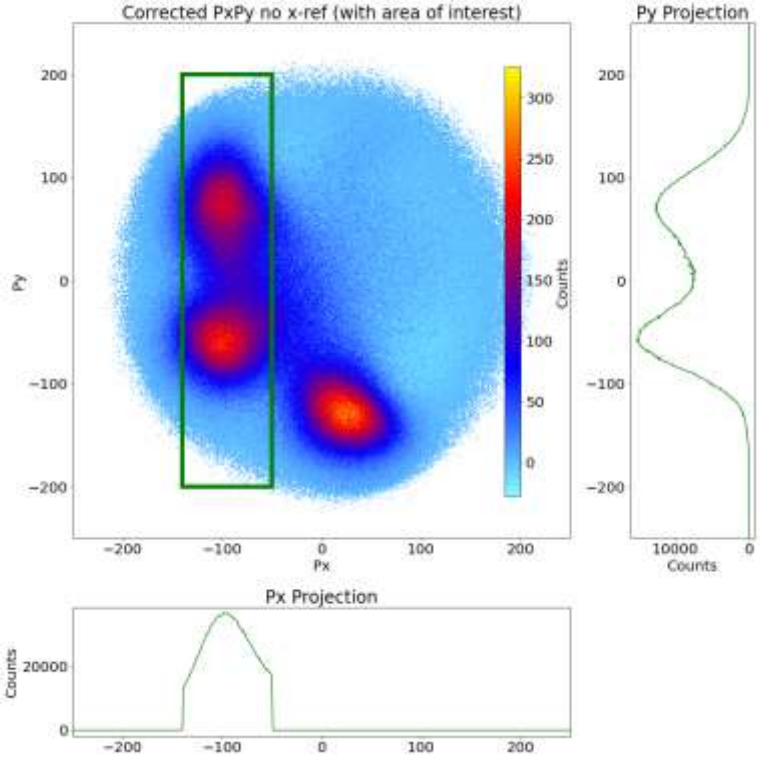
Isoxazole: Channel H+ C+ N+ O+ PyPz
Uncorrected PxPy
Boolean Model Four Body
Corrected PyPz
Py
Pz

Corrected Gated 4-Fold Coincidence

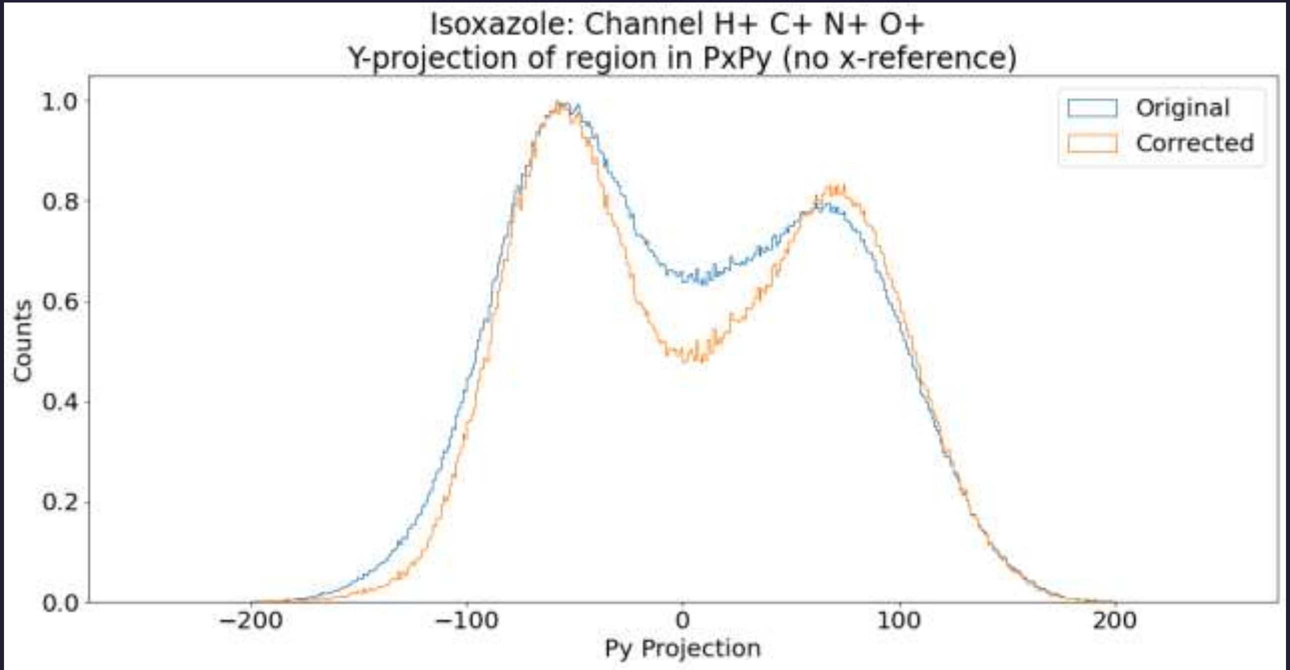


Projections in region where we want the peak to trough ratio



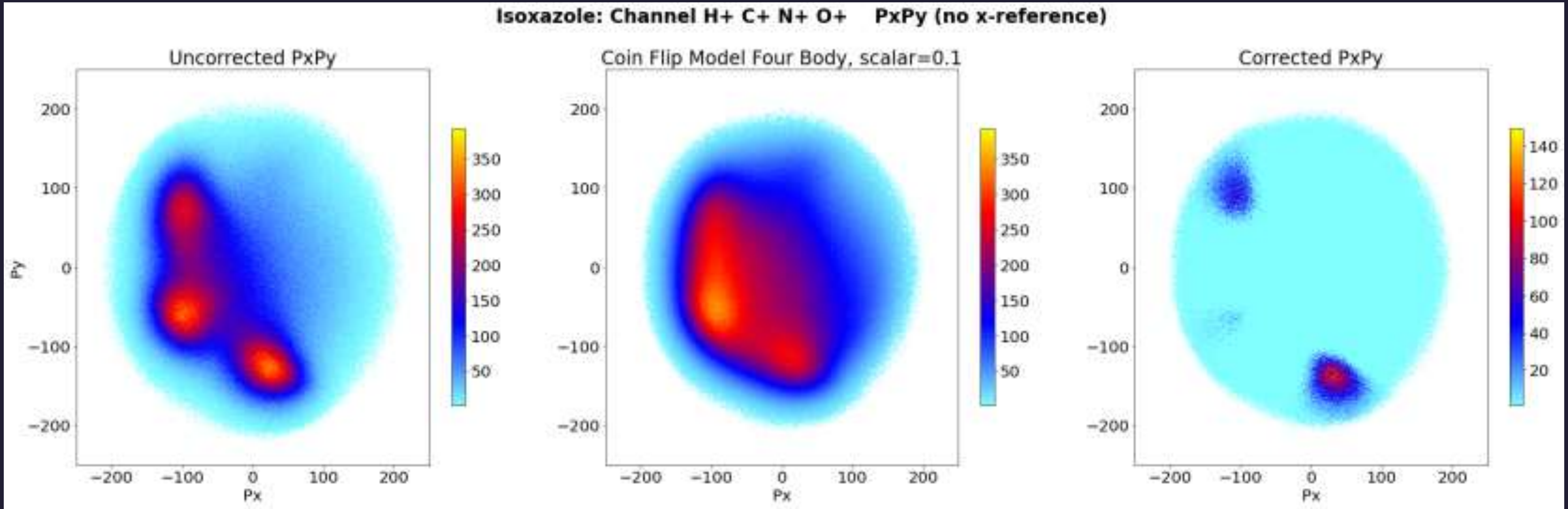


Normalized Py projection in region where we want the peak to trough ratio



Over-Subtraction

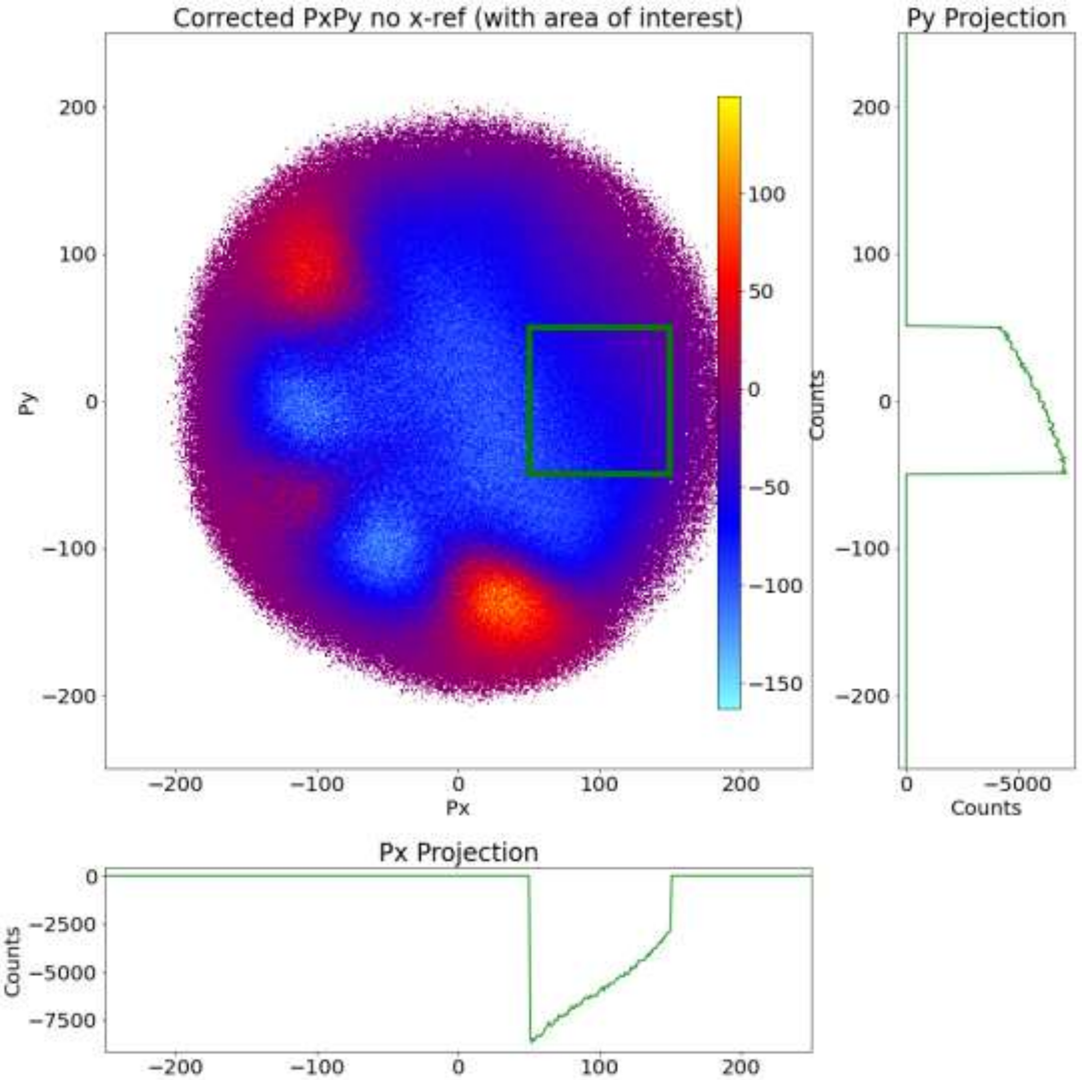
Uncorrected Data | Model | "Corrected"



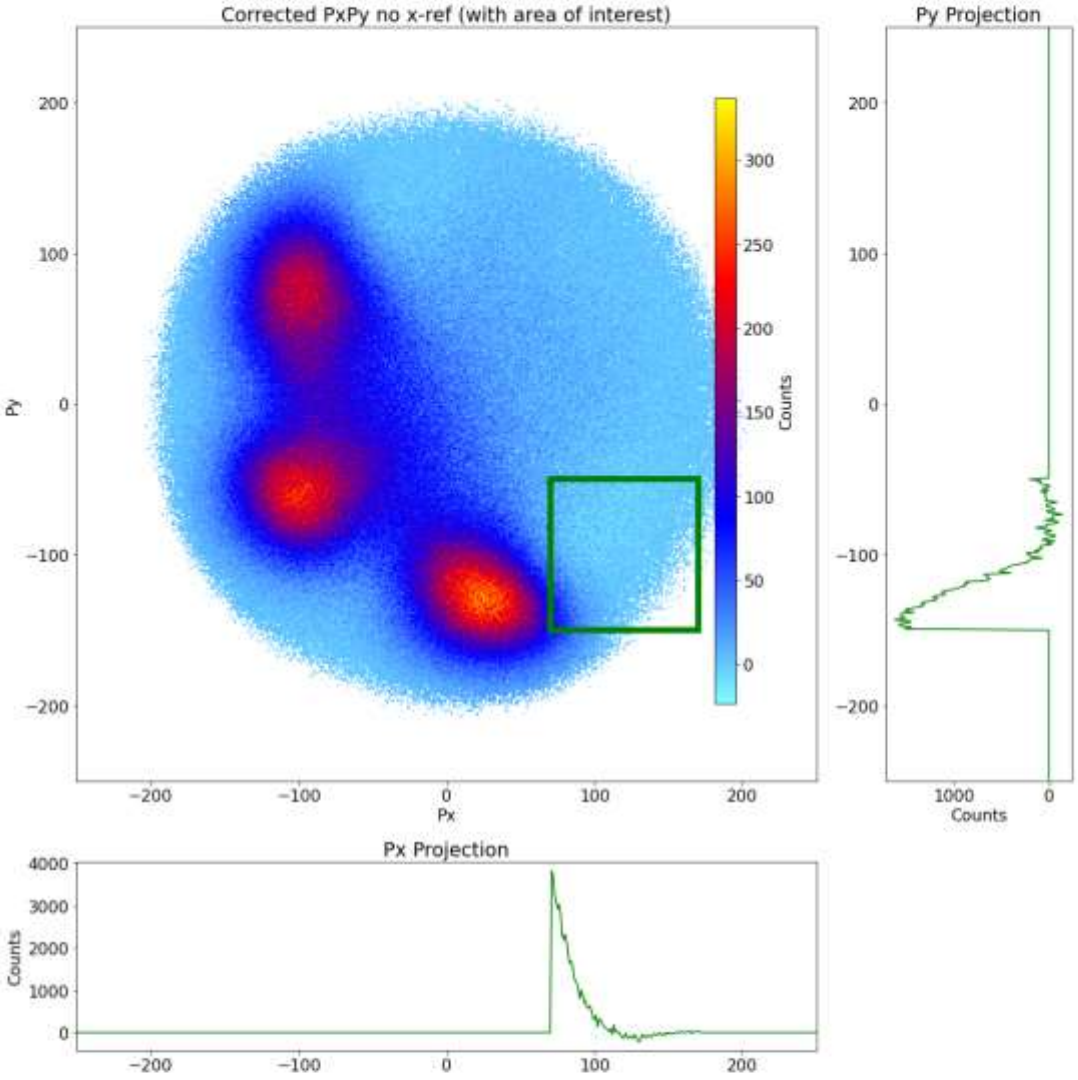
Subtracts too much and leave negative counts, which make no physical sense (points < 0 not plotted)

Plotting the negative counts, we see that there are regions with lots of negative points.

Scale down to reduce negative points

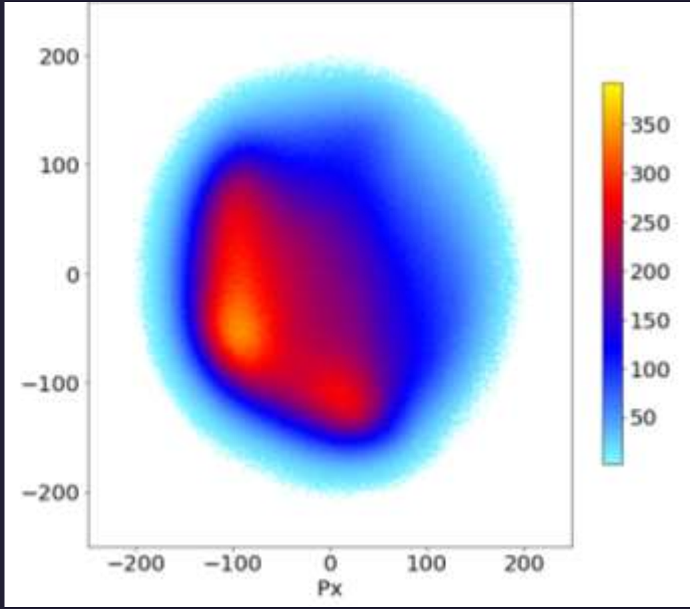


It’s okay if one tiny square is negative, so long as the average is ~0

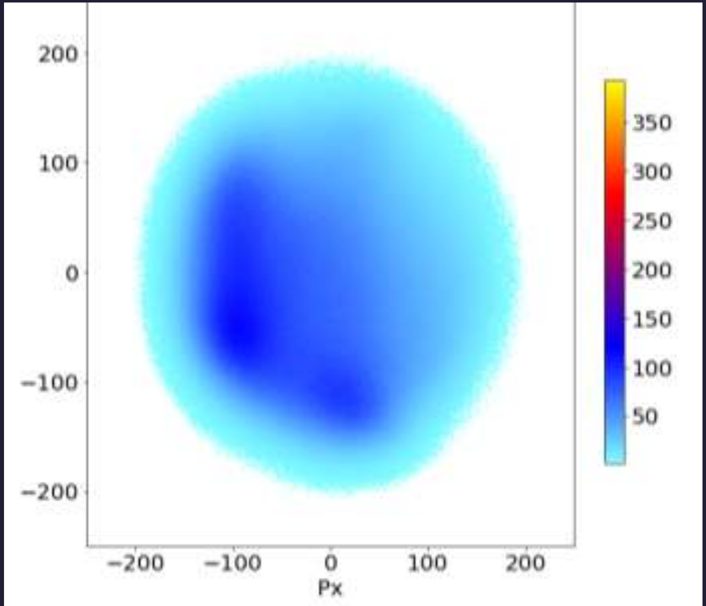


Scalar Change for Reduced Negatives

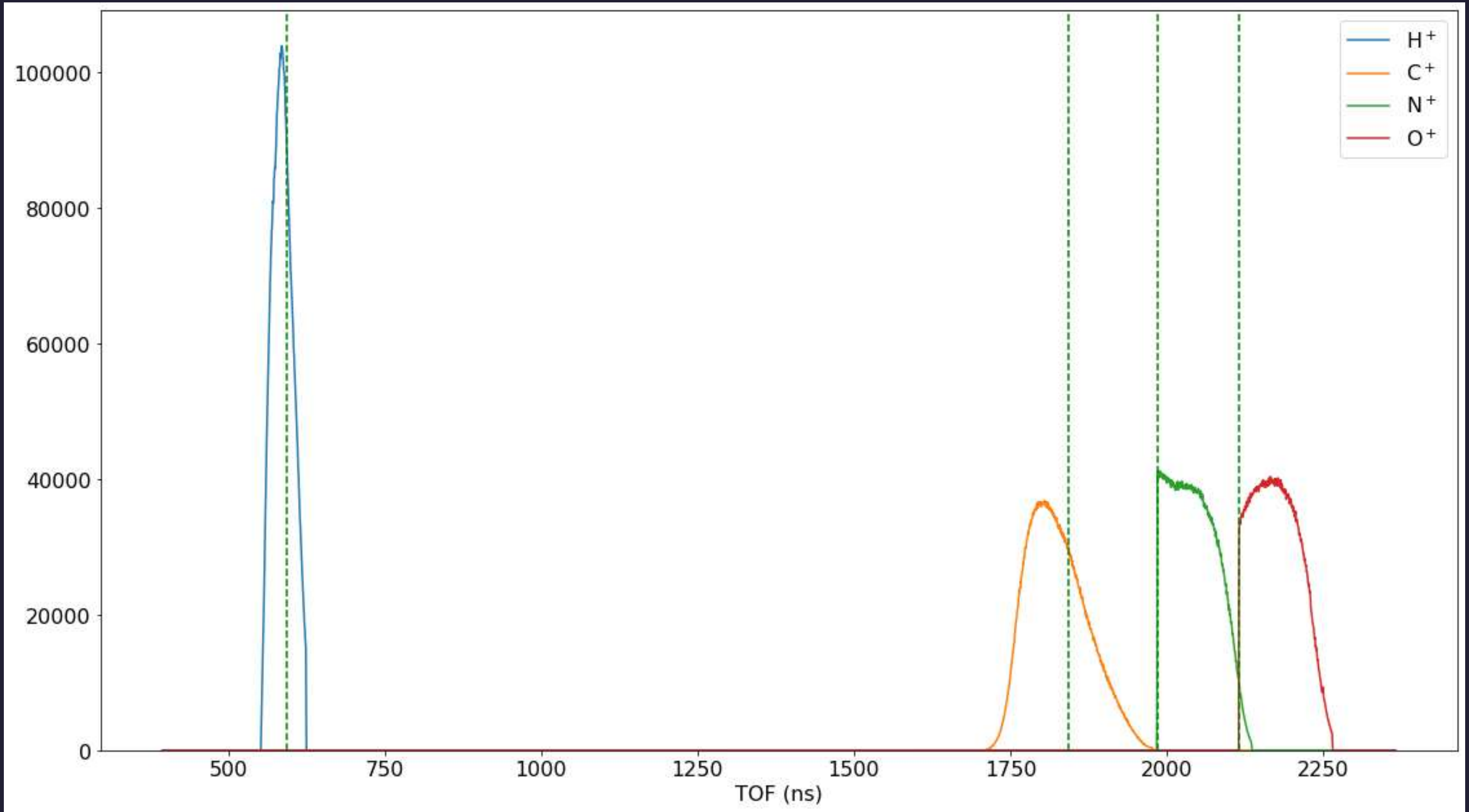
Scalar = 0.1, too much subtraction



Scalar = 0.0303, just enough

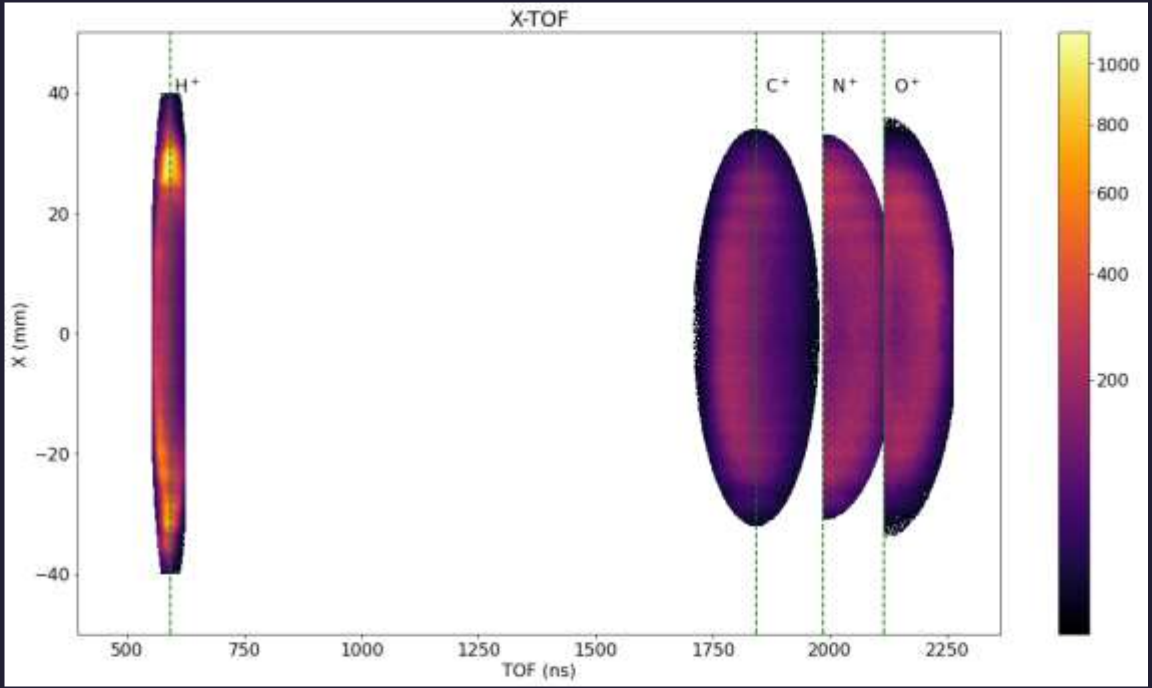


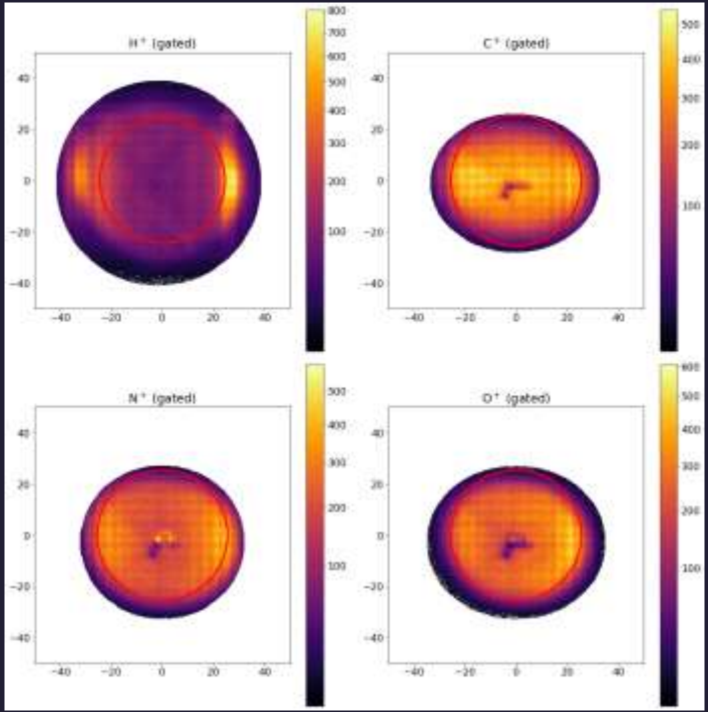
TOF Gating



H+
C+
N+
O+
100000
80000
60000
40000
20000
0
500
750
1000
1250
1500
1750
2000
2250
TOF (ns)

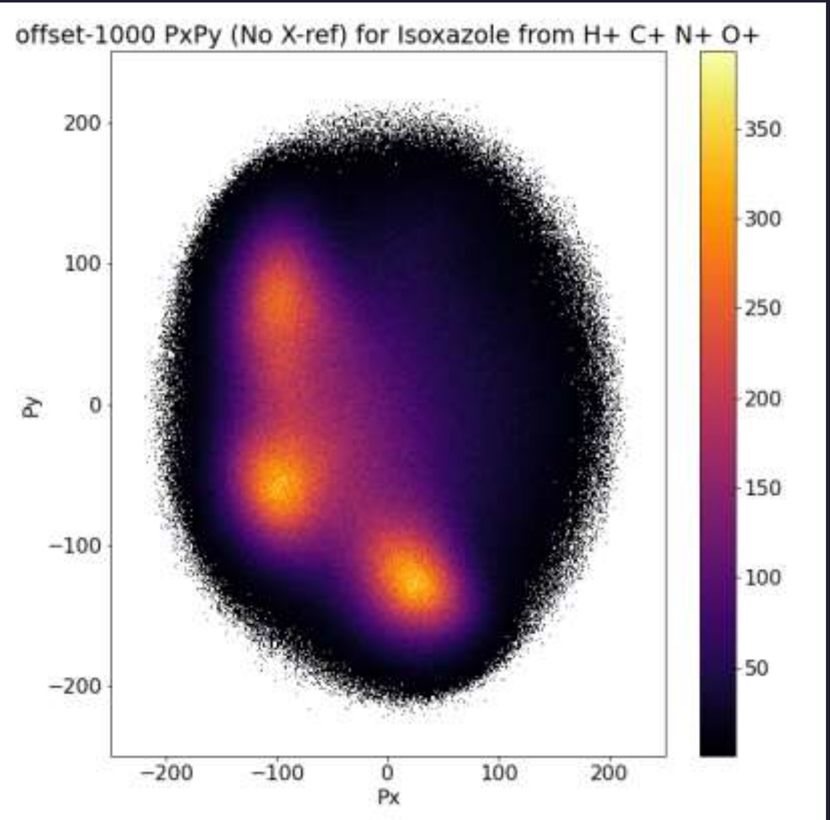
X and Y – TOF Gating





Hydrogen isn’t plotted, so no adjustment is seen

Hydrogen Offset



Original