How can we adjust the probabilities of a pair of dice to make the odds of rolling the different sums as close to equal as possible? The average variance of the probabilities of the 11 possible sums is the sum of the squared differences between each probability and $1/11$, divided by 11.

For fair dice the probability distribution of the various sums is:

The average variance among the 11 sums for fair dice is therefore:

$$
\left(2 \sum_{i=1}^{5} \left(\frac{i}{36} - \frac{1}{11}\right)^2 + \left(\frac{1}{6} - \frac{1}{11}\right)^2\right)/11 \approx 0.001976839098
$$

I wrote a Python program which tried swapping a small and shrinking probability increment between two random faces of both dice, retaining any swap that reduced the average variance. It settled fairly quickly on this probability arrangement for faces 1 through 6:

$p$(faces 1 and 6) $\approx 0.243882629$

$p$(faces 2 and 5) $\approx 0.137479186$

$p$(faces 3 and 4) $\approx 0.118638185$

This gives a probability distribution of the various sums as:

The average variance for this arrangement is approximately $0.001217583329$, which is about 61.6% of the value for fair dice, a noticeable improvement. It makes sense to see the probabilities of 1 and 6 larger than the other values, since you want to increase the probabilities of the extreme sums at the expense of those in the middle.
When I started working on this problem, I assumed that the two dice could be different, and I didn’t see Zach’s tweet that they must be the same until I had already worked out a solution. So, here are the results where the two dice are allowed to have different probabilities from each other:

Die #1
\[ p(\text{faces 1 and 6}) = \frac{1}{2} \]
\[ p(\text{faces 2, 3, 4 and 5}) = 0 \text{ (or arbitrarily small if you consider zero cheating)} \]

Die #2
\[ p(\text{faces 1 and 6}) = \frac{1}{8} \]
\[ p(\text{faces 2, 3, 4 and 5}) = \frac{3}{16} \]

This gives a probability distribution of the various sums as:

The average variance for this arrangement is approximately \textbf{0.000258264}, which is about 13\% of the value for fair dice, a significant improvement.

A nice feature of this version of the problem is that the probability for each face turned out to be a very simple exact fraction. Another nice feature of this version is that it would be possible to construct physical dice (where both dice are rectangular prisms using the traditional arrangement of faces) that have probabilities arbitrarily close to these values!

Die #1 would have large squares for faces 1 and 6, and extremely thin rectangles for 2, 3, 4, and 5. Die #2 would have medium squares for faces 1 and 6, and slightly larger rectangles for 2, 3, 4, and 5.

It would require some experimentation to find the ideal ratio of the short edges to the long edges for Die #2, but the symmetry of the shape would guarantee that the probabilities for faces 1 and 6, and for faces 2, 3, 4, and 5 would be equal. Here is a layout for such a pair of dice:

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