I see 2 questions here:

1: How many pokeballs are in Coit Tower?

2: After removing them all from the tower, how many will Team Rocket actually leave the premesis with?

1:

If you look at the anime, pokeballs, when shrunk down to their smallest size, are about 1 inch diameter.

The volume of a pokeball in inches is 4/3\*pi\*0.5^3, which is about 0.52 cubic inches. If you pack spheres perfectly (and, if Coit Tower is a super-expensive pokeball storage unit, they probably would be packed close to perfectly in there to maximize space), they take up about 74% of the volume of their container.

According to wikipedia, the tower is 210 feet tall. And if you look at it with satellite view via Google Maps, the tower has about a 50’ diameter. However, you said the “tower” was entirely packed with pokeballs. That can’t include the crown-like structure at the top, as it is too porous to hold pokeballs, or the base, since that’s obviously where all of the virtual reality and pokeball-inspired space distortion technology is housed to trick tourists into thinking they’re going up into the tower.

So I interpret the “tower” to be the shaft protruding from the base of the building. From the base of the “crown structure” atop the tower to its base is about 124’(1488”) according to my measurements from a 3d modeling program. Assuming the walls of the tower aren’t paper-thin, the diameter of the hollow inside of the tower is maybe around 33’(396”).

A cylinder’s volume is pi \*r^2 \* h, or pi \*198^2 \*1488, or 1.8 \*10^8 cubic inches. 74% of this, or 1.4\*10^8, is pokeball. Divide this by 0.52, and the tower of the building itself is filled with 260,802,386 pokeballs.

2:

Now, let’s assume Team Rocket has extracted these pokeballs and loaded them into a bunch of trucks. How many did Team Rocket get away with?

None.

You see, the Snorlax is clearly there to do more than seal the entrance when the tower is closed to visitors. Anyone can Google how to get a snorlax to wake up and move. So what use are they in a security system? A canary in the mineshaft. That Snorlax was probably sitting on a pressure-sensitive switch. As soon as Team Rocket got the Snorlax to move, a silent alarm was triggered. By the time Team Rocket loaded up all of the Pokeballs, they were surrounded by police and by 10-year-olds weilding gods as pets.

Plus, the pokeballs are useless to them anyway; since pokemon are basically weapons, California’s strict laws clearly state that all pokeballs must be inert until activated by a licensed vendor who has verified the legitimacy of the purchaser’s trainer’s license.