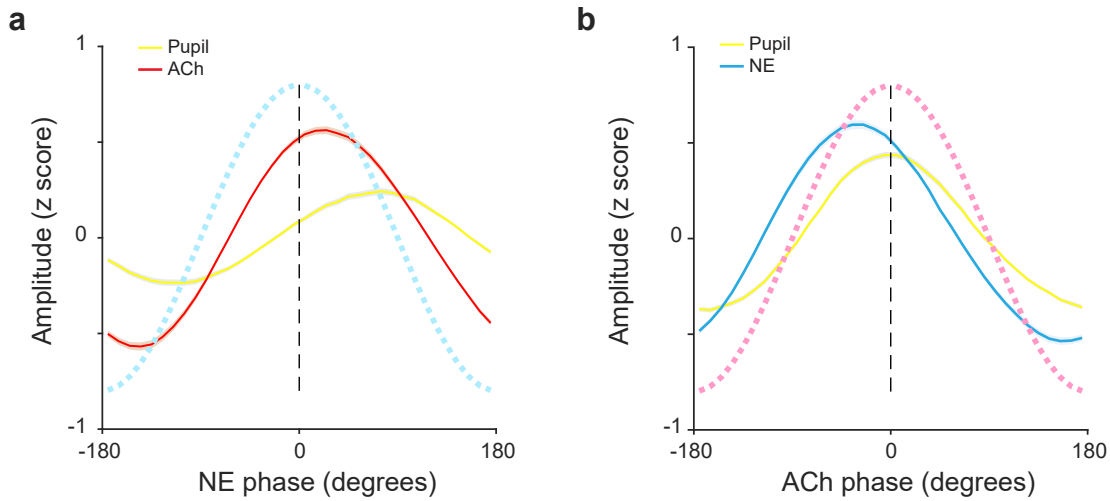
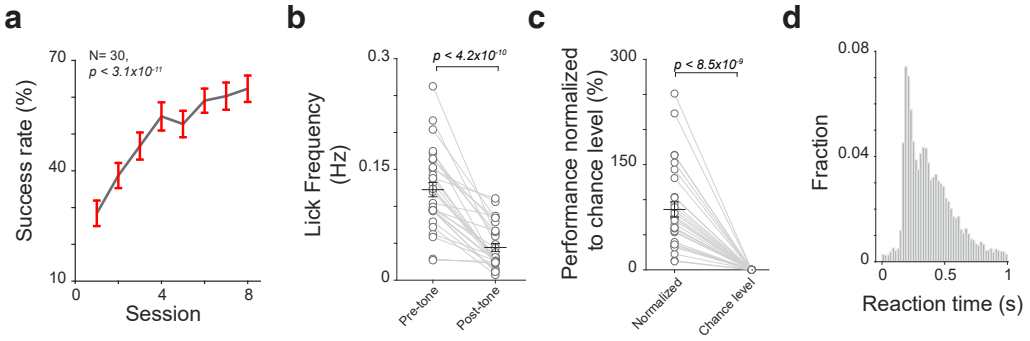
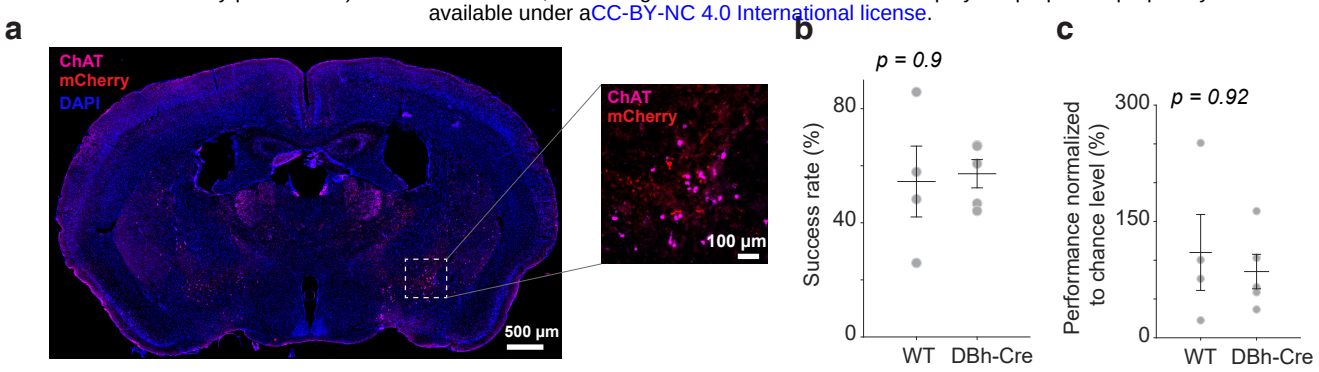


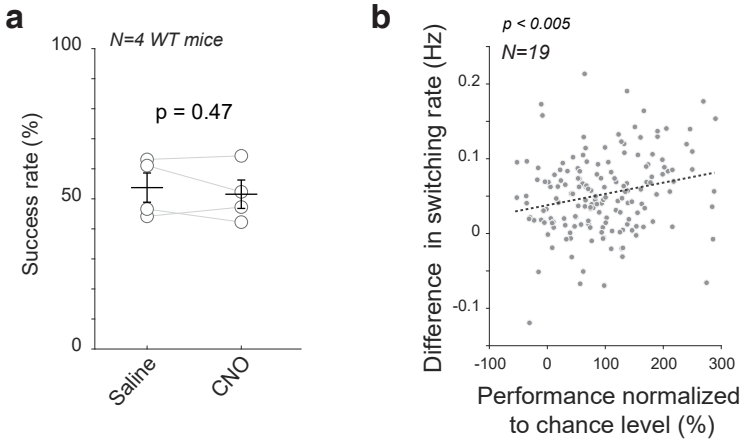
Supplemental figure 1. The phase relationship among prefrontal NE, ACh and pupil fluctuations. a) *ACh and pupil phase referenced to NE phase.* ***b)*** *NE and pupil phase referenced to ACh phase.*



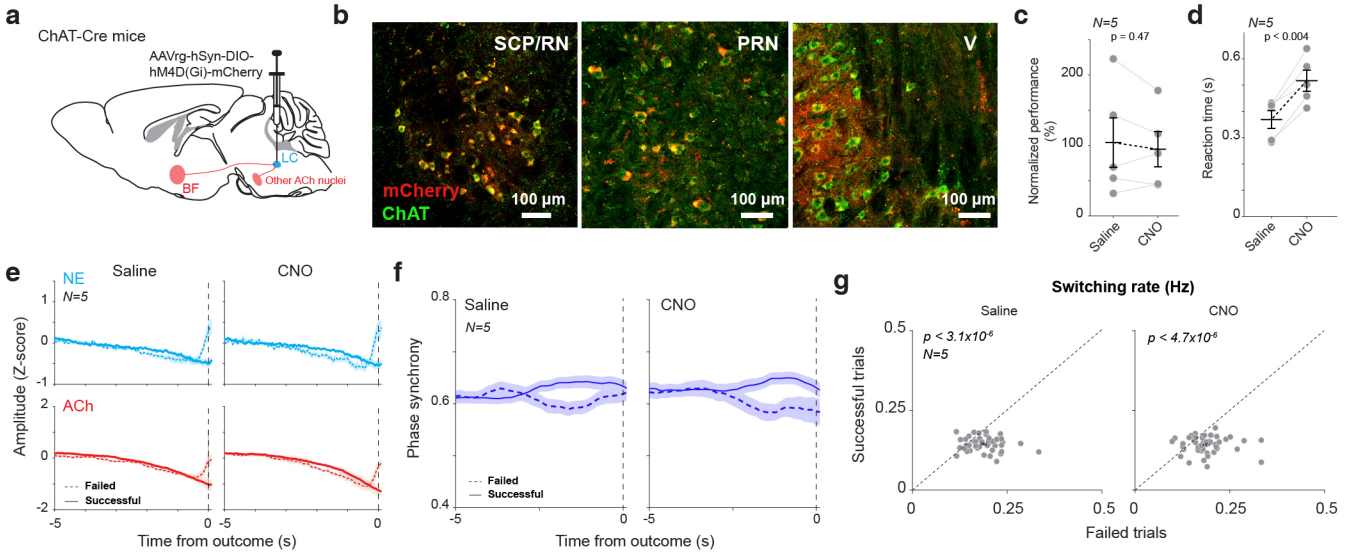
Supplemental figure 2. The inhibitory control task performance. a) *The increase in success rate during the initial training sessions indicated that the suppression of impulsive licking was a learned behavior.* ***b)*** *Mean licking frequency within a 2-second window prior to vs. after the onset of inhibition tone.* ***c)*** *Inhibitory control task performance normalized to the chance level performance.* ***d)*** *Histogram of reaction time in successful trials.*



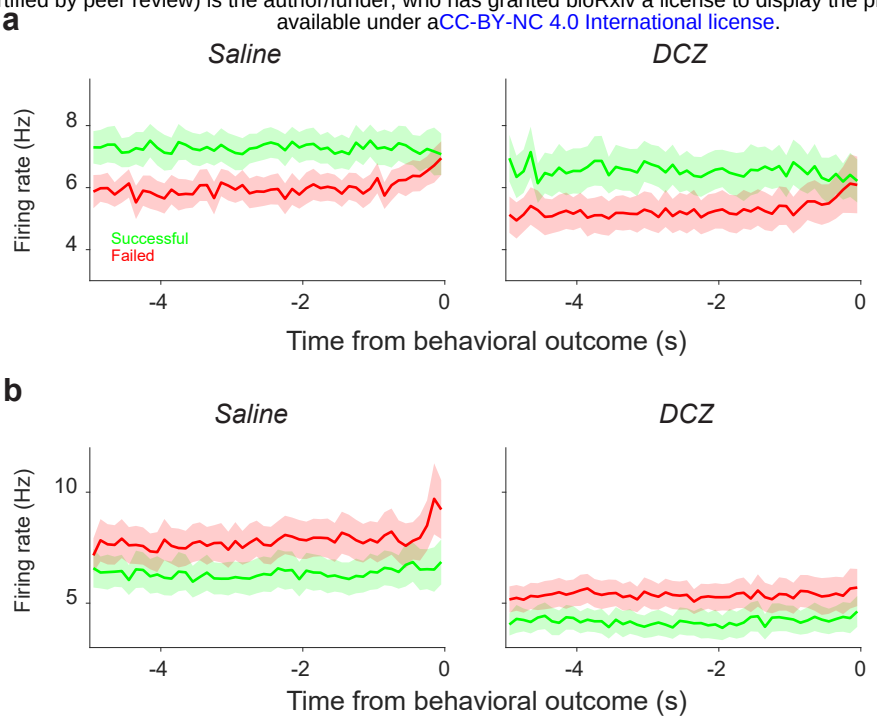
Supplemental figure 3. ***a)*** *mCherry expression in putative LC axons in the basal forebrain region in DBh-Cre mice.* ***b)*** *Raw success rate of WT and DBh-Cre mice in the saline control sessions.* ***c)*** *Normalized behavioral performance of WT and DBh-Cre mice in saline control sessions.*



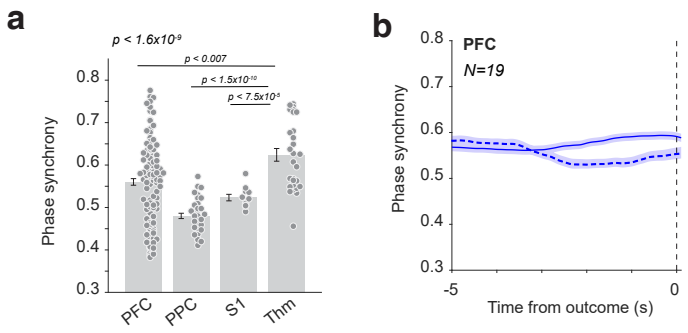
Supplemental figure 4. a) *CNO alone did not affect animals' inhibitory control.* ***b)*** *Difference in switching rate between the successful and failed trials was positively correlated with inhibitory control performance.*



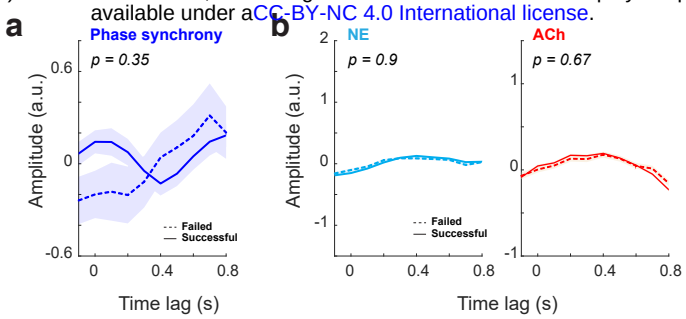
Supplemental figure 5. Chemogenetically silencing cholinergic neurons that project to the LC area did not impair inhibitory control, nor did it change prefrontal NE-ACh phase synchrony. a) *Diagram of retrograde expression of DREADD receptors in cholinergic neurons that project to the LC area.* ***b)*** *Histological confirmation of mCherry expression in the superior cerebellar peduncle (SCP)/Red nucleus (RN), pontine reticular nucleus (PRN), and trigeminal motor nucleus (V).* ***c)*** *Chemogenetic inhibition of cholinergic neurons that project to the LC region had no effect on inhibitory control.* ***d)*** *Chemogenetic inhibition of cholinergic neurons that project to the LC region slowed down the reaction time.* ***e)*** *Prefrontal NE/ACh signals prior to behavioral outcomes in the successful and failed trials under saline or CNO treatment.* ***f)*** *Prefrontal NE-ACh phase synchrony prior to behavioral outcomes in the successful and failed trials under saline and CNO treatment.* ***g)*** *Mean switching rate prior to behavioral outcomes in the successful vs. failed trials under saline or CNO treatment. All data are from 43 saline sessions and 43 CNO sessions from 5 animals.*



Supplemental figure 6. Firing rate of prefrontal encoding neurons during inhibitory control. a) *Population firing rate prior to behavioral outcomes in the successful and failed trials under saline and DCZ treatment for encoding neurons with a higher firing rate in successful trials.* ***b)*** *Population firing rate prior to behavioral outcomes in the successful and failed trials under saline and DCZ treatment for encoding neurons with a lower firing rate in successful trials. All data are from 15 saline sessions and 15 DCZ sessions from 3 animals.*



Supplemental figure 7. NE-ACh phase synchrony in different brain structures during inhibitory control. ***a)*** *NE-ACh phase synchrony is higher in the thalamus than in the cortex. PFC: 165 sessions from 19 animals; PPC: 38 sessions from 4 animals; S1: 11 sessions from 1 animal; Thm: 31 sessions from 3 animals.* ***b)*** *NE-ACh phase synchrony in the prefrontal cortex prior to behavioral outcomes in the successful and failed trials. 165 sessions from 19 animals.*



Supplemental figure 8. Temporal response functions (TRFs) during the free period. a) *TRF mapping prefrontal NE-ACh phase synchrony to pupil size in the successful and failed trials during the free period.* ***b)*** *TRF maping NE (left) and ACh (right) signals to pupil size in the successful and failed trials during the free period.*