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Reduction in NOX emissions during the COVID-19 lockdown did not result in a comparable reduction in secondary PM levels

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The COVID-19 lockdown is viewed as a natural experiment that can put our current understanding of the contribution of secondary $PM_{2.5}$ sources to the test. In ten German metropolitan areas, mean meteorology-accounted for $PM_{2.5}$ concentrations dropped by 5 % during the 2020 lockdown period (spring) compared to 2019, but meteorology-accounted for NO_2 concentrations decreased by 23 % during the same time. Furthermore, meteorology-accounted for SO_2 and CO concentrations show no significant differences between the 2020 lockdown period and 2019. The GEOS-Chem model simulation with COVID-19 emission reduction scenario (23 % reduction in NO_X emission with unchanged VOC and SO2) supports our findings of only a marginal decrease in $PM_{2.5}$ and a significant decrease in NO_2 levels and reveals that the atmosphere's oxidative capacity is increased in all three important oxidants, OH, O_3, and night-time NO_3. The night-time increase in O_3 is the main cause of increase in night-time NO_3 radical. The increase in OH does not compensate for the strong reductions in NO_2, whereas the increase in NO_3 radical at night roughly balances the effects of the NO_2 reduction. As a result, compared to the Business As Usual condition, i.e., no lockdown, day-time PM nitrates are reduced while night-time PM nitrate formation is relatively unaffected. In addition to the above, slightly enhanced sulfate formation and decreased ammonium explain the small reduction in the total $PM_{2.5}$ during the lockdown period. We also investigated the annual spring high $PM_{2.5}$ episodes in German metropolitan areas. Satellite measurements show high ammonia (NH_3) concentrations in the early spring and summer months, when high $PM_{2.5}$ episodes are associated with high NH_3 concentrations in the spring. We find that high atmospheric ammonia concentrations, combined with low temperature and low boundary layer height, are the most favorable conditions for $PM_{2.5}$ formation. Based on our findings, we suggest that emission control policies should be more focused on limiting ozone that should also reduce $PM_{2.5}$. Furthermore, ammonia emissions should be limited in order to control the high $PM_{2.5}$ episodes in winter and spring.