General comments

The manuscript "Chlorine enhances nocturnal heterogeneous uptake of NO_2 in coastal atmosphere under sea-land breeze circulation" investigates the role of chlorine in nocturnal reactive nitrogen cycling during the sea-land breeze (SLB) events using field observations, machine learning, and a chemical box model. It provides valuable observational evidence on the relationship between chloride concentrations and the heterogeneous reaction rate constant of NO_2 (kNO_2), with potentially important implications for coastal atmospheric chemistry. However, the key finding regarding chloride-enhanced NO_2 uptake is not well supported by the current results. There are several methodological issues in the data analysis and model setup, as well as overinterpretations of the results. Major revisions are required before the manuscript can be considered for publication in *Atmospheric Chemistry and Physics*.

Major comments

1. **The relationship between Cl_{total} and kNO_2**

   The key evidence in the manuscript for the chloride-enhanced NO_2 uptake is that Cl_{total} (defined as the sum of gaseous HCl and particulate Cl^-) shows the highest factor importance for predicting kNO_2 in the random forest model (Fig. 3). However, gaseous HCl is itself a product of the heterogeneous reaction of NO_2, as shown by R2-R3 (lines 248-250) and discussed in relation to Fig. 2 (lines 238-244). This means that gaseous HCl is a dependent variable of the NO_2 heterogeneous reaction, so higher gaseous HCl concentrations are expected to be correlated with enhanced NO_2 heterogeneous reaction, regardless of whether chloride is mechanistically promoting the reaction. The random forest model identifies statistical associations, but not causal relationships. Yet the authors used the random forest model results to draw a mechanistic conclusion about the role of chlorine. Moreover, as indicated in the manuscript (lines 241-247), chloride depletion could occur, and once particulate Cl^- is converted to HCl and partitioned into the gas phase, it no longer participates in or affects the aerosol surface properties relevant to NO_2 uptake. To put it simply, why do the authors conclude the role of chloride based on Cl_{total}? This is a fundamental question that the authors need to address. The authors should also clarify and justify the design of the random forest input variables and discuss how including a reaction product as a predictor affects the interpretation of the factor importance.

2. **Concerns regarding kNO_2 calculation**

   (1) The text states that kNO_2 is calculated with the $HONO_{corr}/NO_2$ ratio (lines 147 & 269), but Eq.5 (line 149) calculates kNO_2 using the difference in $HONO_{corr}$ between two time points divided by the mean concentrations of NO_2 over the time interval, multiplied by the time interval. These two calculations are not mathematically equivalent, especially when NO_2 concentrations change over the time interval, which could happen in field observations. The authors should reconcile the text and equation.

(2) For $HONO_{corr}$ calculation, Eq.1 (line 127) combines quantities with inconsistent units – $HONO_t$ and $E_{vehicle,t}$ are concentrations, while $P_{NO+OH,t}$ is defined as a formation rate (Eq.2, line 133). The time step used to convert the rate into concentration should be specified.

(3) For $E_{vehicle,t}$, a single fixed "lower-limit" emission factor of 0.005 is applied for the entire campaign. Given the site's proximity to major traffic arteries, this is likely to be oversimplified without temporal dependence. The authors could consider whether the emission factor can be estimated from observations (e.g., using approaches such as those in Xu et al., *AE*, 2015) or at least conduct a sensitivity analysis. Because the choice of emission factor further affects the calculation of $HONO_{corr}$, kNO_2, and the subsequent evaluation of the contribution of heterogeneous NO_2 on reactive nitrogen cycling with chemical box models (Section 3.3), a clear clarification on this choice and sensitivity analysis might be needed.

3. Concerns regarding the multiphase chemical box model

(1) For model setup, the authors should specify whether the calculated mean kNO_2 on SLB days is applied to scenarios (3) and (4), or whether specific kNO_2 values derived on SLB and non-SLB days are applied to the respective scenarios (lines 178-179). In addition, the authors should compare the HONO deposition velocity used in the box model (4 cm/s, Table S1) with values reported in the cited literature (0.6-3.2 cm/s, Zhang et al., 2003) and justify their choice of this parameter, as it directly affects the simulated HONO concentrations and budgets.

(2) In the model results section, the authors should report the absolute improvement in simulated HONO concentrations and NO_3^- production rates compared to the scenarios excluding NO_2 uptake mechanisms, in addition to the reported improvement in r (lines 336-339). In addition, the authors state that "This massive overestimation implies that the actual difference in kNO_2 between SLB and non-SLB days is likely greater than 1.44-fold, underscoring the amplified importance of NO_2 uptake on SLB days." (lines 342-344). If SLB-derived kNO_2 values were applied to scenario (4) (see comment (1) above), the authors should consider whether the overestimation exceeding the calculated kNO_2 difference between SLB days and non-SLB days could arise from differences in boundary layer height (BLH) and aerosol surface area (S_a), which could influence the simulated HONO concentrations and NO_3^- production rates. This would improve the discussions beyond simply attributing this excess overestimation to amplified importance of NO_2 uptake.

(3) In evaluating the NO_3^- formation enhancement by NO_2 uptake mechanism, the authors compare the simulated NO_3^- production rate against the observed concentrations (Fig. 4b). However, the production rate and concentration are different quantities. This comparison needs to be explained in more detail. In addition, when quantitatively comparing the contribution of NO_2 uptake and N_2O_5 uptake (lines 359-377), the applicability and

uncertainties of the sulfate-based N_2O_5 uptake coefficient (γN_2O_5) parameterization (Table S2) should be discussed.

Minor comments

1. In Fig. 1, the distribution of the observed parameters is fitted with a normal distribution and mean values are used to describe their characteristics. Most species are not normally distributed but show skewness. It would be better to describe the skewed data with median and percentiles.
2. In Fig. 2, the range of observed $HONO_{corr}$ on SLB is substantially higher than on non-SLB days, although there are few data points above 1.5ppb on SLB days. This difference in data range can affect the Pearson r on SLB days relative to non-SLB days. When comparing the overlapped $HONO_{corr}$ range (<1.5ppb), the two groups of data points do not appear to show a substantial difference in data scattering. The author should discuss how the choice of the ranges for correlations may affect their interpretations.
3. Based on Fig. 3h, the authors suggest a synergistic interaction between Cl_{total} and RH (line 311), but this interaction is not obvious in the figure and needs to be more clearly demonstrated. In addition, the authors interpreted the role of RH as supplying water molecules for heterogeneous NO_2 uptake (lines 304-306). Given that the mean observed RH exceeds 60% on both SLB and non-SLB days (Fig. 1f), it is not clear if the amount of water is the limiting factor for NO_2 heterogeneous uptake, and more details should be provided.
4. Some descriptions of the results in the manuscript are overstated and could be revised for accuracy. For example, several moderate correlation coefficients are described as “strong”, e.g., line 213 “the Na^+-Cl^- correlation was strong on SLB days, r = 0.67”; lines 233-235, “a strong correlation between $HONO_{corr}$ and NO_3^-, with the correlation coefficient (r) increasing from 0.56 on non-SLB days to 0.76 on SLB days.” Similarly, some statements overstate the strength of the evidence. For example, lines 297-299, “Subsequent interaction analysis provides evidence that the mechanisms described in reactions 4-5, originally identified under laboratory conditions, indeed occur within the coastal boundary layer”. These should be toned down.

Formatting issues

1. Line 134: the unit for k_{NO+OH} should be written as “cm^3 $molecule^{-1}$ s^{-1}”
2. Line 162: “Cl_{total}” should be defined when it first occurs.
3. Line 235: “reaction 1” should be written as “R1”, consistent with equation numbering below. Similar for other equation numbering.
4. Line 269: in “HONOcorr”, the “corr” should be subscripted
5. Line 340: the figure reference should be “Fig. S10”.