Response to Referee #2
Our responses to the referee's comments are in blue.

1. Since these are the first ambient measurements of HOCl, and the presence of sea spray is mentioned: Can the authors please discuss what interference (if any) could be expected from the presence of sea salt aerosol deposited on the inner walls of the inlet? For instance on pg 8122, line 20, it is stated that the inlet capillary clogged. Assuming that the clog consisted of mainly sea salt, how much of an interference could that have caused? In other words, can the measured HOCl include part of HOCl(aq)? The answer is probably no, but it would strengthen the manuscript if this was discussed.

Added this paragraph after the description of the HOCl calibration technique:
"The observed HOCl signal is very unlikely to be due to reactions producing HOCl on the inner surfaces of the inlet. As discussed later in the manuscript, there are few reactions known that can produce HOCl in the aqueous phase and no known surface-enhanced mechanisms. There is also no indication that sea salt accumulation at the inlet capillary is a source of reactive chlorine signal. Most of the sampled sea salt aerosol is likely very fresh and unlikely to contain measurable levels of reactive chlorine. Lower signals accompany gradual buildup of salt around the capillary."

2. Fig. 2 shows two distinct periods (referred to as high Clx and low Clx days on line 19, pg 8123), separated by a "power outage". Prior to the power outage, high mixing ratios of HOCl and Cl2 are observed, whereas after the outage, the levels appear to be an order of magnitude lower. During a power outage, a lot of things can happen, including a full pressurization of the mass spectrometer. The coincidence of the power outage with such a large change in Cl2 and HOCl mixing ratios is suspicious. Can the authors strengthen the manuscript and make a more convincing case that the sensitivity of the mass spectrometer was not systematically affected by the power outage? Such a case could be made, for example, by including time series of calibrations.

As we point out, we assess the sensitivity of the instrument regularly, and the sensitivity did change significantly over the course of measurements. If sensitivity is overestimated, then ambient mixing ratios are underestimated. HOCl sensitivity did increase after the power outage, but this was a result of a voltage change we made on an electrostatic lens in the front end of the instrument. Cl2 sensitivity decreased as a result of this change. If the changes in calculated ambient mixing ratios were due to changes in calculated instrument sensitivity, one would expect the post-outage HOCl to be lower and the post-outage Cl2 to be higher than before the outage. Yet both Cl2 and HOCl mixing ratios were found to decrease after the power outage.

The only post-outage change we can imagine that could plausibly have caused a systematic bias is a change in the permeation rate of the Cl2 perm tube. For a few hours after the outage it was not flushed with fresh nitrogen. This usually results in higher-than-average permeation rates when the nitrogen flow is returned, and then it returns to usual permeation rates within a few hours. If for some reason the Cl2 permeation rate were irreversibly increased, calculated instrument sensitivity to *both* HOCl and Cl2 would increase, and ambient mixing ratios for both would be underestimated. However, it is unclear what could cause such a change in a perm tube, and as pointed out above, the Cl2 sensitivity in fact decreased after the power outage.

We have included a supplemental figure showing the time series of instrument sensitivity to Cl2 and HOCl over the measurement period.

3. On page 8123, it is stated that "The observed variability in Cl2 and HOCl is likely at- tributable to changes in air mass origin and chemistry." Can this be more substantiated, e.g., by using pollution tracers such as CO or NOy?

We state relationships between Clx and NOy and between Clx and O3 immediately after this statement, and then describe the different air mass regimes for the high and low Clx periods. We also added the statement (also noted later in response to Referee 2's final comment):
"The combination of continental back trajectories, high NOy levels, and low NOx levels indicates that this is aged polluted air which likely originated in Europe."

Comments on the modeling:

4. In Table 2, only one set of input parameters is given, even though in the data there are two distinct periods, identified as "high" and "low" Clx periods. Why is only the "high Clx" case modeled? It seems to me that the model outputs are in greater agreement with the "low Clx period". Consider widening the model-measurement comparison and also discuss how the model outputs compare to the low Clx case.

We pointed out in the first paragraph of Section 5 that we decided to only model the high Clx period because the low Clx period HOCl and Cl2 were often at levels below the detection limit. It is hard to even establish a diurnal pattern such as the one apparent in the high Clx period. We would actually argue that the Cl2 levels in the low Clx period are different from the model simulations, based on what we can see. There was not a clear nighttime increase in Cl2 during this period, and on a couple nights Cl2 was below the detection limit essentially the whole night.

We tried adding text to compare the model simulations to the low Clx observational period, but we found this to be potentially distracting and hard to motivate.

5. Section 5 "Model Simulations" (from page 8124 to the conclusion on page 8131, line 12 "The MECCA modeling results show that it is not possible to simulate the high ob- served levels of HOCl in aged polluted air at Cape Verde, without imposing unrealistic boundary conditions or introducing a new source of Clx.") could be condensed considerably. Personally, I am not convinced that turning reactions on or off in the model with the goal to match observed HOCl mixing ratios is the best approach as there are many unconstrained variables and a match could be due to a fluky cancellation of errors.
Overall, the entire section seems like a long way to go to conclude that there is missing chemistry.

There are really two main points in this section: we seem to be missing an HOCl source in the model, and the conversion of HOCl to Cl2 in aerosols is too efficient in the model. The first point was made fairly succinctly (we think) in the first part of the section. We explored the second of these points in some detail, and perhaps this seemed speculative or digressive. However, we think that we have raised important issues that need to be addressed in order to answer this question. We changed the text of the first paragraph of the modeling section to clarify the approach of the modeling section:

"…often below the detection limit. A base case simulation (described below) did not fit the observed HOCl and Cl2 levels well. Further simulations were then carried out to explore possible means to bring the model closer to the observations.
The model included…"

6. Since the HOCl data are new, I would have personally preferred a narrower approach in the modeling, at least initially. For instance, why not constrain the model to observed levels of Cl2, BrCl, etc. and then answer relatively simple question such as: "Are the observed ratios of HOCl/Cl2 consistent with model predictions?" and go from there.

We think both our and the reviewer's suggested approaches are sensible and would lead to a similar place. The 'best guess' base case simulation does roughly match the observed Cl2 levels, and it is clear that it does not match the observed HOCl levels. This point seems to be what the reviewer's approach aims to address as well.

Also, it is probably not possible to establish model conditions that yield observed levels of Cl2 without the presence of a few ppt of BrCl at night. BrCl was not monitored during the measurement period described here. However, in the week before the measurements described in this manuscipt, and in the published 3-week-long 2007 measurements, BrCl was monitored but never observed above the detection limit.

Minor comments
- the manuscript is internally inconsistent with respect to the use of the subscript (aq). For example, in R1, some ions have the subscript, others do not

Fixed.

- R11 and R12 are not balanced

Fixed.

- pg 8118, line 17 "In photochemical models of marine air, the photolysis of BrCl is a major source of oxidized chlorine in marine air (Pechtl and von Glasow, 2007; Vogt et al., 1996). Surprisingly, BrCl has not yet been detected in coastal or open ocean marine air (Finley and Saltzman, 2008; Lawler et al., 2009)." Is there any reason to trust the modeling work more than the observations? If not, consider rephrasing to say that model outputs and observations are not in agreement.

Instead of ‘surprisingly’ put in ‘however’

pg 8120, line 9. Insert "mixing ratios" between "ozone" and "ranged".

Done.

pg 8121, lines 23-24. "Instrument sensitivity was assumed to vary linearly between calibrations". Please state by how much the sensitivity typically changed between calibrations. Consider adding a time series of calibrations (e.g., as supplemental material)

Added this in supplemental material.

pg 8122, line 18-19 "but it varied by less than 90% between calibrations under similar conditions." I am not sure I understand this correctly. Please rephrase. Is "less than 90%" close to a factor of 2?

Rephrased as suggested.

pg 8124, line 24 and page 8125, line 10. "..... are listed in Tables 2 and 3." I am not sure the reader needs to be told this multiple times.

Removed second instance.

pg 8125, paragraph 5.1. "Best guess simulation"
This paragraph is very confusing. If it’s a "best guess", why does this model give the worst results? Consider labeling this "first guess" or simply "Base case".

Called it “Base simulation”

line 14 "... of only 23 ppt" Please add a statement how much HOCl you wanted the model to achieve (rather than expecting the reader to search for this in tables).

Added phrase “whereas ambient levels were often over 100 ppt.”

Line 14 - line 21 "Increasing O3 and gas phase acids ..." Sounds that model runs other than "base case" are described here. Consider summarizing these under a different heading and justifying each model perturbation. For instance, why increase ozone?

Changed the text to:
“O3 in the *base case* is 45 ppb, already higher than the maximum observed level of 40 ppb, so it does not make sense to increase this further to try to increase HOCl levels. Two independent perturbations to the *base case* were performed to assess the sensitivity of the model results to levels of HCl and NMHCs. Increasing HCl contributes to both aerosol acidity via aerosol uptake and to Cl atom production via HCl + OH. NMHCs reduce the rate of Clx cycling by removing Cl atoms. Fixing model gas phase HCl at 10 ppb results in HOCl levels comparable to observations, but HCl never exceeded 700 ppt during the same season in 2007. Eliminating all NMHCs results in only a 50 ppt HOCl maximum. The base case simulation does not demonstrate good agreement with the HOCl and Cl2 observations, even when modified with major perturbations aimed at improving agreement.”

Line 21. "It seems likely that an additional source of Clx exists." Please add "other than reactions ..."

In response to Referee 1’s comment on this, this statement was removed (see response to Referee 1).

Page 8126, R13. Interesting hypothetical reaction. Is this reaction inspired by K W Oum et al., Science 279 (1998) 74-77?

Not particularly. This was just an easy way to make a reaction that was confined to the daytime.

page 8131 line 5 "We ran a total of 6 box model simulations". I lost count but it seems as if there are more (see comment on page 8125).

Good point. We described 6 simulations in detail, but we mentioned perturbations to the base case as well. Changed this to:
"We ran a number of box model simulations"

pg 8132 line 26. ".. and less acidic aerosols" Can you be more specific here? How much less acidic? In particular, how constrained is aerosol acidity? If aerosol acidity is a critical parameter, how is this used as a model input?

In the lowacid case we state that the pH is 4.3 and this achieves desired HOCl levels.
Added the italicized sentence in paragraph 5.1:

"… were based on measurements made during previous studies at Cape Verde (Read et al., 2009; Lawler et al., 2009). *The fixed levels of HNO3 and HCl are the primary control on coarse (sea salt) aerosol acidity and result in a coarse aerosol pH of 3.4*."

pg 8133. "This study suggests that human influence on Cl chemistry continues for several days over the oceans." I am confused by this statement. The back trajectories in Figure 1 appear to point to the middle of nowhere (or the Saharan dessert, which is sparsely populated) and it's earlier stated that there are no local sources of pollution. Please clarify.

We suspect that we did not state plainly enough that we think the air during at least the first part of the high Clx period is aged polluted air from Europe. Added the italicized sentence to page 8123 at line 27:
"These days showed O3 levels consistently near or above 30 ppb (mean 34.5 ppb), and rising NOy (200–700 ppt). Cl2 and HOCl were at their highest levels during these days. *The combination of continental back trajectories, high NOy levels, and low NOx levels indicates that this is aged polluted air which likely originated in Europe."*