Reply on Anonymous Reviewer No. 2 (AR#2)

We thank the reviewer for acknowledging the relevance of our gas data.

(1a) AR#2: The discussions about the Earth tide are divided into two sections in Discussion and it is difficult for readers to get the key points and the situations you mentioned. Please make it clear about the image of Earth-tide-induced displacements that can occur in the volcanic and magmatic systems, and then discuss about the situation of magmatic systems of Cotopaxi volcano.

Change: We collected the discussion on the Earth tides in a new section 5.4 in order to improve the readability. Further, we switched the order of the remaining section 5.2 and 5.3 for better readability.

(1b) AR#2: The influence of the Earth tides on the ratio of volcanic gas is also unclear. Please mention about it in Discussions.

Change: We added the following clarification to section 5.4:
"(...) In this reasoning, the shallow emplacement of magma prior to the phreatomagmatic explosion in August 2015 may have (temporarily?) shifted the geometry of the Cotopaxi magma plumbing system from a non-excitable to an excitable state. Additionally, the explosive activity very likely gave rise to a transition from closed system to open system degassing, and thus made the volcano more perceptive to external influences. Further, combining our results and the interpretation from Gaunt et al. (2016) suggests a possible tide-induced repetitive plug formation and destruction, causing an alternation of "open and almost shut condition" as proposed by Fischer et al. (2002) for explosive activity at Karymsky volcano. All those effects can result in a periodic variation of the pressure regime in the shallow magmatic system, leading to a periodic variation of the volatile solubility in the magmatic melt, which in turn may vary the magnitude and/or composition of volcanic gas emissions."

(1c) AR#2: Here I list some papers about Earth tides and the volcanic activity that you did not cite:...

We thank the reviewer for providing this supporting literature!

Change: We use those literature for clarifying our interpretation (see section 5.4).

(2a) AR#2: To discuss about the magmatic system in Discussion, I think you need to give us some information about the location and depth of the magma chamber. Are there any previous studies of geophysics (distributions of hypocenters, location and geometry of source of the ground deformation) on Cotopaxi volcano? If so, you should cite such kind of papers.

Change: We added to section 2.1 *"Observations of ground deformation and hypocenter distributions of volcanic earthquakes made in 2001/2002 (Hickey et al., 2015) and 2015 (Morales Rivera et al., 2017) suggest that Cotopaxi currently has a shallow magma reservoir beneath the southwestern flank, which is located at a depth of approximately 5-12 km below the summit."*

(2b) AR#2: And it would be better to write the details about the plug formation and its reliability.

The possibility of a repetitive plug formation and destruction was proposed by Gaunt et al., 2016, as a possible interpretation of their "enigmatic" ash data. We think this was rather a plausible speculation than a empirical evidence and do not want to discuss the reliability. Nevertheless, we think this possible co-occurrence should be included to the list of possible causes.

Change: (none)

(3) AR#2: In Section 3.2, you did not mention about the effect of volcanic ash in retrieval of the SO2 and BrO column amounts. There is some dilution effect in scanning DOAS systems (e.g., Mori et al., 2006, GRL; Kern et al., 2010, BV) and the existence of volcanic ash can result in underestimation of the column amounts. Please mentioned about the problem in this section.

This effect was already discussed in the introduction, but there is no harm to highlight this issue again in the section 3.2. Further, we added the debate on ash scavenging.

Change: We added in section 3.2 *"From August 2015 to February 2016, Cotopaxi also emitted large amounts of ash. These ash emissions can alter the atmospheric radiative transport and thus result in an underestimation of the retrieved SCDs (e.g., Mori et al., 2006; Kern et al., 2010). Nevertheless, those underestimations are approximately the same for both gas species thus their ratio is almost not affected by those ash emissions (Lübcke et al., 2014). Further, it is under debate whether ash has a differential scavenging effect on sulphur and halogens, respectively (Bagnato et al., 2013; Delmelle et al., 2014)."*

(4) AR#2: Minor changes

Change: All applied as proposed.

(5) Further changes

The conclusions focused on the observation and interpretation of the periodic signal. However, also the trend of the BrO/SO_2 molar ratios is an important result of the observations. This trend has been discussed in the manuscript but we did not include it in the conclusions. We added also those findings to the conclusions:
"Previous studies on the volcanic gas plumes of several volcanoes (Mt. Etna, Nevado del Ruiz, Tungurahua) observed relatively low BrO/SO_2 molar ratios prior to volcanic explosions and an increasing trend in BrO/SO_2 molar ratios afterwards. Those consistent observations raised the question whether the BrO/SO_2 molar ratios can be interpreted as a precursor of volcanic activity. We observed a similar behaviour at Cotopaxi during its unrest period in 2015, extending the empirical foundation of this claim. At Cotopaxi, the BrO/SO_2 molar ratios were almost vanishing prior to the phreatomagmatic explosions in August 2015, significantly higher after the explosions, and further increased from September 2015 to December 2015. After December 2015, the unrest calmed down accompanied by a decrease in SO_2-SCDs to a level lower than prior to the explosions, however, the BrO-SCDs remained relatively large. The latter observation suggests that bromine degassed at Cotopaxi predominately after sulphur from the magmatic melt."

Addition to the abstract:
"The BrO/SO_2 molar ratios were very small prior to the phreatomagmatic explosions in August 2015, significantly higher after the explosions, and continuously increasing until the end of the unrest period in December 2015. These observations together with similar findings in previous studies at other volcanoes (Mt. Etna, Nevado del Ruiz, Tungurahua) suggest a possible link between a drop in BrO/SO_2 and a future explosion."