Reply on Anonymous Reviewer No. 1 (AR#1)

We thank the reviewer for the thoughtful and encouraging comments and for acknowledging the relevance of our gas data. The reviewer, first, highlights some misleading statements on the Earth tides themselves, second, and then goes on to question the reliability of the correlation results, and third, asks about a possible explanation for the discrepancy between the observed period of 13.7 days and the astronomic forcing with a period of 14.8 days.
We mostly agree on the criticism of the singular misleading statements due to bad phrasing in our former version and we therefore corrected them (see below). However, we do not share the reviewer's pessimistic view on a correlation coefficient of 47 %. Figure 6 illustrates clearly a sound correlation between the gas data and tidal data. Finally, we have shown that the period in the gas data of 13.7 days matches the temporal variations of the (modelled) North-South component of the tide-induced surface displacement (rather than the periodicity of the total/vertical tidal forcing). It was not the aim of this paper to identify the cause for this partial correlation. Nevertheless, the last ten lines of the discussion discuss the plausibility of a causal relationship between the correlated data, based on the specific environment of Cotopaxi. We just wanted to offer some possible directions for future investigations. Below we give our detailed responses to the individual points of this reviewer.

(1) AR#1: The introduction outlining the history of DOAS measurement is also interesting, but given its length and weight, is slightly beside the point.

We agree with the reviewer.

Change: We shortened the introduction by 12 lines (from 65 to 53 lines, see new manuscript), predominately cutting some of the DOAS part. The new introduction is structured to give an overall motivation for the research on volcanic degassing (10 lines), an overview of the data source we are using (13 lines), an introduction in the analysis of BrO/SO_2 molar ratios (17 lines), and literature on the comparison of NOVAC data and Earth tides (13 lines).

(2) AR#1: The semi diurnal peak-to-peak modulation between S2(K2) and M2 is depicted in Fig1 and has a period of 14.7 days as the excitation mechanism, while the periodigram in Fig 3 reveals a period of 13.7 days describing the response. What is your take on the difference.

We gave a good reason for this apparent discrepancy, which we clarify in the revised manuscript, see below.

Change: The interpretation of the correlation with the Earth tides was formerly split in section 5.2 and 5.3. In the revised manuscript, we collected all interpretation in the new section 5.4 in order to improve the readability of the discussion (see also reply on

the second review). There you can now find, among others, the following interpretation:

"Tide-induced processes are intuitively expected to strictly follow the periodicity of the strongest tidal long-term pattern, that is the spring-neap tide cycle with period of 14.8 days. The observed periodic pattern of 13.7 days in our BrO/SO_2 data, however, matches much better with the temporal intraday amplitude variation of the North-South component of the tide-induced surface displacement, which follows a rather irregular pattern with maxima occurring roughly every 13-14 days. In other words, BrO/SO_2 ratios were elevated when the tidal amplitude variations in North-South direction were most pronounced. Our results accordingly suggest that the volcanic system of Cotopaxi (currently?) is more sensitive to tide-induced stresses acting in the North-South direction, rather than to stresses in the vertical and East-West directions.
This interpretation is further supported by the orientation of the local fault system and associated ambient stress field at Cotopaxi, which is located in a transfer fault zone with greatest principal stress acting in the ENE-WSW direction and the weakest principal stress in the North-South, i.e. also in horizontal direction (Fiorini and Tibaldi, 2012). Such a setting favours the intrusion and ascent of magma along East-West striking planar structures, which is further corroborated by the observation of the inclined sheet intrusion beneath the south-western flank of the volcano (Morales Rivera et al., 2017). Thus, the additional tide-generated stresses probably have a much higher relative impact, when they act in the direction of weakest principle stress, i.e. normal to East-West striking compressible magma pathways, if compared to the other directions. Such a directional dependency is indeed well known for the tidal response of inclined planar aquifers, which cross-cut borehole wells. Bower (1983) e.g. reported oscillations of water levels in boreholes in Canada, which indicated a strong response to the horizontal component of the semi-diurnal M2 tide acting normal to the strike direction of the intersecting aquifers."

(3) AR#1 The statement "The North-South component of the tide has no unique periodicity but a mean periodicity of 13-14 days" in Fig1 reveals the partial understanding of the authors about the tidal potential; this general statement should be removed.

We argue that the content of this statement is crucial for the discussion (see (2)) but agree that "mean periodicity" is not an appropriate term.

Change: We replaced the statement by *"The North-South component of the tide has no strictly regular periodicity but reaches a maximum roughly every 13-14 days and is increasing from September to December 2015."*

The details of the tidal dynamics are virtually numberless and are even more complex when discussing the particular spatial components of the tidal potential. We nevertheless believe that our general understanding of the tidal dynamics is sufficient to interpret the tidal potential (for any given form) as well as its interaction with the volcanic system. Further, the analysis as presented in this paper is not affected by missing knowledge

on the ultimate origin of the pattern in the North-South component, i.e. the tidal time series are just treated as input data.

This said, we can frankly admit that we have not completely understood the ultimate origin of the irregular pattern in the North-South component. Nevertheless, we can give a plausible interpretation of the pattern in the North-South component, although we can not back it up with literature. This interpretation is now included in the manuscript.

Change: We revised Appendix A in order to motivate the irregular pattern in the North-South component. In particular, we added:

"(...) the North-South component follows a rather irregular pattern with a maximum rough every 13-14 days. The discrepancy in beat rate can be explained by the relative impact of the tidal harmonics on the different spatial dimensions. In particular, if the Earth would not be tilted with respect to the ecliptic, there would be no displacement in North-South direction at the equator. Accordingly, the North-South displacement is more sensitive for those tidal harmonics which contribute due to the inclination of the Earth, which is primarily K2. In this reasoning, the tidal harmonics with the strongest North-South component are K2 and M2, thus the dominant long-term pattern has a beat of 13.66 days (K2+M2), rather than the spring-neap tide cycle. The irregularities in the beat rate of the North-South component may be manifestations of the interferences (N2+M2), (N2+S2), and (N2+K2). Furthermore, the North-South component also follows the semi-annual modulation (K2+S2)."

(4) AR#1: Other peaks in the periodigram in Fig 3 are attributed to "probable just artifacts due to spectral leakage" without any further comment. Spectral leakage is caused by the taper length of the time window, and could have been properly defined, if it is indeed the reason for the additional peaks.

We are not sure whether the review refers to the common Fourier transform analysis or to the here applied Lomb-Scargle analysis (because our time series has an uneven sampling). For Lomb-Scargle, things are typically more complicated and we do not know a proper way to check/prove spectral leakage. Instead we tested the Lomb-Scargle for possible artefacts as follows: We generated a sinusoidal signal with a period of 13.7 days and with a length of 6 periods and a sampling rate of twice a day. Then we removed random data points such that we got an uneven sampling which had the same number of data points as our BrO/SO2 data. Finally, we applied the Lomb-Scargle analysis on this "gapped" sinusoidal signal.

We found that the amplitude and position of the side lobes vary for different sets of randomly removed data points. Further, the side lobes are not symmetric around the central maxima. Nevertheless, the peaks observed in Fig 3 are close to the mean amplitude and position of the set of sinusoidal test data. Thus, we can not exclude that those are just artefacts. However, we agree with the reviewer that we have not proven that those are indeed just artefacts.

Change: (1) We removed the sentence on the spectral leakage in order to avoid a

misleading interpretation of those minor peaks which are actually not important for the further analysis. (2) For completeness, we added the further results of the Lomb-Scargle analysis: *"On lower confidence levels, the Lomb-Scargle analysis proposes further peaks at a periodicity of 7.1 days, 9.8 days, and 18.8 days, respectively."*

(5) AR#1: Fig 5: the expectation of a phase shift between excitation and response is indeed justified and could provide important information about the underlying mechanism. In this way sediment porosity, e.g., has been determined by evaluating the response of water-filled boreholes to the tidal potential. After applying a phase shift of about 1.7 and 10 days, respectively, the resulting correlation between tides and volatile ratio is merely 0.47, which is not convincing at all. Fig 6 (left panel) demonstrates the weak significance. In the conclusions the authors describe the correlation with humidity as only 33% while 36% is considered a promising explanation??

In contrast to the reviewer, we consider a correlation of 47% to be unexpectedly high, convincing that gas data and North-South tide are partially correlated (see Fig 6, right panel). We see this statement justified already mathematically but also justified by Fig. 6 right panel (which is just a different graphical representation of Fig. 6 left panel), which clearly shows that there is a partial correlation between the tide-induced displacement and the gas ratio. Further, we did not aim to label correlation coefficient of 33% of the relative humidity as insignificant. Finally, we have to highlight that we have not claimed any "promising explanation" but just described the results of the statistical analysis.

We thank the reviewer for mentioning the response of water-filled boreholes. We had a similar mechanism in mind but missed the literature on the empirical studies.

Change: (1) We added literature on borehole observations. (2) We clarified our interpretation of the correlation results by adding the following paragraph (in the newly created section 5.4): *"The observed periodicity in the time series of the BrO/SO_2 molar ratios is superimposed by an increasing trend and a large scatter in the data. The latter highlights the complexity of the interpretation of BrO/SO_2 molar ratios, which potentially depends on an unknown number of volcanological and atmospherical mechanisms and the fluctuations of their parameters. Despite the large scattering, we nevertheless found an unexpectedly high correlation between the BrO/SO_2 molar ratios on the one hand and the Earth tidal forcing or the relative humidity on the other hand, with correlation coefficients of 47% and 33%, respectively. Thus, the tidal forcing as well as the relative humidity are the most probable candidates to explain a part of the variability of the BrO/SO_2 molar ratios. Accordingly, both mechanisms may independently contribute to the variation of the BrO/SO_2 molar ratios at the same time. In the following, we focus on the plausibility of a causality between the BrO/SO_2 molar ratios and the North-South component of the tidal forcing, which nevertheless appears to be the best candidate"*

(6) AR#1: Appendix A: Addressing the response of the Earth to the tides "The water in the oceans responds..." the authors seem to confuse the amplitude response with the phase. Ocean tides can be completely out of phase with the body tides due to eigen oscillations in bays and estuaries, while the response of solid rock in the crust is smaller than 1 degree, because it is elastic. Love numbers describe exactly this effect. Hence, the final conclusion about the relative displacement between melt and elastic rock needs to be re-considered in that light.

The sentence on the ocean tides has been included in order to illustrate the impact of viscosity, rather than discussing the ocean tides in detail. The reviewer is correct about the complexity of the ocean tides (anyway, our sentence was meant for the open sea). Because the ocean tides are of no relevance in the manuscript and in order to avoid misleading content, we removed the sentence on the ocean tides.

Change: The sentence *"The water in the oceans responds to variations of the tidal forcing almost immediately and is thus displaced always with the theoretical value, e.g. 1m at spring tide."* is removed.

In the next sentences of the manuscript, we used the terms "slower" and "faster" which are typically attributed to velocity or time (probably the reviewer's criticism focuses on those?!). In our manuscript, those should refer to the amplitude of the maximum displacement rather than a phase shift (i.e. whether the magmatic melt reach the maximum is rather a question of time than of elasticity). But the reviewer is definitively right that those terms are much more plausible when talking about a phase shift and are thus misleading here.

Change: We changed the two sentences to *"Solid rock is displaced by about 0.2980 (radial Love number of the SNREI Earth, see Agnew, 2007) times the theoretical value, e.g. $\pm 0.3\,m$ at spring tide. In contrast, magmatic melt is a fluid with a higher compressibility than solid rock and may therefore adopt stronger to the tidal potential."*

(7) 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 explo-

sions 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."