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Atmospheric Measurement Techniques Discussions



# *Interactive comment on* "Theoretical description of functionality, applications, and limitations of $SO_2$ cameras for the remote sensing of volcanic plumes" *by* C. Kern et al.

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## 1 Introduction

We would like to thank the reviewers for their helpful comments on the submitted manuscript. We agree with most of the mentioned points and have modified the manuscript accordingly. We feel that the revised manuscript is much improved now that their suggestions have been incorporated. The responses to the individual reviewer comments are listed below.



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## 2 Responses to Reviewer 1, Anonymous

### 2.1 General comments

***RC1:*** *The main problem of this paper is the inadequate description of the assumptions used in the measurement principle and the lack of analysis of impacts on the measurements due to deviations from these assumptions.*

**AC1:** It is one of the main goals of this paper to discuss the theoretical background of the $SO_2$-Camera technique. Therefore, we agree that a careful description of the assumptions on which the presented calculations are based is of fundamental importance. The manuscript was modified to describe these assumptions more explicitly. Details are listed in the points below.

***RC2:*** *Specifically, the first assumption was implicitly made on page 535, in writing down equations (1) to (3). The authors assume that all photons that reach the CCD detectors in the camera have to pass the* $SO_2$ *plume. This assumption should be explicitly stated in the paper. It is valid if photons scattered into the camera by air mass between the camera and the plume, and photons reflected by aerosols in the plume are negligible. Clearly when photon contribution deviates from this assumption, such as in the cases of long distances between the camera and the plume and bright aerosols within the plume, accuracy of the derived* $SO_2$ *column amount decreases, as more photons arriving at the camera without experiencing the* $SO_2$ *absorption.*

**AC2:** The effects mentioned above are radiative transfer effects which have been known for some time (e.g., Millan, 1980). We refer to them as radiation dilution (radiation entering the instrument's field of view between the plume and the instrument) and



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multiple scattering (enhanced light paths in the volcanic plume due to scattering). A recent study by Kern et al. (2010) has shown that both radiation dilution and multiple scattering in a volcanic plume can be corrected for using moderate resolution spectral data, as measured by a DOAS instrument (cited on page 536, line 2). Unfortunately, this method cannot be applied to data taken using the $SO_2$ camera technique, and it is true that the accuracy of this technique is to a large part determined by the radiative transfer effects. However, no implicit assumptions are made in writing down equations (1) to (3). Instead, these equations are kept very general, and we refer to the "effective light path". We explicitly state that the effective light path is equal to a straight line through the volcanic plume only under ideal circumstances. To clarify the issue more, we have expanded the paragraph ensuing equation (3). It now reads: "Unfortunately, the effective light path L can deviate substantially from the line of sight through the plume. Especially in cases where volcanic plumes contain large amounts of $SO_2$ or scattering aerosols or are viewed from a large distance, the effective light path may be significantly shorter or longer than a straight line through the area of interest. Radiation entering the field of view between the plume and the instrument ('radiation dilution') shortens the effective light path, while multiple scattering inside the plume elongates it. In fact, the absorption of radiation by $SO_2$ in volcanic plumes is oftentimes sufficiently strong in itself to influence the effective light path. Therefore, the column density S is typically a function of the wavelength $\lambda$. All of these effects were recently described and quantified by Kern et al. (2010), along with a suggested method for their correction using moderate resolution spectral data e.g. obtained from a DOAS instrument."

***RC3:*** *Additional assumptions about aerosol absorption and scattering are also implicitly made in section 2.1 of the paper. As correctly described in section 2.1, aerosols can reduce the number of photons that pass through the* $SO_2$ *plume by scattering away the radiation (this intensity reduction is denoted by* $I_{-}(\lambda)$*) entering from the back side, and at the same time can increase the number of photons without experience the* $SO_2$ *absorption by scattering radiation into the field of view (this intensity addition is*



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*denoted by $I_+(\lambda)$). In writing down equation 10, the authors have made an implicit assumption that the photon reduction $I_-(\lambda)$ and addition $I_+(\lambda)$ can be neglected.*

**AC3:** Please see AC5.

***RC4:*** *Another assumption about aerosol absorption is made in equation 10, i.e., the aerosol absorptions in wavelength band A and band B are the same. Though not exactly true due to the spectral dependence of aerosol absorption, this assumption is probably quite good because of the small wavelength separation between bands A and B.*

**AC4:** The concept of the normalized optical density or apparent absorption does rely on the assumption that both aerosol absorption and aerosol scattering are only weakly dependent on wavelength and therefore approximately equal in the filter transmittance windows of filters A and B. To emphasize this fact and give an estimate of the associated error, we have added the following passage after equation (10): "Obviously, this approach assumes that the influence of aerosol scattering on the measured intensities is equal in both observed wavelength ranges. As typical $°Angstrom exponents of volcanic aerosols lie between 0 and approximately 2 (e.g. Spinetti and Buongiornio, 2007), depending on their size distributions, the maximum error associated with this assumption is about 10%, and typical errors are around 5%. However, the radiative transfer between the sun and the instrument can be dramatically changed by aerosols in the volcanic plume, thus possibly changing the effective light path by 70% or more (Kern et al., 2010). This needs to be taken into account when calculating concentrations or fluxes from the measured optical densities. The condition that $SO_2$ must be the dominant narrow-band absorber in the filter A wavelength window remains a prerequisite for an accurate retrieval of the SO2 column density S."



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***RC5:*** *The assumption about aerosol scattering contributions is valid only for small scattering optical thickness, but fails when it is large. One can easily see this by writing* $I_A(\lambda)$ *explicitly with addition and subtraction of scattering intensity,* $I_A(\lambda)$ = $(I_{0,A}(\lambda) - I_-(\lambda))exp(-\tau_g(\lambda) - \tau_a(\lambda)) + I_+(\lambda)$*, where* $\tau_g(\lambda) = \sigma(\lambda)S$ *is the* $SO_2$ *absorption optical thickness, and* $\tau_a(\lambda)$ *is the aerosol absorption optical thickness. Putting this equation into* $I_{M,A}(\lambda)$*, and similarly into* $I_{M,B}(\lambda)$*, one notices that in general the aerosol scattering terms from bands A and B do not cancel out each other in equation 10. Only with the assumption that* $I_-(\lambda)$ *and* $I_+(\lambda)$ *are negligible and that* $\tau_a(\lambda)$ *is independent of* $\lambda$*, can one arrive at* $\tau = AA$*, that is directly related to the* $SO_2$ *column density.*

**AC5:** Together with RC3, this comment again deals with radiative transfer. Due to the complexity of this issue, we intentionally separated the problems of radiative transfer from those associated with the $SO_2$-Camera technology itself in this manuscript. Conceptually, aerosol scattering and absorption along the "effective light path" can be corrected by measuring the intensity around 325 nm and normalizing by this value. This works well as long as $\tau_a$ is largely independent of wavelength (see AC4). No constraints on the actual magnitude of $\tau_a$ are necessary. The contributions $I_-(\lambda)$ and $I_+(\lambda)$ introduced by the reviewer are simply another way of formulating the influence of a change in effective light path of the measured incident radiation. However, this is an oversimplified approach to a complex problem. E.g., simply subtracting an additive term $I_-(\lambda)$ from the original spectral radiance $I_0(\lambda)$ only compensates radiation scattered out of the ideal light path at the very back edge of the plume, but it does not incorporate radiation scattered into the viewing direction somewhere in the middle of the plume. The true radiative transfer situation can only be accurately assessed by simulation with a 3 dimensional radiative transfer model, which is beyond the scope of this study and is discussed in detail in Kern et al. (2010). Since the mentioned radiative transfer effects influence all passive remote sensing techniques that use scattered solar radiation as a light source, we believe it is best to discuss these effects



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separately and only include appropriate references in this manuscript. In section 5.3, we also specifically recommend collocating a DOAS instrument with the camera in order to be able to correct for radiative transfer effects.

***RC6:*** *In summary, adding measurement with filter B does not necessary take care the error due to scattering. The statement in the abstract (Page 532, lines 6 - 8), "the effect of aerosol scattering can be eliminated by additionally measuring the incident radiation around 325 nm where the absorption of* $SO_2$ *is no longer significant, thus rendering the method applicable to optically opaque plumes", is not correct in two aspects: 1) the word 'eliminated' is too strong, replacing it with 'reduce', which is more appropriate due to the* $\lambda$ *dependence of aerosol absorption, 2) replace 'optically opaque' with 'optically thin', because* $SO_2$ *measurement can not be achieved for optically opaque plume, simply because little or no photon experienced its absorption would reach the camera, therefore this statement need to be rewritten.*

**AC6:** The sentence was changed to: "The effect of aerosol scattering can in part be compensated by additionally measuring the incident radiation around 325 nm, where the absorption of $SO_2$ is about 30 times weaker, thus rendering the method applicable to optically thin plumes. For plumes with high aerosol optical densities, collocation of an additional moderate resolution spectrometer is desirable to enable a correction of radiative transfer effects."

***RC7:*** *I recommend the authors rewrite section 2.1, with explicit expression about aerosol scattering and absorption, and derive the apparent absorption for SO2, and clearly state the approximations needed for accurate* $SO_2$ *retrievals.*



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**AC7:** Please see AC5.

***RC8:*** *Also the statement, "...cameras for remote around 325 nm where the absorption of $SO_2$ is no longer significant", is true for small $SO_2$ loading only. When the loading is large, the absorption is quite significant at 325 nm, resulting in $SO_2$ estimate error. Due to non-linear $SO_2$ absorption, the radiation intensity at band A is reduced disproportionally compared to that at band B for large $SO_2$ absorption, making the calibration of the instrument even more difficult. In other words, this $SO_2$ camera is more accurate when measuring low loading $SO_2$ plume.*

**AC8:** The sentence has been changed to "...where the absorption of $SO_2$ is about 30 times weaker". The effect of nonlinearity is discussed in section 2.3.3. The measured normalized optical density of the instrument is not directly proportional to the column density. Deviations occur especially for high $SO_2$ columns. Empirical calibration using gas cells filled with $SO_2$ can capture this effect if the column density of the calibration cells are similar to those of the volcanic plume. In this case, the only additional error introduced is again the deviation of the effective light path from a straight line through the plume. This can only be corrected for by collocating a DOAS instrument and retrieving the radiative transfer according to Kern et al. (2010).

2.2 Technical corrections

***RC9:*** *Page 532, line 17, change "The thus" to "Thus the"*

**AC9:** "Thus" was removed (see AC17).



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***RC10:*** *Page 534, line 5 - 8, move 'only' to after 'a single direction'.*

**AC10:** This has been corrected.

***RC11:*** *Page 536, line 2 - 3, "Therefore, the column density S is typically a function of the wavelength $\lambda$". Please rephrase this statement, because column density by definition is not a function of wavelength. Photons at different wavelength go through different average paths to reach the detectors, therefore experience different amount of the absorption due to the same column density. If a correct radiative transfer is done, different photon paths will be correctly accounted for, and the inversion should yield the same column amount for different wavelengths.*

**AC11:** This is a question of definition. In DOAS-related literature, the column density S is usually defined as the integrated concentration along the effective light path taken by the measured photons (e.g., Frieß et al., 2006; Marquard et al., 2000). In addition, the term "vertical column density" (VCD) is often found. The VCD is defined as the integrated concentration of an absorber along a straight vertical line. The so-called "air mass factor" (AMF) is defined as the ratio S/VCD, and is typically wavelength dependent because the effective light path (and therefore S) varies with wavelength.

***RC12:*** *Page 537, line 16, "In all other cases there is no analytical solution of equation (8) for the column density S, because the incident scattered solar radiation spectrum, the filter spectral transmittance and the quantum yield of the detector are not analytical functions." This statement seem to be out of place: no solution of equation 8 has nothing to do with whether analytical expressions are available or not for $I_S(\lambda)$, $T_A(\lambda)$, and $Q(\lambda)$. Equation (8) can not be solved (neither analytically or numerically), because $I_S(\lambda)$ is not known and is not directly measured. Assuming that $I_S(\lambda)$, $T_A(\lambda)$, and*



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*$Q(\lambda)$ can be measured with sufficient accuracy, conceivably one can numerically solve equation 8 by adjusting the value of S until the equation is satisfied. Getting the value of S is the goal. However one achieves this, analytically or numerically, is not quite important.*

**AC12:** The sentence has been changed and now reads: "In all other cases, Equation (8) cannot easily be solved for the column density S because the incident scattered solar radiation spectrum $I_S(\lambda)$, the filter spectral transmittance $T_A(\lambda)$ and the quantum yield of the detector $Q(\lambda)$ are not well known. They must either be accurately measured (which is especially difficult for $I_S(\lambda)$, as it is constantly changing and needs to be measured behind the plume) or an empirical instrument calibration must be conducted to determine the relationship between the weighted average optical density and the SO2 column density S."

## 3 Responses to Reviewer 2, Fred Prata

### 3.1 General comments

***RC13:*** *The only major item that I would like to see in this paper is a summary Table showing the errors arising from the different effects described in the paper (e.g. optics, choice of filters, interference from other absorbers, calibration, radiative transfer). This should be followed by an error analysis from the results obtained with the prototype camera in comparison to the DOAS measurements. This would give readers a succinct way to evaluate the potential usefulness of an SO2 camera for their applications.*



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**AC13:** A table has now been included (see AC44).

3.2 Technical corrections

***RC14:*** *Abstract, P532, L1. Change "technique" to "device".*

**AC14:** This has been corrected.

***RC15:*** *Abstract, P532, L8. I suppose if the plume were really optically opaque then it would be difficult to determine SO2 accurately. So there must be an optical thickness were the retrieval becomes difficult.*

**AC15:** This sentence has been rewritten (see AC6). A moderate resolution spectrometer (e.g. DOAS) is needed to correct for radiative transfer effects. Retrievals become impossible if the optical thickness is too high to allow light to pass through the plume.

***RC16:*** *Abstract, P532, L10. What is so significant about sampling at 1 Hz? Is this a volcanological requirement or something to do with plume dynamics or perhaps a fundamental sampling limit for these UV CCDs? Would 10 Hz sampling be better?*

**AC16:** A high sampling rate allows the direct observation of processes occurring on the associated time scale. Examples could include dynamical processes in the atmosphere (e.g. turbulent diffusion mixing ambient air into the plume in the first seconds or minutes), or perhaps case studies of the progression of recurring gas/ash eruptions. In principle, 10 Hz would of course be even better for certain case studies (see e.g. Taddeucci et al. (2009)), although 1 Hz is probably sufficient for monitoring



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applications.

***RC17:*** *Abstract, P532, L17. Delete "thus" after "The".*

**AC17:** This has been corrected.

***RC18:*** *Abstract, P532, L21. Change "In addition" to "Thirdly".*

**AC18:** This has been corrected.

***RC19:*** *Abstract, P533, L1. Change "chosen setup" to "setup chosen".*

**AC19:** This has been corrected.

***RC20:*** *Abstract, P533, L2. Change "the instrument" to "an instrument".*

**AC20:** This has been corrected.

***RC21:*** *Introduction, P533, L16. Delete comma after "both".*

**AC21:** This has been corrected.

***RC22:*** *Introduction, P533, L25. Delete "e.g.".*



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**AC22:** This has been corrected.

***RC23:*** *Introduction, P534, L5. Change from "... allow to determine the ..." to "... allow determination of the..."*

**AC23:** This has been corrected.

***RC24:*** *Introduction, P534, L19. So if the time-scale is order of seconds, why do we need 1 Hz sampling?*

**AC24:** According to the sampling theorem, only processes occurring at frequencies at or below the sampling frequency can be correctly captured. If a process occurs on the order of seconds, a sampling rate at or below this frequency is needed. Therefore, a 1 Hz (order of magnitude) sampling rate is desirable.

***RC25:*** *Introduction, P534, L29. Change "discussed methods" to "methods discussed".*

**AC25:** I don't think this makes sense.

***RC26:*** *P535, L17. I think the correct name for this law is "Beer-Lambert-Bouguer". Bouguer was first to describe this law in a paper published in 1729 "Essai d'optique sur la gradation de la lumiere", which predates both Beer's and Lambert's expositions, but somehow it is often quoted as the Beer-Lambert law or often just "Beer's Law".*

**AC26:** You're right! Credit is now given to those who deserve it...



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***RC27:*** *P536, L8-L9. I don't really see any fundamental difference between the spectroscopic measurement principle and that of the SO2 camera, at least in a theoretical sense. The SO2 camera could, in principle, be an imaging spectrometer.*

**AC27:** The $SO_2$ camera can indeed be seen as an imaging spectrometer, albeit one with an optical resolution which is typically insufficient to resolve the $SO_2$ absorption bands. We have now explicitly included the wavelength dependency of the optical density $\tau(\lambda)$ in the text and changed the sentence to: "A spectroscopic instrument with sufficient resolution would measure the optical density $\tau(\lambda)$ to retrieve the column density S, but the $SO_2$ camera instead measures the integral $I_{M,A}$ of the incident intensity $I_A(\lambda)$ over the transmission range of the filter..."

***RC28:*** *P537, L16. I don't think the reason for there not being an analytical solution is because these aren't analytic functions, although this is obviously true. More important, I think, is that any errors in the $I_S(\lambda)$ will ensure that there is no unique solution and maybe not even a stable solution.*

**AC28:** This sentence has been changed (see AC12).

***RC29:*** *P537, L24. Change "to analyze where" to "analysis of where the".*

**AC29:** This has been corrected.

***RC30:*** *P538, L28. Delete "radiation".*



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**AC30:** This has been corrected.

***RC31:*** *P545, L28, L29. Need spaces in the words in the braces.*

**AC31:** This has been corrected.

***RC32:*** *P548, L1. Change "not linear" to "non-linear"?*

**AC32:** This has been corrected.

***RC33:*** *P547, L7. Stating that the zenith angle is time dependent is not really the important point. The zenith angle is also location dependent. In the Arctic circle, for example, during summer, the Sun is always above the horizon. Likewise, it is possible to have the same SZA at different times of day depending on where you are making the measurements. I understand the point but I think it could be better explained.*

**AC33:** The segment has been rephrased and now reads: "The spectrum of incident scattered radiation $I_S$ depends on the solar zenith angle (SZA) and on the prevailing weather conditions (clouds and fog can significantly alter $I_S$). Therefore, the spectrum of incident solar radiation typically changes with time."

***RC34:*** *P549, L14. Change "exchanges" to "exchange".*

**AC34:** This has been corrected.



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***RC35:*** *P550, L17 onwards. Maybe consideration of the integration time and sample averaging are also important here.*

**AC35:** In this section, the ideal maximum transmission wavelength $\lambda_C$ and transmittance curve FWHM of the applied band-pass filters are investigated. The ideal configuration is found by determining the maximum signal-to-noise-ratio (SNR). The integration time / sample averaging are independent of this. The higher the SNR, the smaller the statistical error of a measurement conducted for any given integration time.

***RC36:*** *P550, L18 onwards. Does this also depend on the SO2 amount? For example might one choose different filter combinations if only interested in low concentrations, for example, from ships or boundary layer industrial pollution sources. Likewise, is it possible that different combinations may be better suited for industrial power plant emissions with highly concentrated SO2 emissions.*

**AC36:** This is a very interesting point. The camera design presented in the manuscript is optimized in sensitivity for $SO_2$. This is definitely the best for applications with low $SO_2$ concentrations where the measurement is limited by the sensitivity. However, for very high $SO_2$ column densities (e.g. in volcanic plumes with high $SO_2$ loads), it could be advantageous to move filter A towards longer wavelengths. This decreases the sensitivity of the system, but the effective light path becomes less dependent on wavelength. In fact, the effective light path will be closer to a straight line (i.e. the air mass factor (AMF) will be closer to unity) in many cases. If radiative transfer is neglected in the $SO_2$ retrieval, this is of course an advantage. However, the results will not always be more accurate. In situations where the measured plume contains a large amount of scattering aerosols, the effective light path is typically significantly enhanced by multiple scattering in the plume. In these cases, the reduction of the effective light path by strong $SO_2$ absorption at shorter wavelengths can compensate



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the enhancement by multiple scattering, thus leading to a more accurate result (see Kern et al. (2010) for details). Therefore, while there are some situations where a camera with lower sensitivity will give more accurate results, the only way to really assess this is by retrieving the true AMF with a collocated DOAS (also discussed in Kern et al. (2010)).

***RC37:*** *P554, L20 onwards. Can the effects of ash be corrected by including more filters? For example, OMI and TOMS (previously) have an absorbing index that is sensitive to ash (apparently).*

**AC37:** Both wavelength channels of the $SO_2$ camera are sensitive to ash. If only one channel is considered, absorption by ash is clearly visible as was e.g. shown by Yamamoto et al. (2008), although this holds true for any UV/vis wavelength. When the data is normalized by the second channel, the ash absorption cancels out as can be seen in Figure 12. However, ash clearly influences the radiative transfer. It might be possible to detect the presence of ash without the introduction of an additional wavelength by simply comparing the measured intensity at 325 nm with the intensity simulated by a radiative transfer model that assumes a pure Rayleigh atmosphere. This is how TOMS and OMI do it, although they need an additional wavelength to retrieve the surface albedo (e.g., Torres et al., 1998). However, this approach will not work in the presence of clouds (either in front of or behind the plume), and is susceptible to errors when both scattering (sulfate) and absorbing (ash) aerosols are present. Simply adding an additional wavelength or two in the UV/vis range is not likely to resolve these problems. The thermal IR is better suited for quantitatively measuring ash (Prata and Bernardo, 2009). We prefer not to speculate on these issues in the manuscript.



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***RC38:*** *P554, L26. "... radiative transfer can effectively be retrieved ...". This does not make sense. Radiative transfer is a process. I assume the authors mean that RT can be used to retrieve something. Please re-word.*

**AC38:** "radiative transfer" was replaced with "the effective light path".

***RC39:*** *P555, L22. Delete "with" after "cells".*

**AC39:** This has been corrected.

***RC40:*** *P556, L14. Change "co-located" to "collocated" (Latin root).*

**AC40:** This has been corrected.

***RC41:*** *P556, L19. Delete "the" before "perhaps".*

**AC41:** This sentence was re-worded.

***RC42:*** *P557, L6. Use of the expression "For one, ..." suggest some follow-on, e.g. secondly, thirdly... I suggest this be re-worded.*

**AC42:** "For one" was removed.

***RC43:*** *P558, Limitations of SO2 cameras. I'm not sure that the inability of the camera to measure other trace gases is a "limitation". The camera is presumably designed to*



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*measure SO2 and not other trace gases. I suppose it might be possible to add more filters to measure different gases that absorb through the UV.*

**AC43:** To our knowledge, it is not possible to measure other compounds by simply adding additional filters. No other known UV absorbers exhibit optical densities high enough to dominate absorption at a particular UV/vis wavelength in volcanic plumes. Therefore, the method cannot easily be applied to other gases. This is the mentioned limitation in comparison to some other spectroscopic techniques (DOAS, FTIR).

***RC44:*** *Limitations of SO2 cameras. I suggest that a Table be included here (or perhaps before) showing the magnitudes of the main error sources. This could be corroborated by listing the differences between the prototype camera measurements and the DOAS data (e.g. calculate the bias and rms error from Figure 14).*

**AC44:** A table has now been included in the manuscript giving an overview of the error sources, their magnitudes, and possible correction methods. However, we feel that calculating the bias and rms from Figure 14 could be misleading, as the $SO_2$ camera's results were obtained by calibration with the DOAS system and the values are therefore not independently obtained. As mentioned in the text, the main discrepancies in measured column density are thought to originate in an imperfect match of the field of view of the two measurements.

***RC45:*** *P560, Future work. I think the cost specified (€0,000) is a factor 2 too low.*

**AC45:** You are probably right, we now estimate 20,000€˙



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***RC46:*** *Figures. I suggest replacing the "commas" with "decimal points" in the ordinates of all relevant Figures (e.g. Figs. 1, 3, 4, 5(a), 6, 8, and 9).*

**AC46:** This has been corrected.

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