First of all we would like to thank Jeff Severinghaus for his comments and suggestions. By incorporating these suggestions we considerably improved the paper.

The authors show convincingly that krypton (specifically, low-energy doubly-charged Kr-86 ions appearing in the tail of the beam) interferes with methane measurements in atmospheric samples that are analyzed as CO_2 in the mass/charge 44, 45, and 46 Faraday cups. The paper is very well documented, treats the subject thoroughly, and is written very clearly. It is relevant to the journal and is appropriate for the readership. Overall, this is an excellent paper and should be published with only very minor revisions.

It would be interesting to explore a little further why such a large tail exists for krypton. The authors mention interaction with the helium in the source, and point out that pure krypton has a much narrower peak. This is probably a correct hypothesis. I suspect that the basic reason for the big tail has to do in some way or other with low-energy ions, which are more strongly deflected by the magnetic field than ions having the nominal accelerating energy. It would be good to mention "low-energy ions" in the paper **[1]**. [By the way this suggests that one possible solution might include an electrostatic analyzer, to eliminate low-energy ions.] I once had a problem with graphite ferrules adsorbing krypton, and wonder if the systems discussed by the authors have any of these ferrules. Not that this would explain the interference - rather, this adsorption led to a very large memory effect lasting many hours after putting pure krypton into the machine **[2]**. But it might exacerbate the problem by boosting the amount of krypton above what would normally be encountered in air. Another idea for dealing with the issue might be to put a m/z=43 cup in the mass spec so that the krypton problem could be monitored in real time **[3]**. This problem is reminiscent of a problem we have long been dealing with in the lab, the fact that the large low-energy tail of the argon-40 beam falls into the m/z = 38 cup. It adds to the mass 38 peak by about 1%, which makes the delta values about 1% too small (too near zero). For precise 40/38 measurements it is necessary to make a correction for this issue, much as the earlier generation of mass spectrometrists corrected for the CO2 tails.

Response by Schmitt et al.

Your comments and description of your own experiences with similar phenomena were very useful and helped improve the final version. To respond to your individual statements, we added numbers in brackets in your review, i.e. **[1]**, and respond to them individually below:

[1] Regarding your statement that low-energy ions are more deflected by the magnetic field, we want to add that for the m/z 43 beam, $^{86}Kr^{2+}$, this actually leads to a tail into the low-mass region and not to higher masses, what we observe. So your theory with low-energy ions rather applies to the more abundant singly charged ions which occur in the m/z range between m/z 78 and m/z 86. In this case the m/z distance from m/z 44-46 to m/z 78 is larger, implying a large energy loss in the source, yet would explain the flat Kr signal in the m/z range of m/z 46. An experimental hint to effects at such wide m/z distances are our observations during the measurement of CF_4, where we observed strong Kr signals in the m/z range of 71-74.

[2] While we have a vespel ferrule at the reducing union at the source valve we do not see long-term memory effects for krypton. For example, the peak shape of the Kr peak in Fig. 3 or 4 is very close to CO_2 or has even less memory.

[3] Actually, we do have a cup simultaneously monitoring $^{84}Kr^{3+}$ (m/z 28, see Fig. 28) while measuring $\delta^{13}C$-CO_2 in our mass spec, thus, this information could have been used. But we identified this $^{84}Kr^{3+}$ signal at m/z 28 only after we had identified the Kr problem (and one has to closely look at the m/z 28 signal because the $^{84}Kr^{3+}$ signal produces only a shoulder since CO dominates the m/z 28. In the case of the Kr problem during $\delta^{13}C$ CH_4 analysis there is no other way but to get rid of the Kr in the long run using separation techniques. Correcting for this effect should be only an option for already measured data sets.

minor edits:

pg 1424, line 17 "inferred" rather than "derived" corrected

pg 1425, line 24 "an AV scan" corrected

pg 1428, line 16 "account for or eliminate" corrected

pg 1431, line 17 LN2 (it looks like lN2 at present) corrected