

## ***Interactive comment on “Preparation of primary standard mixtures for atmospheric oxygen measurements with uncertainty less than 1 ppm for oxygen mole fractions” by Nobuyuki Aoki et al.***

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We wish to express our appreciation for your significant and useful comments. We have revised the manuscript, considering your comments and suggestions.

Reviewer #2:

This paper describes an improved method for preparing synthetic gas mixtures of oxygen in artificial air by gravimetry (weighing). The use of a new mass comparator in the automatic weighing system and a thorough uncertainty evaluation allows for a suite of mixtures that have exceptionally low uncertainties. These have been verified with

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high level analytical methods that show a very good consistency within the suite and with other/previous high level standard mixtures. Nevertheless there are some principal comments and specific issues that need to be revised. I therefore recommend resubmission after major revisions. General comments The metrics and terminology lack to some extent concordance with international recommendations, standards and good practice. Even if some quantity and unit ‘habits’ are well established in atmospheric science, they are not to be taken as a role model because they are very often source of misunderstanding and misconception. Some examples are given in the following points: 1. The use of ‘mole fraction’ as a quantity denomination is depreciated and should be replaced by ‘amount (of substance) fraction’ or ‘molar fraction’. Derived quantities should be defined by quantities and not by units (mole is a unit). Angles can be defined as ‘length ratios’ and not as ‘meter ratios’. A mass fraction is not called gram fraction either. ‘Mixing ratio’ or ‘atomic weight’ are established use of quantity denominations but misleading because they mean ‘molar fraction’ and ‘atomic mass’. Further literature is ISO 80000-9, IUPAC gold book, T. Cvitas, metrologia 2003. Response: We revised from mole fraction to molar fraction in accordance with your comments 2. The use of the unit ppm for  $\mu\text{mol/mol}$  is also depreciated because it is not obvious if it is a relative or absolute unit. Please keep  $\mu\text{mol/mol}$ , it is not that long. Response: We kept  $\mu\text{mol/mol}$  in this paper in accordance with your comments

3. The definition of  $\delta$  ( $\text{O}_2/\text{N}_2$ ) in ‘per meg’ is misleading because it contains the factor 106 (equations 1 to 4). All indications in ‘per meg’ are redundant but need a mention of the standard. We would prefer to omit this notation or use it correctly. See also Coplen (DOI: 10.1002/rcm.5129) Note 7 page 2541 and Milton et al. (DOI: 10.1002/rcm.836) Response: We revised the equation 1 to 4 in accordance with your comments

The aspects of pressure dependent adsorption and desorption of analytes inside the pressurised cylinders is not discussed but may be relevant for interpreting results of certain gases (carbon dioxide). Response: We added the sentences for aspects of

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pressure dependent adsorption and desorption in this paper (P13, L14L18).

The issue of analytical interference when comparing standards to real air samples is not discussed but may also be relevant (water-issue). Response:. We added the sentences of the interferences in this paper (P12, L17-22) .

Specific comments:

Page 1, line 3: Replace mole fraction by molar fraction (throughout the text) Response:. We replace mole fraction by molar fraction.

Page 1, line 4: Correct name Matsumoto Response:. We revised the name.

Page 1, line 10: Omit per meg information in the abstract without introduction and replace ppm by  $\mu\text{mol/mol}$  Response:. We omitted per meg in the abstract without introduction and replaced ppm by  $\mu\text{mol/mol}$  according to your comment.

Page 2 line 2: omit (per meg) and ' $\times 10^6$ ' and in equations 2 to 4) Response:. We revised equation 2 to 4 according to your comment.

Page 2 line 24: use linear calibration function instead of calibration line (all instances)

Page 2 line 31: word order: ... have not yet been ... Response:. This sentence was removed.

Page 2 line 33: Replace weight measurement by mass measurement (you indicate mg which is the unit of mass and not N which would be the unit of weight (gravitational force)) Response:. We replace weight measurement by mass measurement in accordance with your comment (P3, L13)

Page 3 line 2: ... were validated ... Response:. We revised the word according to your comment.(P3, L17)

Page 3 line 26: the expression of 'gravimetric cylinder' is misleading (further instances). In fact it is the cylinder containing the gravimetrically prepared mixture. Be clear in describing the procedure. Response:. We revised the sentences of 'gravimetric cylinder'

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to 'sample cylinder' through this paper.

Page 3 line 36: ... were traced to the International ... Response:. We revised the sentence according to your comment (P4 L31)

Page 4 line 14: these may not be ratios of CO<sub>2</sub> to Ar but molar fractions? Response:. We replaced ratios of CO<sub>2</sub> to Ar by molar ratios of CO<sub>2</sub> to Ar (P4,L1-L2)

Page 6 line 8: ... factors of uncertainty... Response:. We revised the sentence according to your comment (P6, L32).

Page 6 line 24: Sentence difficult to understand. Please rephrase Response:. We revised the sentences according to your comment (P7, L14-L16)

Page 12 line 6: Why are the ratios absolute? Is there a convention to reference to AIST Response:. We revised the caption of section 5.1.

Table 1 last column: the isotope ratios should be expressed as ... =  $(x.xxx \pm y.yyy) \%$  Response:. We expressed the isotope ratios as ... =  $(x.xxx \pm y.yyy) \%$ .

Table 2 is hardly readable. Please rearrange for better reading. Response:. We rearranged the table 2

Table 5 title: The indicated numbers represent ratios not fractions Response:. We revised from fractions to ratios

Figure 5 a: The x-axis concerns also Ar/N<sub>2</sub>. Response:. We revised the x-axis according to your comment

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