Nanostructured metal oxide semiconductor-based sensors for greenhouse gas detection: progress and challenges

Yogendra K. Gautam, Kavita Sharma, Shrestha Tyagi, Anit K. Ambedkar, Manika Chaudhary and Beer Pal Singh

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Review timeline
Original submission: 11 September 2020
Revised submission: 3 February 2021
Final acceptance: 10 February 2021

Note: Reports are unedited and appear as submitted by the referee. The review history appears in chronological order.

Review History
RSOS-201324.R0 (Original submission)

Review form: Reviewer 1

Is the manuscript scientifically sound in its present form?
No

Are the interpretations and conclusions justified by the results?
No

Is the language acceptable?
No

Do you have any ethical concerns with this paper?
No

Have you any concerns about statistical analyses in this paper?
No
Recommendation?
Major revision is needed (please make suggestions in comments)

Comments to the Author(s)
Comments

The abstract
1) It presents nitrogen oxides (NOx) as a greenhouse gas together with carbon dioxide (CO2) and methane (CH4). You can't use it, because only nitrous oxide (N2O) is a greenhouse gas or other oxides of nitrogen (N0, NO2 and NO3) are not greenhouses. This generalization cannot be done. See references for reliable sources (greenhouse gases) see below:

   a) EPA – US (Environmental Protection Agency)
   https://www.epa.gov/ghgemissions/overview-greenhouse-gases
   Gases estufa:  (CO2) ,  (CH4),   (N2O) and  Fluorinated Gases

   b) (IPCC) The Intergovernmental Panel on Climate Change – ONU.
     https://www.ipcc.ch/

2) When only the title of the article is read, we have the impression of a review on greenhouse gas sensors based on metallic oxide semiconductor (MOS). But the article has lost its focus, and makes further revisions on the achievements of climate change and its solutions (renewable energies). These themes could be approached as a justification and motivation for the article. But it has an excess of pages, sometimes in a simple language but more suitable for a book than for a scientific text (article). The review (MOS) does not mention other gas detection techniques such as photoacoustic spectroscopy and chemiluminency etc ... There could be a comparison of the advantages and disadvantages with other techniques. The NO2 gas sensors (which is not a greenhouse) are widely discussed, so it is beyond the scope of the article. The greenhouse gas that is (N2O), has no comment in the text, nor reference !! .And the article still lacked comments on the part of the sensors (MOS).

3) “The article needs a new writing that defines its objectives well”

4) Nothing is said about other GHC sensors, only satellite sensing. Why ? there are many gas detection techniques . (The introduction)

5) Let's see the statements in the abstract
   a) “GHGs sensors and their sensing mechanism have been discussed briefly”
   This should be better discussed, rather than discussing details of climate change.

   b) 'The status of global GHGs emissions during the COVID-19 pandemic lockdown has also been discussed'.

Why discuss this topic in the article? I don't see this essential topic for a sensor review.

The introduction

1) This topic is about environmental sustainability, this topic is really important, but it will be for this article.
In the paragraphs below !!
The quality of life in terms of threatening water supplies, compromising ecosystems and impeding growth for future generations. The environment around us is an essential part of human survival. The more we don’t care about our environment, the more it will become intoxicated with contaminants and toxins that have a harmful impact on our lives in numerous ways.

“In particular, the pace of global economic growth during the past decades, paved the way to a continuous decline in the availability of natural resources such as forests (cut down for agriculture/demand for wood), air/land/water pollution. This in turn imposes severe health problems, damage to the productivity of land and seas, sources of oil/coal/gas, loss of fishing stocks due to overfishing and loss of species diversity etc. to the mankind. Our environment is composed of some basic components such as water, air, soil, flora, fauna, and human beings and hence economic development must move forward with the stability between these components”

2) See the statements below

a) On satellite observations as an additional or alternative tool for the detection and monitoring of anthropogenic GHGs emissions such as CO₂, CH₄, NOₓ etc.

b) NMOS-based sensors have been shown to be sensitive to a large range of GHGs, mainly to CO₂, CH₄, NOₓ and O₃.

c) It is noteworthy here that the present review centered around only on the performance analysis of available NMOS GHGs (CO₂, CH₄, NOₓ, O₃ etc.) sensors

NOₓ !!!! it’s greenhouse gas?

In the section ‘Concentration of GHGs in earth’s environment’
In the first paragraph - Look, nitrous oxide (N₂O), greenhouse gas

‘The primary GHGs in the atmosphere are water vapor, carbon dioxide, methane, nitrous oxide, and ozone’.

3) Concentração de GEEs no meio ambiente da Terra

Table 1 shows NOₓ as a greenhouse, in fact it is N₂O
The reference in Table 1 is (28) being the same reference as in Figure 2,
Both table and graph show 2000 data
Shows the figure 2. data 20 years ago. Reference below
https://commons.wikimedia.org/wiki/File:Greenhouse_gas_by_sector_2000.svg

Reference 28 is presented in the article, where the consultation was made 2020, but data are very old.

4) It has a section 3.3 MOS-based NOₓ gas sensors, entirely dedicated to NO₂ gas, which is not a greenhouse! It is a dangerous gas for human health, it generates acid rain and attacks the ozone layer. But the title of the article is sensors for greenhouse gases. Nothing is talked about sensors for N₂O gas

5) It has a section 4. Status of global GHGs emissions during the COVID-19 pandemic lockdown. The theme is important (of course), but it may not be for the central theme of the article (gas sensors)
6) The conclusions must be completely rewritten, according to the objectives of the work (focus). It is certainly not renewable energy or the consequences of climate change.

Review form: Reviewer 2 (Nitin Shelke)

Is the manuscript scientifically sound in its present form?  
Yes

Are the interpretations and conclusions justified by the results?  
Yes

Is the language acceptable?  
Yes

Do you have any ethical concerns with this paper?  
No

Have you any concerns about statistical analyses in this paper?  
No

Recommendation?  
Accept with minor revision (please list in comments)

Comments to the Author(s)  
The manuscript can be accept with minor revision.

Decision letter (RSOS-201324.R0)

The editorial office reopened on 4 January 2021. We are working hard to catch up after the festive break. If you need advice or an extension to a deadline, please do not hesitate to let us know – we will continue to be as flexible as possible to accommodate the changing COVID situation. We wish you a happy New Year, and hope 2021 proves to be a better year for everyone.

Dear Dr Sharma:

Title: Nanostructured Metal Oxide Semiconductor-based Sensors for Greenhouse Gas Detection: Progress and Challenges  
Manuscript ID: RSOS-201324

Thank you for your submission to Royal Society Open Science. The chemistry content of Royal Society Open Science is published in collaboration with the Royal Society of Chemistry.

The editor assigned to your manuscript has now received comments from reviewers. We would like you to revise your paper in accordance with the referee and Subject Editor suggestions which can be found below (not including confidential reports to the Editor). Please note this decision does not guarantee eventual acceptance.
Please submit your revised paper before 31-Jan-2021. Please note that the revision deadline will expire at 00.00am on this date. If we do not hear from you within this time then it will be assumed that the paper has been withdrawn. In exceptional circumstances, extensions may be possible if agreed with the Editorial Office in advance. We do not allow multiple rounds of revision so we urge you to make every effort to fully address all of the comments at this stage. If deemed necessary by the Editors, your manuscript will be sent back to one or more of the original reviewers for assessment. If the original reviewers are not available we may invite new reviewers.

To revise your manuscript, log into http://mc.manuscriptcentral.com/rsos and enter your Author Centre, where you will find your manuscript title listed under "Manuscripts with Decisions." Under "Actions," click on "Create a Revision." Your manuscript number has been appended to denote a revision. Revise your manuscript and upload a new version through your Author Centre.

When submitting your revised manuscript, you must respond to the comments made by the referees and upload a file "Response to Referees" in "Section 6 - File Upload". Please use this to document how you have responded to the comments, and the adjustments you have made. In order to expedite the processing of the revised manuscript, please be as specific as possible in your response.

Once again, thank you for submitting your manuscript to Royal Society Open Science and I look forward to receiving your revision. If you have any questions at all, please do not hesitate to get in touch.

Yours sincerely,
Dr Laura Smith
Publishing Editor, Journals

Royal Society of Chemistry
Thomas Graham House
Science Park, Milton Road
Cambridge, CB4 0WF
Royal Society Open Science - Chemistry Editorial Office

On behalf of the Subject Editor Professor Anthony Stace and the Associate Editor Dr Dattatray Late.

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RSC Associate Editor:
Comments to the Author:
Major Revision needed.

RSC Subject Editor:
Comments to the Author:
(There are no comments.)

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Reviewers' Comments to Author:
Reviewer: 1
Comments to the Author(s)
Comments
The abstract
1) It presents nitrogen oxides (NOx) as a greenhouse gas together with carbon dioxide (CO2) and methane (CH4). You can't use it, because only nitrous oxide (N2O) is a greenhouse gas or other oxides of nitrogen (N0, NO2 and NO3) are not greenhouses. This generalization cannot be done. See references for reliable sources (greenhouse gases) see below:

a) EPA – US (Environmental Protection Agency)
https://www.epa.gov/ghgemissions/overview-greenhouse-gases
Gases estufa: (CO2), (CH4), (N2O) and Fluorinated Gases

b) (IPCC) The Intergovernmental Panel on Climate Change – ONU.
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2) When only the title of the article is read, we have the impression of a review on greenhouse gas sensors based on metallic oxide semiconductor (MOS).
But the article has lost its focus, and makes further revisions on the achievements of climate change and its solutions (renewable energies). These themes could be approached as a justification and motivation for the article. But it has an excess of pages, sometimes in a simple language but more suitable for a book than for a scientific text (article).
The review (MOS) does not mention other gas detection techniques such as photoacoustic spectroscopy and chemiluminescence etc... There could be a comparison of the advantages and disadvantages with other techniques. The NO2 gas sensors (which is not a greenhouse) are widely discussed, so it is beyond the scope of the article. The greenhouse gas that is (N2O), has no comment in the text, nor reference !! And the article still lacked comments on the part of the sensors (MOS).

3) “The article needs a new writing that defines its objectives well”

4) Nothing is said about other GHC sensors, only satellite sensing. Why ? there are many gas detection techniques . (The introduction)

5) Let's see the statements in the abstract
a) “GHGs sensors and their sensing mechanism have been discussed briefly”
This should be better discussed, rather than discussing details of climate change.

b) 'The status of global GHGs emissions during the COVID-19 pandemic lockdown has also been discussed'.

Why discuss this topic in the article? I don't see this essential topic for a sensor review.

The introduction
1) This topic is about environmental sustainability, this topic is really important, but it will be for this article.
In the paragraphs below !!
- quality of life in terms of threatening water supplies, compromising ecosystems and impeding growth for future generations. The environment around us is an essential part of human survival.
The more we don’t care about our environment, the more it will become intoxicated with contaminants and toxins that have a harmful impact on our lives in numerous ways
- “In particular, the pace of global economic growth during the past decades, paved the way to a continuous decline in the availability of natural resources such as forests (cut down for agriculture/demand for wood), air/land/water pollution. This in turn imposes severe health problems, damage to the productivity of land and seas, sources of oil/coal/gas, loss of fishing stocks due to overfishing and loss of species diversity etc. to the mankind. Our environment is composed of some basic components such as water, air, soil, flora, fauna, and human beings and hence economic development must move forward with the stability between these components.”

2) See the statements below

a) On satellite observations as a new additional or alternative tool for the detection and monitoring of anthropogenic GHGs emissions such as CO2, CH4, NOx etc

b) NMOS-based sensors have been shown to be sensitive to a large range of GHGs, mainly to CO2, CH4, NOx and O3,

c) It is noteworthy here that the present review centered around only on the performance analysis of available NMOS GHGs (CO2, CH4, NOx, O3 etc.) sensors

NOx!!! It’s greenhouse gas?

In the section ‘Concentration of GHGs in earth’s environment’
In the first paragraph - Look, nitrous oxide (N2O), greenhouse gas

‘The primary GHGs in the atmosphere are water vapor, carbon dioxide, methane, nitrous oxide, and ozone’.  

3) Concentração de GEEs no meio ambiente da Terra

Table 1 shows NOx as a greenhouse, in fact it is N2O
The reference in Table 1 is (28) being the same reference as in Figure 2, Both table and graph show 2000 data
Shows the figure 2. data 20 years ago. Reference below
https://commons.wikimedia.org/wiki/File:Greenhouse_gas_by_sector_2000.svg

Reference 28 is presented in the article, where the consultation was made 2020, but data are very old.

4) It has a section 3.3 MOS-based NOx gas sensors, entirely dedicated to NO2 gas, which is not a greenhouse! It is a dangerous gas for human health, it generates acid rain and attacks the ozone layer. But the title of the article is sensors for greenhouse gases. Nothing is talked about sensors for N2O gas

5) It has a section 4. Status of global GHGs emissions during the COVID-19 pandemic lockdown. The theme is important (of course), but it may not be for the central theme of the article (gas sensors)

6) The conclusions must be completely rewritten, according to the objectives of the work (focus). It is certainly not renewable energy or the consequences of climate change.
Reviewer: 2

Comments to the Author(s)
The manuscript can be accept with minor revision.

Author's Response to Decision Letter for (RSOS-201324.R0)

See Appendix A.

Decision letter (RSOS-201324.R1)

We hope you are keeping well at this difficult and unusual time. We continue to value your support of the journal in these challenging circumstances. If Royal Society Open Science can assist you at all, please don't hesitate to let us know at the email address below.

Dear Dr Sharma:

Title: Nanostructured Metal Oxide Semiconductor-based Sensors for Greenhouse Gas Detection: Progress and Challenges
Manuscript ID: RSOS-201324.R1

It is a pleasure to accept your manuscript in its current form for publication in Royal Society Open Science. The chemistry content of Royal Society Open Science is published in collaboration with the Royal Society of Chemistry.

The comments of the reviewer(s) who reviewed your manuscript are included at the end of this email.

Please see the Royal Society Publishing guidance on how you may share your accepted author manuscript at https://royalsociety.org/journals/ethics-policies/media-embargo/. After publication, some additional ways to effectively promote your article can also be found here https://royalsociety.org/blog/2020/07/promoting-your-latest-paper-and-tracking-your-results/.

Thank you for your fine contribution. On behalf of the Editors of Royal Society Open Science and the Royal Society of Chemistry, I look forward to your continued contributions to the Journal.

Yours sincerely,
Dr Laura Smith
Publishing Editor, Journals

Royal Society of Chemistry
Thomas Graham House
Science Park, Milton Road
Cambridge, CB4 0WF
Royal Society Open Science - Chemistry Editorial Office
On behalf of the Subject Editor Professor Anthony Stace and the Associate Editor Dr Dattatray Late.

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RSC Associate Editor
Comments to the Author:
Accept as is

*******

Reviewer(s)' Comments to Author:
Response letter to Referees” for the manuscript ID: RSOS-201324 “Nanostructured Metal Oxide Semiconductor-based Sensors for Greenhouse Gas Detection: Progress and Challenges”

Dear Editor,

We are greatly thankful to you and reviewers for the valuable comments and suggestions on our research article to improve the quality of manuscript entitled “Nanostructured Metal Oxide Semiconductor-based Sensors for Greenhouse Gas Detection: Progress and Challenges” (RSOS-201324). We have revised the manuscript in the light of reviewer’s comments/suggestions. Kindly, find the attached revised manuscript for your consideration. All the modified/revised part has marked in yellow in the revised manuscript. The point-wise clarifications/respond to the comments made by the referees are given below. We hope this will also be acceptable to the reviewer and editor.

With kind regards,

Dr. Kavita Sharma
Chaudhary Charan Singh University
Meerut, India

Response to reviewer comment

Reviewer: 1 (Major Revision)

The abstract

Comment 1: It presents nitrogen oxides (NOx) as a greenhouse gas together with carbon dioxide (CO2) and methane (CH4). You can't use it, because only nitrous oxide (N2O) is a greenhouse gas or other oxides of nitrogen (NO, NO2 and NO3) are not greenhouses. This generalization cannot be done.

See references for reliable sources (greenhouse gases) see below:

a) EPA – US (Environmental Protection Agency)
https://www.epa.gov/ghgemissions/overview-greenhouse-gases
Gases estufa: (CO2), (CH4), (N2O) and Fluorinated Gases

b) (IPCC) The Intergovernmental Panel on Climate Change – ONU.
https://www.ipcc.ch/

Response: We thank the reviewer for raising this important comment on (NOx). We completely agree with the reviewer. In the revised manuscript, we have replaced NOx with N2O as per the suggestion of learned reviewer through the links suggested in a) and b) and also cite the link in reference list. Now the greenhouse gases referred in revised manuscript are; CO2, CH4, N2O and O3.

Comment 2: When only the title of the article is read, we have the impression of a review on greenhouse gas sensors based on metallic oxide semiconductor (MOS).

But the article has lost its focus, and makes further revisions on the achievements of climate change and its solutions (renewable energies). These themes could be approached as a
justification and motivation for the article. But it has an excess of pages, sometimes in a simple language but more suitable for a book than for a scientific text (article).

The review (MOS) does not mention other gas detection techniques such as photoacoustic spectroscopy and chemiluminescence etc. There could be a comparison of the advantages and disadvantages with other techniques. The NO\textsubscript{2} gas sensor (which is not a greenhouse) are widely discussed, so it is beyond the scope of the article. The greenhouse gas that is (N\textsubscript{2}O), has no comment in the text, nor reference !!. And the article still lacked comments on the part of the sensors (MOS).

**Response:** We agree with the reviewer. We have removed the excess of pages on the achievements of climate change and its solutions (renewable energies) in the revised manuscript accordingly.

As per reviewer’ suggestion, authors have mentioned other gas detection techniques such as photoacoustic spectroscopy, chemiluminescence, electrochemical sensor, thermometric sensors, catalytic sensor, gas chromatography, chemical sensor and mass resistive sensor in the revised manuscript.

Authors have given a comparison of the advantages and disadvantages of MOS-based technique with other techniques as per the suggestion of reviewer and highlighted by yellow colour in Table-2 of the revised manuscript.

Authors are very much thankful to the reviewer for the comment on the NO\textsubscript{2} gas sensors. We agree with the reviewer. As NO\textsubscript{2} is not a greenhouse gas. Therefore, authors have removed the given content for NO\textsubscript{2} gas sensors.

In the revised manuscript, authors have included a wide discussion about the various studies/reports available on MOS-based sensors for N\textsubscript{2}O (greenhouse gas). Authors have also introduced the other recent available techniques for N\textsubscript{2}O detection as per the reviewer suggestion/expectation.

**Comment 3:** “The article needs a new writing that defines its objectives well”

**Response:** We are agreeing with the reviewer. In the revised manuscript, authors have rewritten the objectives of this review and highlighted that with yellow colour.

**Comment 4:** Nothing is said about other GHC sensors, only satellite sensing. Why? there are many gas detection techniques. (The introduction)

**Response:** Although satellite sensing is an emerging interest of climate scientists in order to have a wide spatial and temporal coverage of GHGs emission globally. But authors agree completely with the reviewer’s concern on the theme of this review and therefore authors have removed satellite sensing paragraph in the introduction of revised manuscript. Instead other GHG gas sensors have been included in introduction of revised manuscript and highlighted in yellow colour.

**Comment 5:** Let's see the statements in the abstract;

a) “GHGs sensors and their sensing mechanism have been discussed briefly”

This should be better discussed, rather than discussing details of climate change.

**Response:** We agree with the reviewer. In the revised manuscript, authors have rewritten the abstract as per the suggestion of the reviewer and highlight the rewrite part with yellow color.

b)“The status of global GHGs emissions during the COVID-19 pandemic lockdown has also
been discussed’. Why discuss this topic in the article? I don't see this essential topic for a sensor review.

**Response:** We agree with the reviewer. Authors have discussed the status of global GHGs emissions during the COVID-19 pandemic lockdown because of the interest and motivation followed by the reduction of global GHGs emissions level. However, authors have removed this section 4 in the revised manuscript as per the reviewer suggestion.

**Reviewer: 2 (Minor revision)**
Comments to the Author(s)

The introduction

**Comment 1:** This topic is about environmental sustainability, this topic is really important, but it will be for this article.

In the paragraphs below !!

- quality of life in terms of threatening water supplies, compromising ecosystems and impeding growth for future generations. The environment around us is an essential part of human survival. The more we don’t care about our environment, the more it will become intoxicated with contaminants and toxins that have a harmful impact on our lives in numerous ways

- “In particular, the pace of global economic growth during the past decades, paved the way to a continuous decline in the availability of natural resources such as forests (cut down for agriculture/demand for wood), air/land/water pollution. This in turn imposes severe health problems, damage to the productivity of land and seas, sources of oil/coal/gas, loss of fishing stocks due to overfishing and loss of species diversity etc. to the mankind. Our environment is composed of some basic components such as water, air, soil, flora, fauna, and human beings and hence economic development must move forward with the stability between these components”.

**Response:** We agree with the reviewer. In the revised manuscript, authors have removed all the above said statements/paragraphs about the environment sustainability and modified part of introduction is highlighted by yellow colour as per the reviewer’s suggestion.

**Comment 2:** See the statements below;

a) On satellite observations as an additional or alternative tool for the detection and monitoring of anthropogenic GHGs emissions such as CO2, CH4, NOx etc

b) NMOS-based sensors have been shown to be sensitive to a large range of GHGs, mainly to CO2, CH4, NOx and O3,

c) It is noteworthy here that the present review centred around only on the performance analysis of available NMOS GHGs (CO2, CH4, NOx, O3 etc.) sensors

NOx !!!! it's greenhouse gas?
In the section ‘Concentration of GHGs in earth’s environment”
In the first paragraph - Look, nitrous oxide (N2O), greenhouse gas

‘The primary GHGs in the atmosphere are water vapor, carbon dioxide, methane, nitrous oxide, and ozone’.

Response: We agree with the reviewer as N2O is a greenhouse gas. In the revised manuscript, authors have modified all the above said statements/paragraphs as per the reviewer suggestions.

Comment 3: Concentração de GEEs no meioambiente da Terra

Table 1 shows NOx as a greenhouse, in fact it is N2O
The reference in Table 1 is (28) being the same reference as in Figure 2,
Both table and graph show 2000 data
Shows the figure 2. data 20 years ago. Reference below
https://commons.wikimedia.org/wiki/File:Greenhouse_gas_by_sector_2000.svg

Reference 28 is presented in the article, where the consultation was made 2020, but data are very old.

Response: We agree with the reviewer. We have mentioned N2O in place of NOx in Table-1, in the revised manuscript as per reviewer suggestions.
Authors are very much thankful to the reviewer for the valuable comment on the reference (28) (reference no. 20 in the revised manuscript) in Table 1 and in Figure 2.
In the revised manuscript, we have modified the Table 1 and Figure 2 as per the data available in the references [(a)EPA – US (Environmental Protection Agency) https://www.epa.gov/ghgemissions/overview-greenhouse-gasesGases estufa: (CO2), (CH4), (N2O) and Fluorinated Gases and (b) (IPCC) The Intergovernmental Panel on Climate Change – ONU,https://www.ipcc.ch/] suggested by reviewer 1. Authors also included the suggested reference in the reference list. Please note that all changes have been highlighted by yellow colour in revised manuscript.

Comment 4: It has a section 3.3 MOS-based NOx gas sensors, entirely dedicated to NO2 gas, which is not a greenhouse! It is a dangerous gas for human health, it generates acid rain and attacks the ozone layer. But the title of the article is sensors for greenhouse gases. Nothing is talked about sensors for N2O gas.

Response: We agree with the reviewer. In the revised manuscript, authors have replaced complete section 3.3: MOS-based NOx gas sensors with MOS-based N2O gas sensors as suggested by the reviewer.

Comment 5: It has a section 4. Status of global GHGs emissions during the COVID-19 pandemic lockdown. The theme is important (of course), but it may not be for the central theme of the article (gas sensors).

Response: We agree with the reviewer. Authors have removed the “section 4: Status of global GHGs emissions during the COVID-19 pandemic lockdown” in the revised manuscript accordingly.

Comment 6: The conclusions must be, according to the objectives of the work (focus). It is certainly not renewable energy or the consequences of climate change.
**Response:** We are agreeing with the reviewer. In the revised manuscript, authors have completely rewritten the conclusions according to the objectives of the work as per reviewer suggestion.

In the last, authors are very much thankful to the reviewers for critically reviewing our review article and accepting the manuscript for publication with the major/minor revision.

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