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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Gaseous VOCs rapidly modify particulate matter and its biological effects – Part 1: Simple VOCs and model PM" *by* S. Ebersviller et al.

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Anonymous Referee #1 Received and published: 20 April 2012 Review comments for "Gaseous VOCs rapidly modify particulate matter and its biological effects – Part 1: Simple VOCs and model PM" by Ebersviller et al.

Referee's Comment: The manuscript describes an experimental set up to investigate the toxicological effects of particles, gases and mixtures of the two in-vitro experiments. Especially results of experiments looking at the combined particle/gas effect is novel and interesting. The concept of the experiment design is described repetitively. I suggest that the authors delete some of these paragraphs.

Author's Reply: Thank you for your comment on being novel and interesting. With regard to the conceptual experimental design, we have weighted the response that knowledgeable researchers might have against the need to remind some of our target audience about the complex interplays in our test system (see further comments about this below). In response, however, we have revised the text and removed some material that may have been considered repetitious. Other reviewers have called for additions especially in the area of how the biological samplers work. Altogether the manuscript remains about the same length.

Referee's Comment: p. 5067/5077 (i): It is mentioned that in the GIVES system no particle effects are observed. It would be nice to see some evidence supporting this statement, e.g., particle deposition characteristics, possibly from earlier publications. A detailed schematic of the GIVES instrument might be clarifying.

Author's Reply: Based on both Referees' comments, it is clear that we should have provided more information in the reviewed manuscript about our unusual biological exposure systems. In the reviewed manuscript, we cited three published papers in which the GIVES sampler had been previously described and used; in the revised manuscript we have added additional citations. The GIVES is a commercial device and we had provided the manufacturer and model number. We also gave a six-line brief description and a one-line explanation of why PM does not cause an exposure in the device. The second referee also asked for more information in the revised manuscript about the two samplers. In response, we have expanded the GIVES description (Section 2.6.1) by reporting calculations and including a citation for terminal settling velocities of mass-mode-diameter PM in the chamber (2.86x10ˆ(-3) cm sˆ(-1) for 1 μm particles, of which there were about 82 particles per cmˆ3 in our chamber tests). Applying various simplifying assumptions we have estimated how long it would take in the GIVES to give a deposition similar to that in the EAVES PM sampler. Depending upon the assumptions, this time varies from 12 hours to 150 days. The former requires that all particles that entered the GIVES be deposited uniformly across the footprint of the exposure cham-

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ber, which of course, does not ever happen. Thus, cells in the GIVES do have some exposure to PM, but the extent is so limited that it cannot be detected by biomarker changes, making this sampler 'virtually' a gas-only sampler.

As an observational point, if significant PM settling did occur in the GIVES, the GIVES responses for Exp. D vs. Exp. C in Fig. 4 would not have declined as TOLALD moved from gas-phase to PM (as shown by the large increase in responses in the EAVES). If the cells in GIVES were responding to the PM-borne TOLALD, we would expect to have seen the same (or nearly the same) response from both TOLALD mixtures in the GIVES.

Referee's Comment: p. 5067/5077 (ii): Similarly, it is not clear why there should be no gas phase effects in the EAVES system. There seems no gas/particle separation in place for the EAVES. Thus the cell cultures in the EAVES are constantly exposed to the gas phase as well. Aerosol (gas and particles) is constantly pumped through the EAVES and it seems a similar gas response as in the GIVES should be observed. Exposure times in the EAVES are shorter than in the GIVES but effects in the GIVES are quite pronounced suggesting that also for the shorter exposure time in the EAVES gas phase effects should be observed.

Author's Reply: Both referees raised these issues. While we did cite peer-reviewed publications in which these findings have been described, discussed, and supported, it is clear that this manuscript would be improved by adding a brief summary of the operational principles and previous test findings. This was done in Section 2.6.2 in which we added a brief paragraph describing the physical and flow operations of the EAVES sampler and its operational environment. This is followed by an 18-line summary of the published peer-reviewed findings for the operational characteristics. We also added statements that clarified that cells in the EAVES do have some exposure to the sample gas, but it has been repeatedly demonstrated experimentallyâĂŤincluding in this study's results – that such exposure is so limited that the cells show no detectable response to gases for the 1-h exposure time and unique conditions of the

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sampler. These outcomes make this sampler 'virtually' a PM-only exposure device. Further, in the largely revised Section 4, we have identified comparisons of the experimental graphical results in this manuscript where this operational feature of the EAVES is clearly evident (in the discussions of both the TOLALD and ACRO results).

âĂČ Referee's Comment: p. 5079, line 21: No VOCs are observed in the clean air experiments. What was the detection limit for the VOC analysis method?

Author's Reply: The reviewed manuscript did state that the chamber is flushed and filled with the output of a clean air generator. We have added additional details on the commercial Addco Clean Air Generator (Model 737, 250 L minˆ(-1)) which was designed specifically to remove VOCs by high capacity reverse-flush absorption. A conservative average of the limit of detection for the GCMS used to measure the chamber background is between 1-3 ppbV. This has been added to the manuscript in Section 2.4 and to the footnote on Table 1.

Referee's Comment: p. 5083. Line 17-22: How is the pronounced LDH release effect explained considering that tolualdehyde is not toxic, as mentioned on line 17? The authors mention that the effects of the particle addition to the system with respect to the GIVES and EAVES results are unexpected. What could have caused this unexpected and very pronounced result, especially the essential disappearance of the gas phase toxicity of tolualdehyde and partly also of acrolein?

Author's Reply: Thank you for pointing out this discrepancy. This was a mistake. The toxicity referred to in the reviewed manuscript was for ingestion of small quantities, not inhalation of a gas. As a gas, tolualdehyde is a known respiratory irritant, producing a burning sensation, bronchial constriction, choking and coughing as reported in "Patty's Industrial Hygiene and Toxicology", 1982 and cited as a peer-reviewed source on the US National Institute of Health's Toxnet web site (http://toxnet.nlm.nih.gov). This information has been added to the revised manuscript in Section 2.3 "Choices of gases and particles to test", and a citation to Patty's has been included. It was inappropri-

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ate to describe the results as “unexpected”. Reorganizing the Discussion (section 4) and explicitly addressing the relative changes in biological responses in the different exposures have addressed the second part of the comment.

Referee’s Comment: The authors emphasize repetitively in the manuscript that the addition of tolualdehyde and acrolein to the mineral oil aerosol changes the composition of the mineral oil particles and consequently their toxicity. From an atmospheric chemistry point of view it seems obvious that a change in the organic gas phase composition also changes the composition of the particle phase. The surprising result of this manuscript is that the very small overall amount of tolualdehyde and acrolein absorbed by the mineral oil particles causes such a pronounced effect. As mentioned above this would deserve more discussion, also considering the instrumental questions raised above.

Author’s Reply: Our experience has been that many toxicologists, health effect researchers, and regulatory policymakers have minimal understanding of gas-particle phenomena that are quite familiar to the physics and chemistry community. See, for example, in the reviewed manuscript’s “Abstract” and “Conclusion” sections the citation of the US National Academy of Science report on “Research Priorities for Airborne Particulate Matter,” which calls for evidence of "effects modification" for PM. The quote from that citation is:

“A finding that the effect of particles depends on the concentration of another pollutant – that is, ‘effect modification’ – would have implications for setting NAAQS independently for the various criteria pollutants” Research Priorities for Airborne Particulate Matter, NAS, 2004, p. 99

Clearly they do not think that this is currently true, but call for research.

Because part of our target audience are those conducting biological effects research and regulatory policymakers need to cite peer-reviewed work, the need to emphasize that the atmosphere itself can be a source of PM toxicity is justified here. What seems so rudimentary to one community can be a significant surprise to another.

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To the second point, as we discussed in the reviewed manuscript's Introduction (section 1), PM has great potential to deliver certain types of VOC to the air-liquid interface of cells, where it is able to bypass the diffusion-limited transfer across the aqueous layer above the surface of the cells. Our results demonstrate the power of even 'non-toxic' PM to deliver a significant dose of what was an airborne toxicant to the cells. As a gas, such oxygenated toxicants are mostly removed in humans in the nasal and upper airways, but when taken up on PM, they can be transported into deep areas of the lung and, because of equilibrium considerations, be 'off-gassed' and be delivered much more systemically. In our in vitro exposures, the PM on the cell surface can sustain a much longer exposure than the mere encounter of a few gas molecules would be able to give.

The additions and clarifications made to the sampling and exposure sections of the reviewed manuscript have addressed any issues over what the two exposure methods are responding to and our reorganization of the Discussion section following a step-by-step evaluation of our hypothesis should give the reviewer more confidence in our findings.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/12/C5596/2012/acpd-12-C5596-2012-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 5065, 2012.