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Interactive Comment

Interactive comment on "Effects of methane outgassing on the Black Sea atmosphere" by K. Kourtidis et al.

K. Kourtidis et al.

Received and published: 4 July 2006

Response to comments by Referee #1

Generally, the comments are well-founded and contribute to the improvement of the manuscript. Hence, all will be taken into account in the revised version. We give below in [brackets] the text that will be included in the revised version.

1. The referee makes a justified comment, namely that wind speed measurements are not presented. In the revised version, wind speed will be included in Figure 1 as Fig. 1c:

[Figure 1c (revised). Wind speed measurements during the BIGBLACK cruise. Shaded areas denote ship's location, while areas with darker shading indicate measurements where vessel was anchored directly above active seeps. Areas denoted "Seep" are

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areas with active seeps.]

2. Another comment is that the figures 1 and 2 are difficult to judge without reading the text; hence, indications on the ship's location will be included in Figs. 1 to 3. (as in Fig. 1c above).

3. ppt unit: It is almost a "standard" notation in atmospheric science, namely ppt is parts per trillion per volume. Since, though, it appears this may not apply to some disciplines, to avoid confusion we will explain the notation in the figure caption: [units are in parts per trillion per volume]. Also, ppt will be changed to pptv in the y axis of the figure.

4. Derivation of dispersion model input of 6.25 mmol/m2/s in an area 100X100 m2:

At the time of the first submission, as stated in lines 20-29 of p. 3618, we started from a subsurface (at 2 km depth) emission rate of 22.5 million m3 gas, 90% CH4. This is in the lowest range of the amount of gas expelled during the first few hours from four prominent eruptions (1902-1961), 22.5-495 million m3, (Guliyev and Feizullayev, 1996, quoted extensively in the peer-reviewed literature, e.g. in Dimitrov, 2002; also Guliev, 1992, p. 7, quoted in Kopf, 2002, see also below). Guliev (1992) has performed the only extensive study on quantitative eruptive methane emissions from mud volcanoes. Clearly, our input has been conservative. Then, we used preliminary results from work done in the CRIMEA project on bubble-water column exchange of gases to assume that an upper limit of 0.1 % of the emitted methane would reach the surface, hence arriving at the 6.25 mmol/m2/s value for emission rate at the surface (at a 100 X 100 m2 rectangle). Since then, however, recent calculations by another group in the frame of the CRIMEA project, taking also into account newer findings about the formation of a bubble hydrate skin, have updated this calculation, and indicate that for a substantially large eruption (larger than the one we simulated in the earlier manuscript version) a much larger percentage of the emitted gas reaches the surface (see below for details). Hence we have performed again our calculations using these recent bubble model 6, S1524–S1531, 2006

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results, and the following text will be added in the relevant discussion of the revised manuscript (taking also comments from reviewer #2 into account), while figure 5 will be updated accordingly:

[Guliev (1992), p. 7, quoted extensively, e.g. in Kopf (2002) and Guliyev and Feizullayev (1996), also quoted extensively in the western peer-reviewed literature, e.g. in Dimitrov (2002), states that for four prominent mud volcano eruptions between 1902-1961 (on land) the amount of gas expelled during the first few hours ranged from 22.5-495 million m3. If we assume a 4-hour, constant rate explosion, the upper estimate of 495 million m3 translates to 24,500,000 g/s, if the gas is 100% methane. For 70% methane this would still be 17,150,000 g/s.

The amounts of methane that might rise to the atmosphere from a catastrophic methane release at depth have been estimated in the frame of the CRIMEA project (McGinnis et al., 2006) through a modified version of the Wuest et al. (1992) plume model. Some aspects of the model have been treated in McGinnis and Little (2002). The initial conditions of the model and the results are given in Table 1. It seems it would require a release of about 16,000,000 g/s (Scenario I) of methane in gas bubble form to reach the surface from 2000m. Of this, only roughly 30% reaches the atmosphere in both gaseous and dissolved form (50% gas, 50% dissolved). If we use 1,600,000 g/s then no methane reaches the surface.

On the other hand, simulating a mud volcano gas release of 24,500,000 g/s (100% methane) at 2 km depth, all of the methane reaches the surface (about 20% gas, and 80% dissolved) (McGinnis, personal communication). The plume water when it reaches the surface is much denser, so it is difficult to estimate with certainty how much it will degass the dissolved fraction before settling back to the equilibrium depth. These simulations assume that a hydrate skin exists on the bubble in the stability zone (see, e.g., Rehder et al., 2002; Sauter et al., 2006). If no hydrate skin exists on the bubble, then the plume does not reach the surface.

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Table 1 - Modelling results for two hypothetical scenarios of catastrophic mud volcano outbursts.

Initial Conditions (Source) Scenario I Scenario II

Initial plume radius (m) 100 100

Bubble diameter (mm) 8 8

Methane flux (mol/s) 1,000,000 1,531,000

Methane flux (kg/s) 16,000 24,496

Depth (m) 2,000 2,000

Results

Flux methane in bubbles (mol/s) 143,600 319,073

Flux dissolved methane (mol/s) 158,400 1,218,613

Even the most intensive Black Sea bubble seep that was studied within the BIGBLACK and CRIMEA projects does not transfer methane into the atmosphere through bubble transport. Only at 90 m some of the methane can survive in the bubbles to reach the atmosphere, but this is very minor. At larger depths, all methane is stripped from the bubbles long before they reach the surface. The same holds also for a mud volcano (M/V Dvurechenskiy) gas eruption that was monitored during the CRIMEA project. So, in order to cause a significant methane input into the atmosphere, much more violent eruptions are needed. This seems rather unlikely for the shallower seep areas (100, 250, 600 m) given the nature of the seeping process there. On the other hand, it does not seem impossible for a really large mud volcano eruption to generate a much bigger methane bubble flux, with bigger bubbles and possibly even creating a bubble plume, which could eventually make it up to the atmosphere.

Results from the AERMOD atmospheric dispersion model in the case of a 4-hour ex-

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plosion of a constant rate of 16,000,000 g/s gas at 2,000 m depth are presented. As mentioned above, bubble modelling estimates that 30% of the emitted gas reaches the atmosphere. The equivalent emission rate to the atmosphere would then be 2,400,000 g/s (here we assume that the direct bubble transfer is more efficient than the diffusion of dissolved methane, and hence neglect the latter in the calculation). Assuming the release takes place from an area source 100m x 100m, the maximum increases in the atmospheric levels of methane during the 4-hour eruption as estimated through dispersion modelling (ISC-AERMOD; The et al., 2002), are given in Figure 5.

Figure 5 (revised). Calculated increases in the atmospheric background of methane for the case of a catastrophic eruption under different atmospheric conditions. The wind speed used in the calculations was 3m/s, 5 m/s and 4 m/s, for stability classes A, D and F, respectively.

The plume dispersion modelling results show that the spatial average of the methane perturbation during the eruption over a square receptor area with dimension 100km x 100km centered in the source (assuming a 4h release) is approximately 4 ppmv for unstable conditions (A), 10 ppmv for neutral conditions (D) and 20 ppmv for stable conditions (F), which represent increases of the average background methane mixing ratio of 1.86 ppmv over the Black Sea of 315%, 640% and 1175%, respectively.

Similarly, the spatial 24h-average of the methane perturbation over a square receptor area with dimension 100km x 100km centered in the source (also assuming a 4h release) is approximately 0.7 ppm for unstable conditions (A), 2.7 ppm for neutral conditions (D) and 6.5 ppm for stable conditions (F). The wind velocity field was 3, 8 and 12 (m/s) for the stability classes A, D and F, respectively (the perturbation amount will be generally higher for lighter winds). Hence, given an average background methane mixing ratio of 1.86 ppmv over the Black Sea, the calculated increase (%) of the 24-hr average mixing ratio of methane over the 100 km X 100 km area ranges from 35% for unstable conditions to 350% for stable conditions.]

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5. Plume dispersion model description: The following text will be added in the revised version:

[The ISC-AERMOD (The et al., 2002) steady-state Gaussian plume dispersion model from Lake Environmental Inc. is based on US Environmental Protection Agency's ISCST3 model (EPA, 1995). It calculates ground-level and aloft inert ambient pollutant concentrations and/or deposition fluxes, for continuous and/or accidental releases (per hour). The model can handle multiple sources including point, volume, area, open pit as well as line source types. Source emission rates can be treated as constant or may be varied by month, season, hour-of-day, or other optional periods of variation. These variable emission rate factors may be specified for a single source or for a group of sources. In addition, the underlying ISCST3 model includes several options for addressing complicated problems like site-specific wind profile exponents, vertical potential temperature gradients, time-dependent exponential decay of emissions, plume rise calculated as a function of downwind distance, etc.]

6. Unignited methane simulations: The following text will be added in the revised version:

[Several authors report self-ignition of the expelled gas from mud volcanoes (Kugler, 1939; Bagirov et al., 1996a,b; Aliyev et al., 2002; Jevanshir, 2002), even for underwater eruptions (Sokolov, 1969). However, whether self-ignition is common in underwater eruptions and under which circumstances it might occur in nature, are, to our knowledge, not well documented, hence no simulations for ignited emission were performed.]

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