Atmos. Chem. Phys. Discuss., 11, C13192–C13201, 2011
www.atmos-chem-phys-discuss.net/11/C13192/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Xenon-133 and caesium-137 releases into the atmosphere from the Fukushima Dai-ichi nuclear power plant: determination of the source term, atmospheric dispersion, and deposition" *by* A. Stohl et al.

G. Grasso

giacomo.grasso@enea.it

Received and published: 14 December 2011

Engineering aspects related to the paper by A. Stohl et al. – 2nd Reply

M. Di Giuli, G. Grasso, D. Mattioli, F. Padoani, R. Pergreffi and F. Rocchi

Technical Unit for Reactor Safety and Fuel Cycle Methods (UTFISSM) – Italian National Agency for New Technologies, Energy and Sustainable Economic Development (ENEA)
v. Martiri di Monte Sole, 4, 40129 Bologna (ITALY)

Dear Colleagues,

we want to thank you for your replies to some of our comments: we find this interesting discussion fruitful, and we agree with you that it's possible only because of the open peer review system. We understand that media may have used some of your preliminary conclusions without proper and due caution, nonetheless we think that at least in the final version of your paper it could be helpful if you provide clear statements not only about the robustness of your calculation schemes, but also and most importantly about the uncertainties related to your transport and deposition models.
We still would like to add some remarks and observations concerning the conclusions of your tentative reconstruction of the accident emissions. For clarity's sake, we follow the individual points you put forward both in the reply to us and to Mr. Horst-Michael Prasser.

1. *Xe-133*. Even if you adapt your calculations to show a discrepancy above the maximum physical inventory of the entire Dai-ichi site of 1 EBq or less, this discrepancy is still a huge quantity of activity, and not a small amount that "might well have escaped the general attention". Firstly we want to stress that every NPP is built in order to adopt the most effective and advanced techniques and strategies for the physical protection of the workers and the people living in the hood of the plant. Saying that even small amounts of radioactivity might escape from a plant without being revealed contradicts the philosophy of nuclear safety itself, which is an assert that can be hardly shared. Entering the detail, you state that "it is not entirely clear whether on-site radiation monitors at other NPPs would have captured such releases"; if you refer to emissions originated from NPPs other than Dai-ichi, as mentioned, it's hardly believable, given the fact that Xe would have been detected much earlier than any later possible emission outside the primary containment buildings by the huge number of radiation monitors inside the primary containment buildings themselves. Please consider that it takes a lot of

time to pollutants to eventually leak outside a primary containment which has not been substantially damaged; this implies that there is in no way a possibility that such an emission would have not been measured, even if split among more than one single unit. If on the contrary your statement is related only to Dai-ichi, than we believe that such an extra-release is simply too big, as said at the beginning of this point; it seems a better explanation considering the possibility of a problem with the models used in your calculations or with the data you used. About your procedure concerning the validation of the robustness of your method, we understand that a variation of the parameters may indicate a variation of only +/-20% from the derived 16.7 EBq. But, in order to appreciate the actual accuracy of the results as a function of the variation of the parameters, it would be useful to estimate and report the value of one standard deviation of the nominal results.

2. *Burn Up*. We agree on the fact that the value we suggested for the fuel in the cores of units 1 to 3 would have not affected at all the Xe inventory, but only the Cs one. It is not clear to us why you mentioned a change in the U inventory in the cores as well (it should be kept at 69 t for unit 1 and 94 t for units 2 and 3), which is the actual responsible of the (small) change in the Xe inventory you calculated.

3. *Early Xe-133 emissions - 1*. If media didn't give the possibility to correct the misinterpretation occurred about releases within the earthquake moments and the tsunami arrival, we suggest you to include in the final version of the paper a strong statement to clarify this point. Analyzing also your reply to Mr. Horst-Michael Prasser, we think that some other points are to be touched as well. You clearly state there that the time resolution of the emissions is 3 hours; we understand that 3 hours is also the time resolution used for the calculations: in this case there might be some systematic error propagation as far as time advances too much, just because of the time resolution. You correctly say that the time resolution of most CTBT Xe-133 measurements is 12 h; however, even overlaying many measurements together, these data are time-integrated ones and this means a

loss of accuracy at lower, f.i. 3 h, time resolutions. Overlaying in itself may also be affected by large errors if you consider the many uncertainties in the meteorological data which disturb the transport calculations. The fact that your algorithm gives a value of 8 TBq/3h at March 11, 03 UTC seems an indication that errors at later phases may propagate at the initial times. We think it is important to stress your statement that early emissions are to be considered zero even if the model predicts different values, to better explain to the reader both the effect of the uncertainties on the model and the right key to be used in reading and interpreting the results.

4. *Early Xe-133 emissions - 2*. We suggest you to reconsider the statement that "after the loss of the AC power with the tsunami, there was no cooling to unit 1 core at all". Besides the fact that unit 1 has been cooled for at least 6 hours after the tsunami through the manual actuation of its own emergency cooling systems like the ICs, this point seems of secondary importance: this at least could just provide evidence of the presence of Xe in the reactor vessel and – thinking at the automatic safe depressurization of the latter – also in the primary containment vessel. But it should be recognized that the integrity of the primary containment has been guaranteed for a longer period, the first releases of Xe being due to the venting operation, so no release of Xe outside the primary containment can be assumed even accepting the lack of core cooling after the tsunami.

5. *Model uncertainties*. You state that to reproduce correctly the measurements of Xe-133 at the North American west coast your model requires even higher and earlier Xe-133 emissions: in our opinion this is a further indication that the higher emissions are mostly required by the model, rather than proven by it.

6. *Releases from the IC*. The statement that the operation of the IC 3A valve "could have released a considerable amount of noble gases, and even minor leaks of the containment would have released more and probably earlier" is questionable.

The valve operation (see Fig. 1) simply starts the cooling of the core by IC A. In case of problems on the piping – as mentioned, immediately revealed by the radiation monitors into the reactor building -, valve IC 1A, 2A or 4A may be operated to isolate the damaged line and the alternative line B used instead. Both ICs may be operated in parallel.

7. *Emission of Cs from unit 4 SFP - 1*. We didn't only put forth the argument that the elements in the Unit 4 SFP appear intact to infer that no big damages occurred there, but also advanced that radioactivity measurements made on several occasions indicate that no major release of radioactivity has occurred. We include here some data as an example:

SFP Unit 1	**Half life (y)**	**11th Feb. 2011 (Bq/cm^3)**	**22th June 2011 (Bq/cm^3)**
Cs134	≈ 2	Under detection limit	12000
Cs137	≈ 30	0.078	14000
I131	≈ 8	Under detection limit	68
SFP Unit 3		**8th May 2011 (Bq/cm^3)**	**7th July 2011 (Bq/cm^3)**
Cs134	≈ 2	140000	94000
Cs137	≈ 30	150000	110000
I131	≈ 8	11000	Under detection limit
SFP Unit 4		**4th Mar. 2011 (Bq/cm^3)**	**8th May 2011 (Bq/cm^3)**
Cs134	≈ 2	Under detection limit	56
Cs137	≈ 30	0.13	67
I131	≈ 8	Under detection limit	16

These data are not compatible with any assumption of a significant release of Cs from SFP of unit 4. It could help considering that the inventory of Cs in a single fuel pin burnt at 40000 MWd/kg is of the order of 10^{13} Bq. The Cs concentrations

measured in the water of the SFP of unit 4 seem on the contrary compatible with the hypothesis of a drag of a small amount of radioactive pollutants by means of the hydrogen transported from unit 3 to unit 4, as proposed in the report of the Japanese Government.

8. *Emission of Cs from unit 4 SFP - 2*. We looked carefully at the very interesting presentation by Christoph Müller you indicated us. Essentially Christoph Müller advances an hypothesis to justify the possibility of hydrogen production from the SFP of Unit 4; this is based on two major but absolutely necessary assumptions: a) a large accidental outflow of water (the best candidate phenomenon suggested being syphoning) that must have helped evaporation to uncover completely and quickly the fuel assemblies and, b) some phenomena like those verified in the s.c. CORA-33 experiments. The CORA-31 and CORA-33 experiments1 showed the possibility of cladding failure (at about 1200 - 1400 ℃) without extensive visible damage in case of severe vapor starvation, a condition that in a SFP may occur only in case of a large accidental outflow of water with very rapid drainage; in this case the damage mechanism of the fuel pin is as slow as the heating rate due to the exothermal oxidation reactions. In our opinion, this cannot have occurred at Fukushima:

   - In the case of an hydrogen explosion after the drainage of the SFP, damages to the fuel Assemblies (FAs) should be visible;
   - In the case of drainage of the SFP before the hydrogen explosion (which means, assuming polluted hydrogen coming from the RB of unit 3), two further scenarios should be considered:
     - If the drainage is slow (mainly due to evaporation), the damaging of the FAs which justifies the release of Fission Products should be massive, because of the steam oxidation of the cladding, and localized in the head of the FA, the part depicted in the videos by TEPCO;

– If on the contrary the drainage is fast (so, assuming the intervention of further mechanisms, such as syphoning), therefore causing the steam starvation condition, the release of Fission Products with some cladding failure at around 1200 °C(as reported in the CORA experiments) would have required the reaching of high temperatures in a matter of hours: simple thermodynamic calculations, under the hypothesis of quasi-adiabatic heating (air convection - water cooling being absent by hypothesis - is a negligible heat removal mechanism in these conditions; radiation plays some role, but only above about 800 - 900 °C) of the fuel elements due to their own decay heat, show that the time needed to reach about 1200 °Cis of several hours (with respect to some 1 h required in the CORA experiments, considering the much lower linear power rating of SF). According to this, the following consequences should be also expected:
  * the fast and violent oxidation in atmospheric air (absent in CORA experiments, performed in Argon environment), resulting in a severe and visible damage of the FAs2;
  * the later water spraying (as was done since March 19th), with the elements at temperatures at or above 1200 °C, would have resulted in incredible thermal stress on the structures, again with visible deformations.

  It could be also useful to notice that, once the FAs have reached the temperature of 1200 °C, it's a matter of few hours to reach – in an empty pool – also 1800 °C, the melting temperature of Zircaloy.

We would like to add here a further comment regarding the possibility of emissions before the venting operations executed on units 1 and 3, that your results indicate as probable. As it can be read on the official report from the Japanese Government, every operation of venting has been tempted several times in the hours preceding the

moment assumed as official actuation of the venting. All the preliminary tentative operations ended without complete success, that is: the valves were successfully opened, but they remained in this position for a limited time span because of the sudden lack of pressure from the gas bottles or electricity from the battery packs, both necessary to actuate the electro-pneumatic valves installed on the venting lines. During these short periods of actuation, a sort of "breathing" from the primary containment vessel to the reactor building has been possible, with corresponding releases of radioactivity.

Concluding these comments, in our opinion, simulations should be recognized reliable as far as they can reproduce experiments or facts: only from the comparison with reality the effectiveness of the models used in the simulations can be evaluated and assessed.

We appreciate the fact that you "can't and don't want to claim that your results prove releases exceeding the FD-NPP unit 1-3 inventories"; anyhow we think it's too strong to speak about the results of simulations as "evidences" regarding further emissions from the Fukushima site or from other NPPs in Japan.

What we suggest is therefore some caution in certain part of your work, and in particular regarding the implications of results which are unavoidably affected by uncertainties, in order to discourage as much as possible further indiscriminate and uncritical dissemination of information decontextualized from your work.

Finally, we would like to apologize for the length and maybe excessive detail of our answer, but the matter was very interesting, so it was our desire to be as clear as possible.

[1] S. Hagen *et al*. BWR slow heat up test CORA-31: test results. Report KfK 5383, 1994.
S. Hagen *et al*. Dry core BWR test CORA-33: test results. Report KfK 5261, 1994.

[2] M. Steinbrück. Prototypical experiments relating to air oxidation of Zircaloy-4 at high temperatures. *J. Nucl. Mat.* **392**(2009):531–544.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 28319, 2011.

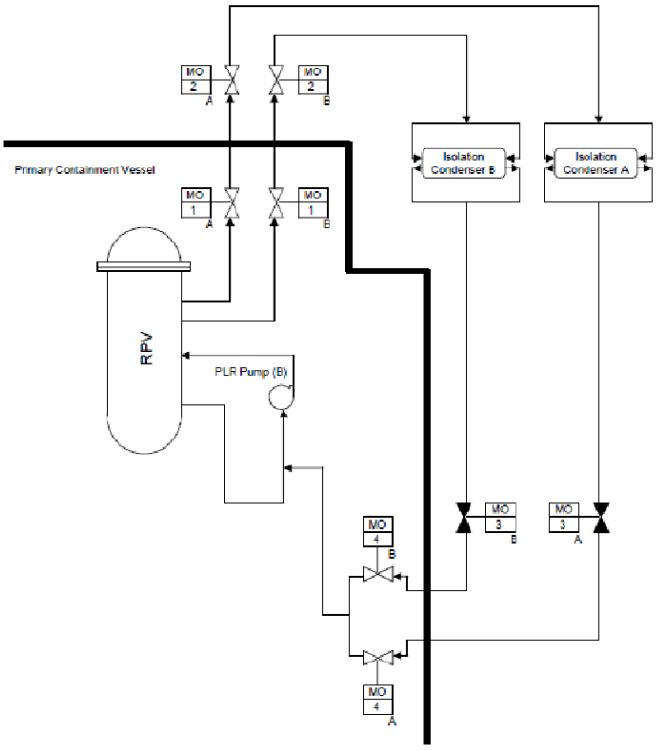

Fig. 1.