On cosmic rays, cloud condensation nuclei and climate

Rao[1], in his study on the role of galactic cosmic ray flux changes on climate, overlooked some of the important assumptions and conditions of validity of some of the reports he uses and, some recent experiments based on detailed physics show that the maximum radiative forcing caused by cosmic ray flux changes is very low (0.005 W/m^2). Also, some researchers Rao cites in his study rescinded their results and opinions in recent communications. The keystone of this study is the mechanistic relationship between GCR flux and cloud condensation nuclei (CCN) concentration through aerosol particle formation. Here, I examine the role and the robustness of this relationship.

The discussion in the study can be broadly broken down into the following questions for the sake of ease and clarity.

(1) How sensitive is aerosol formation to cosmic ray fluxes? The physics of ionization of the earth's atmosphere by GCR particles is well established. The presence of ions on chemical species in the atmosphere facilitates formation of molecular clusters and increases the probability of coagulation of these molecular clusters to form larger conglomerates, which further attain a critical size to continue to exist as aerosol particles[2,3]. These are ultrafine particles that have diameters less than 9 nm (ref. 4). This work by Lee *et al.*[4] is one of the studies that Rao's[1] argument depends on to explain the relation between GCR fluxes and CCN formation. A few key aspects of the study by Lee *et al.*[4] need to be kept in perspective while using their results. They report: (i) the formation of high concentrations of ultrafine particles; (ii) that the concentrations of these newly formed particles via GCR ionization are sensitive to the scavenging by pre-existing aerosol particles; (iii) particle formation in the upper troposphere and the lower stratosphere; (iv) that particles formed at temperatures higher than 270 K.

In the present context, these assumptions and conditions of validity of their results have important implications, as discussed below, for understanding the formation of particles.

(a) The aerosol particles that are formed in the upper troposphere are ultrafine and have very low terminal velocities. To act as CCN, they must be transported to the boundary layer which necessitates large terminal velocities. However, as their terminal velocities are small, their Lagrangian time to reach the boundary layer is very large. While the presence of ions on the molecular clusters enhances aerosol formation, a similar presence of ions on these newly formed aerosol particles increases their chance of scavenging by the larger aerosol particles. So, their probability of reaching the boundary layer to act as CCN is rendered very small.

(b) The observation that the concentrations of these newly formed particles is sensitive to the concentrations of pre-existing particles is a consequence of the fact that the pre-existing particles have sizes larger than these particles and these newly formed particles are scavenged through coagulation. The presence of ionic charge on these newly formed particles increases the rate of scavenging.

(c) Their simulations showed that the particles formed when the temperature was less than 270 K. It is interesting that at 270 K and 60% relative humidity, they found no significant nucleation. This is an important implication that new particles can only form in the middle and upper troposphere, and their formation in the lower troposphere is unlikely because the large concentrations of pre-existing aerosols remove the ultrafine, newly formed particles and also offer greater surface area for gaseous species to condense preferentially.

These inferences also assume importance in the context of correlation between GCR flux variations and 'low cloud cover', because low clouds form due to boundary layer eddies and are seeded in the boundary layer.

(2) How sensitive are CCN concentrations to aerosol concentrations? This is the weakest link in establishing the role of aerosol–cloud interactions in changing the climate. The uncertainty in our knowledge of aerosol–CCN relationship arises from two questions: (i) If there are *n* aerosols, how many CCN will result (aerosol–CCN closure)? (ii) If there are *n* CCN, how many cloud droplets will result (CCN–cloud droplet concentration closure)? These two questions have been defying theoretical understanding for almost two decades. The representation of these two processes in models is undoubtedly the greatest challenge for unravelling these relationships.

CCN is the subset of the population of aerosol particles at a point in the atmosphere that can seed clouds. The ability of an aerosol particle to act as a CCN depends strongly on the thermodynamic state of the atmosphere and the chemical composition and size of the particles[5]. Considering the diverse population of the atmospheres in which clouds evolve, it is generally agreed that the accumulation-mode aerosol particles can be roughly considered as CCN. They have diameters in the range 0.1–1.0 μm. Thus the ultrafine mode particles (5–40 nm) that are generated by the cosmic rays cannot act as CCN unless they grow by diffusing water to larger sizes. However, since the timescale of such growth is longer than that of aerosol-depleting processes like coagulation and scavenging, the contribution of these ultrafine particles to CCN is small.

Much of the aerosol–cloud interaction research using models and observations for the past two decades has been to understand this relationship. Most of the models used Twomey's parameterization of cloud droplet concentration.

As the knowledge of aerosol physical and chemical processes advanced, this representation resulted in inaccurate estimations of cloud droplet concentrations[5]. Sophisticated models to represent the diffusion growth of an aerosol to a cloud droplet are the latest advancement[6,7].

It is to be noted that the study by Yu[8] cited by Ramanathan[9] assumes that aerosol particles with a dry diameter of 30–50 nm can be activated as CCN based on reports of activation of similar-sized particles in deep convection where the supersaturations are very large[10] compared to the low level clouds being considered in Rao's[1] study. With this assumption, Yu[8] assumes that ultrafine particles are activated, which is not true in stratocumulus and small cumulus due to the low supersaturations in these clouds compared to the deep convective clouds.

(3) How well do the GCR flux and low cloud cover correlate? The motivation

for the study by Rao[1] is the correlation reported by Svensmark and Friis-Christensen[11]. It is important to note that Svensmark and Friis-Christensen[11] do not report the correlation between GCR flux and low cloud cover, but report that between GCR flux and total cloud cover, and this discrepancy was later corrected in Marsh and Svensmark[12].

An important remark by Laut[13] was that the correlation was valid only for 1984–1994, and that there is no agreement afterwards. studies showed that these correlations are not as clear as evinced in Svensmark and Friis-Christensen[11]. Laut[13] pointed out many inconsistencies in establishing the correlation in Svensmark and Friis-Christensen[11] and suggested that any conclusions drawn based on the results in this study need to be buttressed by further investigations.

It is known that solar variability affects GCR flux and solar irradiance is the dominant forcing on the earth's atmosphere. So, it is possible that both GCR flux and some geophysical fields on earth which have very high sensitivity to solar irradiance show the same variation. While they may seem correlated because they are forced by the same field (solar irradiance), they may not be mechanistically correlated at all. Thus, any observed correlation is random.

In a latest set of experiments, Kristjánsson *et al.*[14] used satellite data (MODIS) to look for any possible correlation between GCR flux changes and the four most important cloud variables that play a key role in cloud radiative forcing – cloud droplet size, cloud water content, cloud cover and cloud optical depth. For greater robustness of any possible relationship, they chose the regions where such a relationship may have the greatest impact – marine atmosphere in the southern hemisphere and clouds that are most susceptible – stratocumulus. They reported that there is no correlation.

It is interesting to note that Veizer[15], whose report of correlation between cosmic ray flux and low level cloud intensity has been used in Rao[1] (figure 2), rescinded his hypothesis in Came *et al.*[16] and infered that rising CO_2 is largely responsible for climate change.

As Ramanathan[9] has pointed out, the magnitude and periods of correlation among these studies of correlation between GCR flux and low cloud cover vary considerably.

(4) How robust is the relationship between GCR flux, CCN and global climate as speculated by Carslaw *et al.*[17]? Carslaw *et al.*[17] speculated the relationship between GCR flux and CCN concentrations and the linkage to global radiative balance.

Pierce and Adams[18] used a general circulation model with an aerosol microphysics module to study sensitivity of the formation of aerosols due to changes in GCR flux. To address the specific issue of the relationship between cosmic rays and CCN, they computed the number concentration and composition of aerosols as a function of the particle size and time in the whole troposphere and the lower stratosphere for the years with high and low GCR fluxes, approximately corresponding to 1986 and 1990. Between these years, the GCR fluxes changed between 15 and 30% in the free troposphere and by about 10% in the lower troposphere. They assumed that every ion produced resulted in the formation of a particle if there is even a small amount of sulphuric acid in the vicinity. Thus their findings set the upper bounds for particle and CCN concentrations.

Their result that the CCN concentrations changed by just 0.04% between solar minimum and solar maximum years is a remarkable one. Models are imperfect and have uncertainties. But the fact that their results put an upper bound on CCN formation leads to the important conclusion that CCN concentrations are insensitive to GCR flux variations and is a landmark inference.

Pierce and Adams[18] used the computed CCN sensitivity of 0.04% to infer that the change in solar radiation reflected to space would be 0.005 W/m^2. This is insignificant compared to the value of 1.1 Wm^2 computed in the study by Rao[1]. Investigations of aerosol nucleation by GCR flux variations by Pierce and Adams[19], and Spracklen *et al.*[20] lend further credence to the results of Pierce and Adams[18].

Further, Carslaw[17], on whose report the hypothesis that GCR flux variations impact global climate rests in Rao's[1] study, rescinds his speculation in a recent letter[21]. He reasoned that based on the study by Pierce and Adams[19] and other similar reports, it is clear that global climate is insensitive to GCR flux variations.

Contrary to Ramanathan's[9] inference that one cannot conclude about the existence of a relationship as that suggested by Rao[1], Pierce and Adams[18] and Carslaw[21] are confident that the said connection cannot be found. Pierce and Adams used a sophisticated representation of global lifecycle of aerosols starting from their formation to growth over days to weeks to the sizes of CCN. The confidence of Pierce and Adams[18], and Carslaw[21] is borne of two facts that the used representation of aerosols was detailed and further, Pierce and Adams, instead of modelling the detailed ion–aerosol processes, made an assumption that all nucleation is due to ions, which sets the upper bound on the CCN sensitivity to GCR flux variations. So, any detailed treatment of aerosol physics and chemistry would yield CCN sensitivity less than 0.04%.

This also provides credence to the observation by Ramanathan[9] that the GCR flux changes cannot account for the global warming trends.

Recent advances in the understanding of the physical and chemical mechanisms of formation and growth of aerosol particles in the earth's atmosphere have been helpful in understanding the GCR–CCN link and in keeping it in the right perspective. Some of the recent reports to estimate the climate sensitivity to GCR flux variations have been categorical that the contribution of the GCR–CCN link to the radiative forcing is only 0.005 W/m^2, which is small compared to 1.1 W/m^2 reported by Rao[1]. Quantification of radiative forcing by all known or hypothesized forcings on climate is important as our understanding of the evolution of earth's climate has a bearing on this number. The best course would be to examine the mechanistic relationships in this chain of processes to evaluate the validity and limitations of the assumptions and incorporate the ever-advancing theoretical understanding of the physical processes. Since activation of aerosols is the key process in this linkage, field experiments like the Cloud–Aerosol Interaction and Precipitation Enhancement Experiment, and development and use of models that represent detailed aerosol activation physics like those by Donnan[6] and Bhaskar[7] are helpful in understanding this relationship.

1. Rao, U. R., *Curr. Sci.*, 2011, **100**, 223–225.
2. Arnold, F., *Space Sci. Rev.*, 2006, **125**(1), 169–186.

3. Yu, F. and Turco, R. P., *Geophys. Res. Lett.*, 2000, **27**(6), 883–886.
4. Lee, S. H. *et al.*, *Science*, 2003, **301**(5641), 1886.
5. Houghton, J. T. *et al.*, *Climate Change 2001: The Scientific Basis*, Cambridge University Press, Cambridge, UK, 2001, vol. 881.
6. Donnan, S. H., Ph D thesis, Massachusetts Institute of Technology, USA, 2004.
7. Bhaskar, G. U., Ph D thesis, Massachusetts Institute of Technology, USA, 2010.
8. Yu, F., *J. Geophys. Res. A*, 2002, **107**(7), 1118.
9. Ramanathan, V., May 2011; http://moef.nic.in/downloads/public-information/Discussion-paper-INCCA-1-2.pdf
10. Shaw, G. E., Benner, R. L., Cantrell, W. and Clarke, A. D., *Climatic Change*, 1998, **39**(1), 23–33.
11. Svensmark, H. and Friis-Christensen, E., *J. Atmos. Sol-Terr. Phys.*, 1997, **59**(11), 1225–1232.
12. Marsh, N. D. and Svensmark, H., *Phys. Rev. Lett.*, 2000, **85**(23), 5004–5007.
13. Laut, P., *J. Atmos. Sol-Terr. Phys.*, 2003, **65**(7), 801–812.
14. Kristjánsson, J. E., Stjern, C. W., Stordal, F., Fjæraa A. M., Myhre, G. and Jónasson, K., *Atmos. Chem. Phys.*, 2008, **8**(24), 7373–7387.
15. Veizer, J., *Geosci. Canada*, 2005, **32**(1).
16. Came, R. E., Eiler, J. M., Veizer, J., Azmy, K., Brand, U. and Weidman, C. R., *Nature*, 2007, **449**(7159), 198–201.
17. Carslaw, K. S., Harrison, R. G. and Kirkby, J., *Science*, 2002, **298**(5599), 1732.
18. Pierce, J. R. and Adams, P. J., *Geophys. Res. Lett.*, 2009, **36**(9), L09820.
19. Pierce, J. R. and Adams, P. J., *Atmos. Chem. Phys. Discuss.*, 2006, **6**(6), 10991–11023.
20. Spracklen, D. V. *et al.*, *Geophys. Res. Lett.*, 2008, **35**(6).
21. Carslaw, K. S., *Nature*, 2009, **460**(7253), 332–333.

ACKNOWLEDGEMENTS. I thank all the scientists in the MIT Joint Program for their valuable comments and for validating some of the arguments made in this discussion.

Received 7 May 2011; accepted 23 May 2011

GUNTURU UDAYA BHASKAR

The MIT Joint Program on the Science and Policy of Global Change,
Department of Earth, Atmospheric and Planetary Sciences,
Massachusetts Institute of Technology, Cambridge, MA 02139, USA
e-mail:bhaskar@mit.edu