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Report on the PhD thesis of Arianna Varrani, titled "*Onset of motion of compact-shaped microplastics in open-channel flows*".

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The result of fragmentation, weathering, photooxidation or biodegradation of synthetic plastics over several decades is the ubiquitous presence of microplastics in the environment. They are present in rivers, mixed with natural sediment, and are known to enter the food chain. The many awareness campaigns have contributed to limit the amount of plastics that end up in water courses, at least in Europe. However, the fate of the small plastic particles is still a great concern to human communities.

To assess the fate of microplastics, one of the aspects to tackle, as important as the entry points in the food chain and degradation, is the mechanics of its transport in rivers. Arianna Varrani chose, as her PhD thesis topic, a subset of these processes, namely that involving near-bed processes (entrainment and transport as bedload) of particles whose shape is similar to that of natural river sediment of the same size. The methodology is experimental. Arianna builds her arguments based on results obtained by operating a hydraulics flume with research-level velocimetry instrumentation.

I have no doubts that Arianna Varrani achieved complete mastery of laboratory techniques in hydraulics and mechanics of sediment transport and has attained an exceptional level of understanding of data processing strategies. Ariana also shows a firm grasp on the main concepts that emerged over the past century on mechanics of sediment transport and (incompressible) fluid mechanics. She cites most relevant papers I can recall (and some I'm not familiar with) about sediment transport. Ariana's reading of these works does not always coincide with mine but that is not a disadvantage. The state of the art on plastic transport is much less explored, I'll come back to this issue.

Based on her understanding of the past research on sediment mechanics and her awareness of the challenges of microplastic transport, Arianna poses what I believe to be a fundamental research question, at a time when the answer is much needed. The way this question is posed at the conclusion, "Are compact microplastics talking sedimentary processes' language?" is a very good and creative way to put it, one that I fully endorse[1].

So, in Arianna's view, can we deal with microplastic transport with the tools developed over the past 100+ years of sediment research? To answer this, Arianna is very careful to single out as object of study only plastic grains that are convex solids, such as sand grains or water-wore gravel particles (the "compact" microplastics). Arianna does not forget that previous research has used plastic as sediment to test the validity of general formulas based on parameters that incorporate the density of the sediment, such as the Shileds parameter $\theta = \frac{\tau_b}{\rho(\Delta - 1)gd}$, where $\tau_b$ is the bottom boundary shear stress, $\rho$ is the water density, $g$ is the acceleration of gravity, $d$ is a representative grain diameter and $\Delta = \rho_s / \rho$ is the specific gravity of the sediment grains, where $\rho_s$ is the density of the grains. This is classical avenue that is revisited in the thesis but not the main objective. The main objective is, if I can summarize it as so, to understand if the description of microplastic transport at low rates (including the very subjective rates that we may describe as "incipient motion") can be formulated with the same tools of classic research of sediment mechanics.

Considering the type of microplastics (that excludes filaments) and type of natural sediment, my reply to this question, and the more vivid question are "microplastics talking sedimentary processes' language?" would be a resounding yes – it is my working hypothesis, one that I would like to see validated in Arianna's thesis. And that does not mean that the problem is or would be immediately solved. We may have the language and plastics may talk it, but extracting the full answers demands patience and skill, and this is why I found Arianna's approach the right one at the right time.

The main issue, that Arianne is fully aware, is that we are dealing with a complex classification problem, that does not only involve sizes but also densities[2]. The same density class will have different size classes, and the same size fraction of particles may be composed of different types of plastic, with different densities. Arianna's approach here is classical and conservative. And fully justifiable – Arianna picks two types of plastic, with similar shapes, different densities and different grain-size distributions (one is characterized by a single diameter, the other is a distribution) but essentially the same median diameter. She then observes the weak bedload transport of the plastic classes and formulates her arguments about the condition of possibility of casting these transport conditions in the language of sediment mechanics.

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[1] I like less the way the research questions are laid out in the Introduction. Please see below the transcription of my comments in the manuscript.

[2] I believe Arianna cites a colleague, Enrica Viparelli, I cite from memory, as I did not find again the citation in the text (not having a pdf version complicates searching), that showed that most work on sediment mixtures was done on size classes of natural sediment and very few on density classes.

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Arianna's answer to her research questions is somewhat restrained. There is a comment on observation that the amount of plastic particles in the bed surface does not influence the threshold into conditions we might call generalized transport. But since the availably of plastic grains in the surface does influence any quantitative attempt to determine incipient motion conditions. This is rather relevant and quite understated in the text. It shows that our natural predisposition, following the early works by Kramer, to see incipient motion as the flow conditions for which only a subjectively low number of particles is moving presupposes that we are observing bed with grains with the same density. For the same incipient conditions, the availability of the only density class determines the number of particles that are moving – counting particles is not enough, rate of bedload-based estimates is not enough too. The criterion to decide incipient motion conditions must be made relative to the class of sediment and its availability.

This is, I believe, the content of another understated answer, about the uniqueness of the threshold and a range of uncertainty (section 6.3) on identifying threshold conditions. Reading Arianna's thesis, I do not think there are fundamental impediments to address microplastic transport with the tools of sediment mechanics, so I'm surprised that her answers are not more assertive. There are challenges about the classification of the sediment and how to formulate size and density selective formulas, but the language is there.

Some of the methodological options by Arianna may have conditioned the range of the replies. And perhaps lack of time resulted in a sub-exploitation of the database or hindered the production of complementary databases. Results on actual bedload rates, and an attempt at parameterizing such rates to grain, sediment mixtures, flow and fluid quantities may have provided substance for more assertive conclusions.

This does not take merit out of Arianna's thesis. Her PhD work and published material places Arianna as a world reference in plastic transport themes, her methods will be taken as a reference point for future studies and her results will be cited. So, formally, to comply with my contractual obligations as reviewer of Arianna's thesis, I state that

- I have the highest regard for Arianna's knowledge on sediment transport issues and I sure that her skills and knowledge guarantee that she is able to conduct independently scientific research and guide future students on these matters;
- The problem Arianna addressed (microplastic transport in rivers) is relevant and required scientific investigation with the methods Arianna proposed (drawn from experimental sediment and fluid mechanics); the solutions she found are valid and original and a significant contribution to the topic;
- I have a very positive opinion regarding the candidate's admission to the public defence.

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At this point, it is inevitable that I now write about the aspects I liked less in Ariannas's thesis.

1. A first issue is the lack of recent references about studies dealing with plastic transport with sediment in rivers. A quick search the Scopus database reveals hundreds of entries in the past 3 years of papers whose objectives are relevant to this thesis. I will not point out any particular study, but I wat to share the finding that there is information about how microplastics mix with sediment. Less on transport processes, which justifies this thesis, but some using concepts of sediment mechanics. Works that characterize the way microplastics and sediment are mixed in actual riverbeds would be useful to determine the way they could be mixed for the lab experiments.

2. I'm not convinced about the introduction of "incipient motion" and "remobilization" as mutually exclusive concepts (I read dichotomous somewhere in the text). In current language, plastic particles can be remobilized at incipient motion conditions or not so incipient. I can understand that the candidate does not want to use "incipient motion" for beds composed granular mixtures. But the arguments are not convincing. The assertive tone of section 1.3 (almost authoritarian) is not backed by evidence, it is merely a rhetorical movement that is not even scrupulously followed in the text.

In my view "incipient motion" means the state of a granular bed for which a subjectively small number of grains is in motion. We can quantify this, for instance defining a quantitative upper limit to bedload flux or grain flux, but, at this stage of knowledge, the value of such limit is still subjective – there is no necessary value stemming from some transport quantifier. Can a bed made of different size and density classes be in incipient motion? The largest and denser particles will not be mobile but, yes, the bed will be experiencing incipient motion conditions because some of its lighter/smaller fractions are experiencing limited movement.

A different concept is "critical" flow, which I personally find somewhat misleading. It may mean a state of a flow over granular bed, characterized by specific values of flow variables, for which that granular bed is at the threshold of changing from week values of sediment transport to generalised sediment transport. I believe that the concept is only useful in the context of defining beload formulas based on the Shields parameter. The "critical" value of the Shields parameter is the critical value of the bed shear stress normalised by fluid and sediment properties and representative sediment diameter. It allows a good fitting of simple power laws valid for generalised transport and ignores the week transport that may be expressed by more complex formulas[3] but in practice too small to consider. And then, as Arianna shows in this thesis, a numeric threshold cannot be universal for a given size and density class, as it depends on its availability on the bed surface. But that does not mean a hiding/exposure function cannot be devised

[3] Like the 16th power polynomial advocated by Paintal in 1970 or 1971 (I cite from memory).

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for the purpose of constructing a formula for partial transport of different size and density classes.

As for remobilization, it has no tradition in sediment mechanics, at least not in this context of incipient motion (and to my knowledge). In natural language is a noun used for the act of getting something ready to be mobilized again. In what sense plastics move again? I see no need for this term. I would much prefer that Arianna would explore the and make precise the terms already in use.

3. I liked a lot the question “are microplastics talking sedimentary processes' language?”. The questions in section 1.1 seem a bit convoluted. Q1.1 and Q1.2 are confusing (is “critical threshold” a redundancy?) and bit overlapping. How do you cast critical conditions of not based on hydraulic variables? Question Q2.2 seems written after the thesis was completed and there was apparently a Q3 (see section 1.2) that may have been forgotten. I would prefer that the fundamental research question so elegantly formulated in the Conclusion would be in the Introduction.

4. Chapter 2 voices, in my opinion, misconceptions about thresholds of motion in granular beds subjected to free-surface flows and about the significance and scope of the Shields diagram.

Section 2.2.2.2 is about “particle-oriented descriptors” of (2.2.2) “Incipient motion of sediments ...”. This could not be more misleading. The force or torque limit static equilibrium conditions have no necessary connection to incipient motion. Static equilibrium conditions pertain single particles, incipient motion pertains the granular bed, best described with statistics. Every grain has its limit equilibrium conditions, depending on its exposure, geometrical properties and density. And that is true if the bed is under incipient motion conditions or under generalised bedload. I believe that the work of Martina Cechetto and my work with my PhD student Federica Antico showed this.

The link between critical flow and the limit equilibrium conditions for specific grains is statistical, in the wake of the works by Paintal or Grass. My work with Marwan Hassan and Carles Ferrer-Boix is an example of this. Having a statistical distribution of turbulent velocity fluctuations, having statistical distributions of exposure and shape and density of particles, we can construct a predictor for critical flow and, indeed, a predictor for bedload fluxes. It involves statistics which means it needs a spatial domain containing enough particles, highlighting that incipient motion pertains to a completely different scale of analysis relatively to particle equilibrium conditions.

The jumps between 2.29 and 2.30 and 2.31 and 2.32 entail hypothesis that endorse my comments and some simplification that do not. In any case, I find it incorrect that particle equilibrium conditions are mixed with incipient motion conditions without a through explanation of the scale of the approaches and the condition of possibility to have a link between these two scales of analysis.

Section 2.2.2.3 does not clarify the issues above - the explanation based on force balance does not make sense at the scale of analysis inherent to Shields' diagram. And introduces the suspicion that Shields' diagram does apply to turbulent flows. This follows an earlier proposal, in p. 16, in the section about "turbulence-based descriptors". I my view, there have been confusing interpretations of classical works, perhaps induced by the methodology of White 1940. But in our age, I believe we should clarify these matters and stop propagating misconceptions. So, Shields' diagram is cast in statistical terms – uses mean flow variables. That does not mean the effect of near-bed turbulence is not considered. The "critical" bed shear stress in Shields diagram is the bed shear stress – a mean flow quantity – for which a subjectively small number of grains of the granular bed is found moving. Why are they moving? Because the limit equilibrium conditions of those grains were attained and exceeded. Because these grains were too exposed to resist the "instantaneous"[4] hydrodynamic actions. In fact some grains are really (in absolute terms) very exposed and can be entrained by a mild hydrodynamic action (with an associated mild velocity in the vicinity) and some grains were less exposed but subjected to an exceptional hydrodynamic action (a large near-bed velocity, maybe within a large turbulent structure). The value of the Shields' threshold entails a specific preconception of turbulence, as generated in boundary layers over rough elements, and of the statistical distribution of grains in the granular bed. And, of course, Shields' diagram is very ill-equipped to deal with variations of density in the granular material – and again I justify my enthusiasm about this thesis. So, if the structure of turbulence is changed (y seepage for instance), if the distribution of roughness elements is detached from the distribution of mobile factions in the bed surface (as is the case of the seeded beds in this thesis) of course the Shields' thresholds will not be useful – its preconceptions are not met. But this does not mean that the approach is not valid – it means that the preconceptions must be eliminated to make more general.

In this respect, the sentences like the last sentence in p. 23 "They propose... nuanced concept ... overcome particle inertia" and the same sentence in p. 27 are damaging to future progresses and to this thesis. If the candidate can remove them, it would be very beneficial.

As a minor issue, I ask Ariann to check for errors or typos equations in 2.18 and 2.19 and define all variables. I would also find it beneficial that the Shields number and the roughness Reynolds are introduced in section 2.1.2

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[4] Not exactly instantaneous because it takes some time for the forces around the particle to build up, as shown by the works of P Diplas and co-workers.

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5. I have a major concern about some of the experimental tests. The flow is highly non-uniform, if the gradient of the flow depth is 3 mm over 1.5 m. The channel slope is of the order of 0.0001 or 0.00001 (it is not clear both numbers appear in different parts of the text). The gradient of the flow depth is 1 order of magnitude higher. It would be the dominant term if the friction slope would be computed by an integral energy balance.

I see that some form of Clauser's method was chosen to compute the friction velocity. But, in fact, I think the candidate should have tried and contrasted different methods. The adopted method is not well described – equation 3.2 is not what is plotted in Figure 4.2. In the equation, roughness scales are adopted, in the Figure the distance to the wall is normalized with the viscous scale typical of smooth boundary layers. The way to estimate the roughness length is not described in thesis.

But more importantly, in p. 55 there is a (essentially correct) mention to the limits of validity of the logarithmic fitting -between 30 and about 140 wall units. Bu in this case, the upper limit is not respected. In a uniform open-channel flow this would not be a major issue. Bu if, as it seems to be the case, the flow is gradually varied (spatially accelerating or decelerating) the log-law must be complemented with a wake function (Coles), with more fitting parameters. Or used strictly within the cited limits. What we see in Figure 4.2 is that the lines fit well the outer layer – were the log-law in not valid – and are not well-adjusted to the three first points, in the overlapping region. This may be a cause of serious errors in the estimation of the friction velocity.

Moreover, the option to collect data at only 5 locations above the bed is highly questionable. The structure of the boundary layer would always be an important factor in this type of studies. It is not possible to grasp the structure of the boundary layer with only 5 points in the wall-normal direction.

Still on the issue of the experimental tests, I commend Arianna's integrity by showing the less-than-ideal conditions of here flume in Figure 3.3. Laboratory channels are tricky facilities, it is sometimes frustrating to set-up the ideal conditions. The flow in Figure 3.3 seems skewed and that may have had impacts on the estimation of the friction velocity as it surely had an impact on the wall-normal profile of the longitudinal velocity. This is where other estimates of the friction velocity could be useful. I hope that the channel has become more manageable, and I state that I still find Arianna's database valid and useful.

6. Still on the issue of computing the friction velocity, I believe I read about attempts to determine the instantaneous friction velocity or, at least the friction velocity over a short time span (could Figure 5.3 be about such fluctuations of the friction velocity?).

The spatial average of the bulk force exerted by the flow on the granular bed surely fluctuates over time. It is important to know these fluctuations and there are methods to do it, for instance measuring forces on base plate that supports the bed. One of the

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methods that has no theoretical backing is using instantaneous (or short timescale-averaged) wall-normal profiles of the longitudinal velocity to estimate an instantaneous (or short timescale-averaged) friction velocity. The friction velocity that normalizes the ensemble-averaged (time-averaged and/or space-averaged) longitudinal velocity in the log-law

$$\frac{\langle u \rangle}{u_*} = \frac{1}{\kappa} \ln\left(\frac{z u_*}{\nu}\right) + B \qquad \text{(A)}$$

is a kinematic scale that is the expression of the wall shear stress $u_* = \sqrt{\frac{\tau_b}{\rho}}$. This is only true if the certain conditions apply, notably wall similarity, a statistical concept that must involve all scales of the turbulent flow. If wall-similarity holds, the bed shear stress provides the kinematic scale that makes the z-derivative of equation A above (the shear rate) independent of the Reynolds number, i.e. self-similar. Because we know that wall-similarity holds, than the $u_*$ in equation A provides a sound estimate of the bed shar stress.

But this does not mean that any successful logarithmic fit

$$\frac{u(t)}{u_s(t)} = a(t) \ln\left(\frac{z u_s(t)}{\nu}\right) + B(t) \qquad \text{(B)}$$

of instantaneous data provides sound estimates of the hydrodynamic force on the bed per unit area. There is no proof or empirical confirmation that the kinematic scale $u_s$ in equation B is an estimate of an "instantaneous" bed shear-stress. The statistical conditions that allow for this link between different and independent concepts (the kinematic scale the normalizes the velocity profile and the momentum sink at the bed) are not met. Maybe it is not a bad estimate, but there is no sound evidence, and that we should avoid using it without a lengthy explanation of its theoretical insufficiencies.

7. The usage of the older DOP UVP system should come with a lengthy explanation, too. As the candidate surely knows, a single UVP probe measures velocities along its beam. To obtain longitudinal velocities, one needs to assume that the velocities measured along the beam are projections of the true velocity in the longitudinal direction. For that purpose, the probe is deployed at an angle to the bed. But the problem is that the projection is not unique, the same projected velocity along the beam may come from infinite (well, limited by the physics of the flow) combinations of longitudinal and vertical velocity.

As a consequence, in a uniform open-channel flow, the time-averaged longitudinal velocity profile is highly believable, since the time-averaged vertical (or wall-normal) velocity is zero. But if the open-channel flow is not uniform or the velocities are instantaneous rather than time-averages, the profiles will contain errors. All this should

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be explained in the thesis and the impacts on the measurements should be explained and accounted for.

A more detailed explanation on the actual data sampling rate would be welcome. The sampling time is adequate (3 min) but I'm not sure I read correctly that the time series are resampled at 0.3 Hz? Is that correct? If so, why? Are there enough points for a convergence of even the first moments?

8. The last methodological issue concerns the choice for sprinkling the water-worked bed with plastic particles. I think this was a great idea, as it allowed for the discussion of the influence of the availability of this density class (I wrote it above). But the discussion does cover the implication of this method – the plastic particles are on average over-exposed. This has effects on the interpretation of incipient motion conditions. Together with imperfections in estimating the friction velocity, may lead to an imperfect view and discussion of the flow conditions under which this density class undergoes week transport.

I would say this database should be complemented with another in which the plastic material is mixed by volume with the natural sediment and then placed in the bed. There are a lot of difficulties in this method, related with segregation by buoyancy, grain skin friction, and shape that condition the way the bed is laid out. But is done adequately to match what was been observed in field studies (see my comments about the state-of-the-art) with would provide a view of fractional entraiment and week transport not affected by selective exposure.

9. Moving on to the results, I miss a formal dimensional analysis before the principal component analysis is attempted.

I do not agree on the classification of several variables and parameters in p. 76. For instance, $\rho$ and $\nu$ are not flow properties, they are fluid properties. Gravity is a context, in the fact the condition of possibility of open-channel flow, but it is not a flow variable. I do not know what are variables $U^*$ and $\tau^*$ but they are the friction velocity and a bed shear stress, they cannot be in the same list of dimensional parameters. Well, they can be but they should not, as that is a waste of time - they are the same. And, furthermore, the friction velocity is not a property of the bed sediments.

The outcome of the much-abbreviated dimensional analysis in p. 76 is surprising to me. How come the Shields parameter or another type of Froude number, with density effects, is absent from the list of parameters for mixed beds?

The list of parameters in the homogeneous bed does have Fr-type and Re-type parameters. But some are redundant, there is no need for a PCA analysis to verify this. For instance, it seems to me that

$$Fr = Fr_S^* \left( \frac{y}{d_{MP}} \right)^{-1/2} (\Delta + 1)^{-1/2} \, \mathrm{Re}_*^{-1} \, \mathrm{Re}$$

and

$$Fr_S^* = \theta^2$$

I believe this should be checked carefully for future publications.

10. I see the principal component analysis not as an end in itself. It allows the identification of orthogonal groups that are most important to explain the data. If conducted properly, it eliminates parameters we hadn't realised they were redundant and suggests the removal of parameters that may be independent but contribute little to explain the trends in the data.

After this step, it is customary to conduct a curve fitting analysis to find expressions between the relevant combination of non-dimensional parameters and the non-dimensional variable for which we wish to have a predictive model. In this case, the rate of bedload is not readily available but proxy $\chi_B$ has been computed. So why not attempt a multivariate analysis with between $\chi_B$ and the non-parameters found independent (or powers or logarithms of these parameters). I would say the extra effort to do that is small relatively to the huge effort to generate the databases.

The proxy $\chi_B$ does not tell us if the perturbation in the image is caused by the same particle as it entrains and deposits in the same field of view. Or if the particle just arrives to the field of view; or if it just leaves the field of view. It is a measure of bed activity but very conditioned by the size of the field of view relatively to the size of the jumps.

11. I understand the time is always short for a PhD thesis. But not to attempt estimating the bedload rates seems a waste of resources. There a freeware software packages available for computing week fluxes of granular material (Track-Track from Joris Heyman, the tool from Roger Noaks, I forget the name, among many others). Not all software solutions have to be developed in-house by the candidate.

I believe this has somewhat limited the range of discussion of the results. I would be very much available for future collaborations to further explore this database.

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